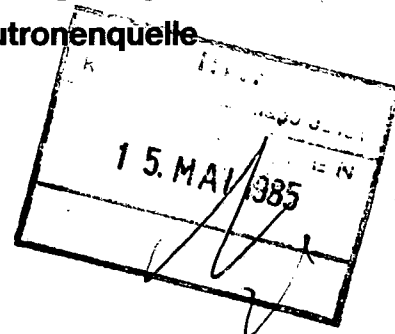




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**Spallations - Neutronenquelle**

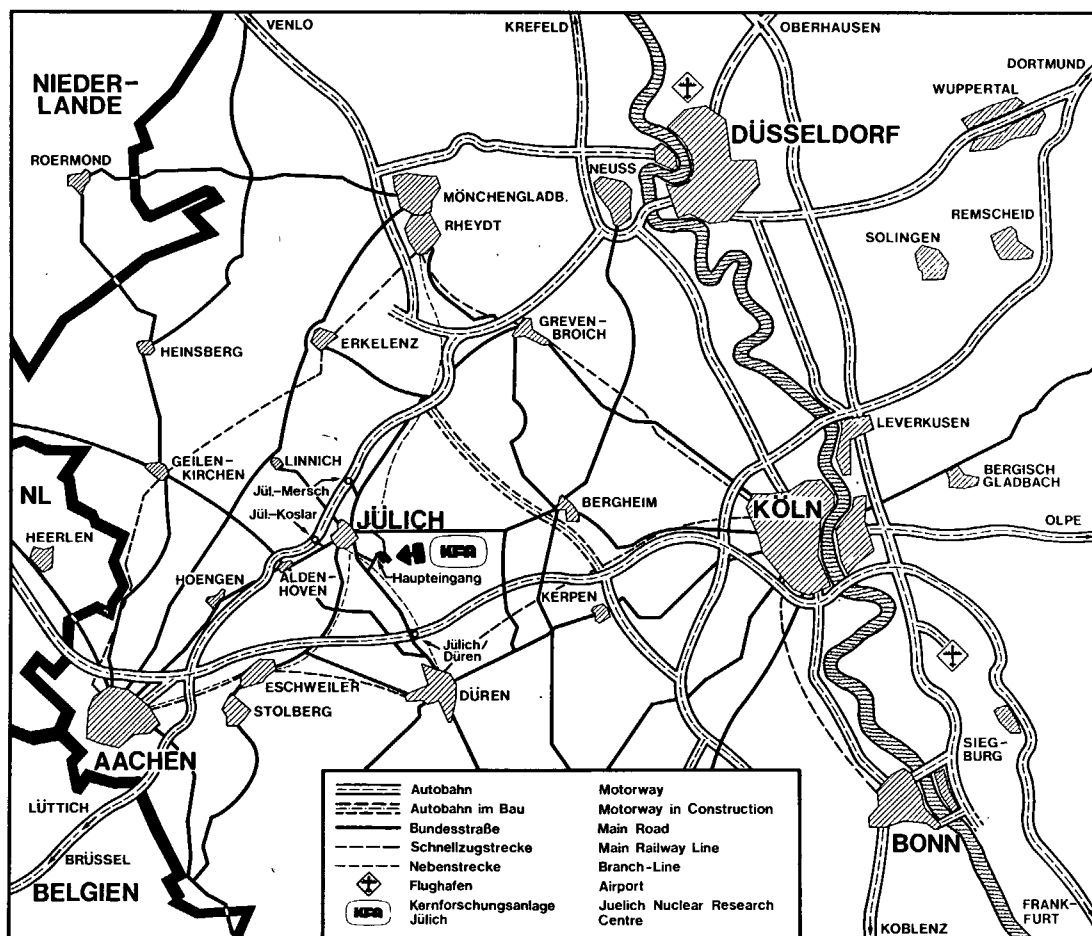


**Proceedings of the Workshop on  
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for SNQ,  
Maria Laach, 3 - 5 Sept. 1984**

**Editors : R. Scherm, H. Stiller**

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## SNQ - Instrumentation

### Preface, Introduction and Summary

R. Scherm

PTB, Braunschweig

Usually we think of big accelerators as high energy machines built to chase more exotic elementary particles. With the recent progress made in the acceleration of high beam currents, spallation neutron sources based on accelerators, generating megawatts of beam power, became a valuable competition to high flux research reactors for the production of neutrons. In contrast to high energy physics, which investigates the basic fundamental forces, condensed matter research deals with a wide variety of physical processes in many body systems. Here neutron scattering plays a dominant role, the neutron being an ideal probe for the investigation of microscopic atomic and magnetic properties of solids and fluids.

Generally the scattering of thermal neutrons by any sample can be described by the scattering function

$$S(\underline{Q}, \omega)$$

which, besides of trivial factors, describes the probability of a neutron being scattered with a transfer of momentum  $\hbar \underline{Q}$  and a transfer of energy  $\hbar \omega$  to the atomic lattice of the sample. Via a Fourier transform  $S(\underline{Q}, \omega)$  is related to the so called pair correlation function  $G(\underline{r}, t)$ , which describes the correlation in space  $\underline{r}$  and time  $t$  between two particles or magnetic electrons in a many body system. This information is the closest one can get to a "hologram movie" i.e. following the movement  $\underline{r}_i(t)$  of an individual particle, an information which would require to register the phases of scattered waves.

In practice we distinguish elastic and inelastic neutron scattering, depending whether or not the energy of neutrons is analysed after the scattering event. Elastic scattering measures  $S(\underline{Q})$  and gives information on the structure i.e. spatial arrangement of atoms or magnetic electrons. Depending on the scales to be investigated, we distinguish diffraction, where the atomic or magnetic



structure can be gained from the intensity of Bragg reflections. To resolve the structure of big molecules such as proteins, it is necessary to measure more than  $10^5$  reflections. Diffuse scattering observes the elastic intensity between Bragg reflections, such unveiling defects, impurities and distortions in imperfect lattices. Large scale objects as e.g. precipitates in alloys, the conformation of polymers or large biological molecules in solutions can be seen by Small Angle Neutron Scattering.

Inelastic Neutron Scattering necessitates to define the energy of neutrons both before and after the scattering event. Here the transfer of energy, typically of the order of 10 meV, combined with the creation or annihilation of an elementary excitation  $Q, \omega$  is observed. Examples are molecular vibrations, molecular rotations, phonons (= lattice vibrations) which describe the atomic motion. Magnetic scattering observes crystal field levels of one magnetic ion, magnons, excitons, charge density waves or solitons, which describe the dynamics of electronic spins in magnetic systems. The development of very high energy resolution in the  $\mu\text{eV}$  region permits the observation of rather slow processes like rotational and translational diffusion in solids, orientational tunnelling or the collective motion of long polymer chains or even biological molecules.

Polarizing the neutron beam and eventually analysing its spin direction after scattering gives access to the spin dependent scattering function, the matrix

$$S_{\alpha\beta}(Q, \omega).$$

This permits to separate the nuclear density correlations from those due to unpaired electronic spins. Through the spin dependence of the neutron-nucleus interaction this method even permits to look into the ordering of nuclear spins at very low temperatures.

To emphasize the impact of the method neutron scattering, we shortly follow the historical development as pictured in table 1. Starting in the early fifties, the power of research reactors increased by a factor of 10 per decade, the available flux density saturating at  $10^{15} \text{ cm}^{-2} \text{ s}^{-1}$  around 1970. Parallel to the development of the sources we follow the development of experimental techniques, starting with neutron diffraction at the relatively weak first generation of 1 MW reactors, then the birth of inelastic scattering at the medium size 10 MW reactors until the introduction of high resolution

Table 1.: History of Neutron Scattering: Sources, Experimental Techniques and Scientific Fields

| YEAR | SOURCE<br>Power Flux<br>MW $\text{cm}^{-2}\text{s}^{-1}$ | SCIENCE:                       | CRYSTALLOGRAPHY<br>1950                  | SOLID<br>STATE<br>1958   | MATERIAL<br>SCIENCE<br>1966 | CHEMISTRY<br>1968        | POLYMER<br>SCIENCE<br>1974 | BIOLOGY<br>1976                |
|------|----------------------------------------------------------|--------------------------------|------------------------------------------|--------------------------|-----------------------------|--------------------------|----------------------------|--------------------------------|
|      |                                                          | EXPERIMENTAL<br>TECHNIQUE      |                                          |                          |                             |                          |                            |                                |
| 1950 | 1 $10^{13}$                                              | Diffraction-<br>metry          | atomic structures<br>magnetic structures | x                        | x                           | x                        | x                          | proteins                       |
| 1960 | 10 $10^{14}$                                             | Inelastic<br>Scattering        |                                          |                          |                             |                          |                            |                                |
| 1970 | 57 $10^{15}$                                             | Small Angle<br>Scattering      |                                          |                          |                             |                          |                            |                                |
|      |                                                          | Diffuse<br>Scattering          |                                          |                          |                             |                          |                            |                                |
| ?    | $10^{16}$                                                | Hi Resolution<br>Inelastic Sc. |                                          | critical<br>fluctuations | alloys                      | diffusion,<br>tunnelling | chain motion               | slow motion<br>of biomolecules |

methods at the ILL high flux source. Parallel to the power of the sources we observe the development of experimental components like monochromator crystals, collimators, choppers, neutron guides, polarizers and multilayered mirrors and last not least position sensitive area detectors.

Looking back in history teaches us another important fact: neutron scattering is a multidisciplinary research method. Starting from crystallography, emerging from the long established X-ray crystallography, neutrons consecutively conquered the fields solid state physics, material research, chemistry, polymer science and molecular biology, such entering areas of basic fundamental as well as applied research.

What comes next? The development of sources obviously became stagnant after a few major high flux sources offered a flux density of  $10^{15} \text{ cm}^{-2} \text{ s}^{-1}$ . Does this mean there are now plenty of neutrons and further increase in source strength is no more urgent? But even in a high flux source the neutron density just reaches a "vacuum" of  $2 \cdot 10^{-8} \text{ mbar}$ , counter rates of inelastic experiments are expressed in  $\text{min}^{-1}$  rather than kHz. It is still the lack of intensity which prohibits a further amelioration of resolution, more precise or more systematic experiments.

It is only very recently, that the possibility had been discussed to design a very high flux reactor, with projected flux densities of  $5 \cdot 10^{15}$  by pushing the present reactor technology /1/. Following another new and very promising line, first the joint nuclear research centers KfK Karlsruhe and KFA Jülich /2/, and since 1982 the KFA alone have thoroughly studied the ambitious project to build the "Spallations-Neutronen-Quelle" SNQ /3/. Its main parameters are summarized in table 2.

The form of the neutron pulse is determined by the rectangular proton pulse of length  $t_p = 250 \text{ } \mu\text{s}$  and the decay time  $\tau$  of the neutrons within the moderators. The resulting pulse form is shown in figure 2 for both thermal and cold moderators. Beyond 0.1 eV  $\tau$  becomes very short, the neutron pulses become more rectangular.

Table 2 SNQ Parameters

|                                 |                       |                                                    |
|---------------------------------|-----------------------|----------------------------------------------------|
| Proton linac                    | 1100 MeV              | 5 mA                                               |
| Proton pulse length:            | 250 $\mu$ s at 100 Hz |                                                    |
| Target material                 | $^{238}\text{U}$      |                                                    |
| Moderator                       | thermal               | cold                                               |
|                                 | $\text{H}_2\text{O}$  | $\text{D}_2$ with C reflector                      |
| neutron pulse length fwhm.      | 260 $\mu$ s           | 390 $\mu$ s                                        |
| time averaged flux $\bar{\phi}$ | $1.2 \cdot 10^{15}$   | $0.8 \cdot 10^{15} \text{ cm}^{-2} \text{ s}^{-1}$ |
| peak flux $\hat{\phi}$          | $45 \cdot 10^{15}$    | $16 \cdot 10^{15} \text{ cm}^{-2} \text{ s}^{-1}$  |
| $\hat{\phi}/\bar{\phi}$         | 37                    | 20                                                 |
| moderator size                  | 10x10 $\text{cm}^2$   |                                                    |
| target block radius             | 6 m                   |                                                    |
| beam holes                      | 14 holes              | 2 beams each                                       |
| neutron guides                  | 5 cold                | 22 m                                               |
|                                 | 5 thermal             |                                                    |

In a future option it is foreseen to shorten the proton pulse to 200 ns in a compressor ring. This will improve  $\hat{\phi}/\bar{\phi}$  by a factor of 2 in the thermal regime. Enormous gains are expected for hot and epithermal neutrons.

While the average flux of  $1.2 \cdot 10^{15}$  is equivalent to the existing high flux reactor at the ILL, the flux density during the peak is enhanced by a factor 37. If it were just  $\phi$  which counts regardless of the pulse structure, this source would be an expensive way just not to copy a reactor. The major advantage of the SNQ is of course its pulse structure. Justifying this high effort is synonymous with properly utilizing the pulse structure. Ideally we aim to invent instruments, whose detectors permanently count relevant neutrons, which emerge from the peak flux  $\hat{\phi}$  during the pulse. This would correspond to a steady state source of  $\phi = 4 \cdot 10^{16}$ .

Interesting enough, time of flight instruments were quite popular in the early pioneering days of neutron scattering. Then they were soon overtaken by crystal spectrometers. The reason was the development of better mosaic crystals as monochromators and the clumsiness of handling the vast amount of data, typical for time of flight results. With modern computers, the last argument

lost all its grip and we expect a reappearance of TOF techniques in a wide variety. To develop a new generation of instruments, mostly based on novel ideas, is not an obvious nor trivial task, particularly because it requires to turn away from thinking in those experimental techniques, which were so successful at reactors. This process requires time and its own harmonic pace. Even if the SNQ is only to become operational a decade from now, it is by no means too early to initiate this rethinking process now. Secondly the case to build a SNQ stands and falls with its application. To prove how a proper instrumentation can make use of the enhanced flux during the pulse is a necessity.

To initiate this process two workshops have been held, sponsored by the SNQ project group of KFA Jülich, in Bad Kissingen and Maria Laach, during the course of 1984. Discussions were organized in five subgroups

- |                                        |              |
|----------------------------------------|--------------|
| • Diffraction                          | W. Prandl    |
| • Diffuse and Small Angle Scattering   | H. Stuhrmann |
| • Inelastic Scattering                 | W. Reichard  |
| • High Resolution Inelastic Scattering | D. Richter   |
| • Special Techniques                   | W. Schmatz   |

The results of these discussions are presented in the papers of this report.

The comparison SNQ versus reactor instruments had been based on the following rules:

1. For a specific application, hence a given resolution, design an instrument at a SNQ beamhole or neutron guide, based on the above defined performance of the linac SNQ, and optimize it such that it makes best use of the pulsed intensity. In this stage spatial interferences between neighbouring instruments should be neglected.
2. Similarly an idealized instrument at a reactor with identical resolution should be defined for the same application. Often, but not necessarily, this reactor instrument will resemble to one of ILL's instruments.
3. Compare intensities of the two "paper designs". This factor  $I(\text{SNQ})/I(\text{reactor})$  expresses much better a true "gain factor", than comparing a calculated SNQ intensity to real measured values at ILL instruments. Intensity estimates tend to be notoriously optimistic, therefore the latter method would unrealistically favour the pulsed instruments.

Further the following questions have been discussed:

1. How would the operation of the compressor ring in a later stage affect the sketched instrument?
2. What would be the ideal time structure, i.e. pulse length and repetition rate, for the above specified instrument, obviously without altering the limits, set by the linac performance? This question seems important in order not to loose flexibility by freezing parameters in an early design state.

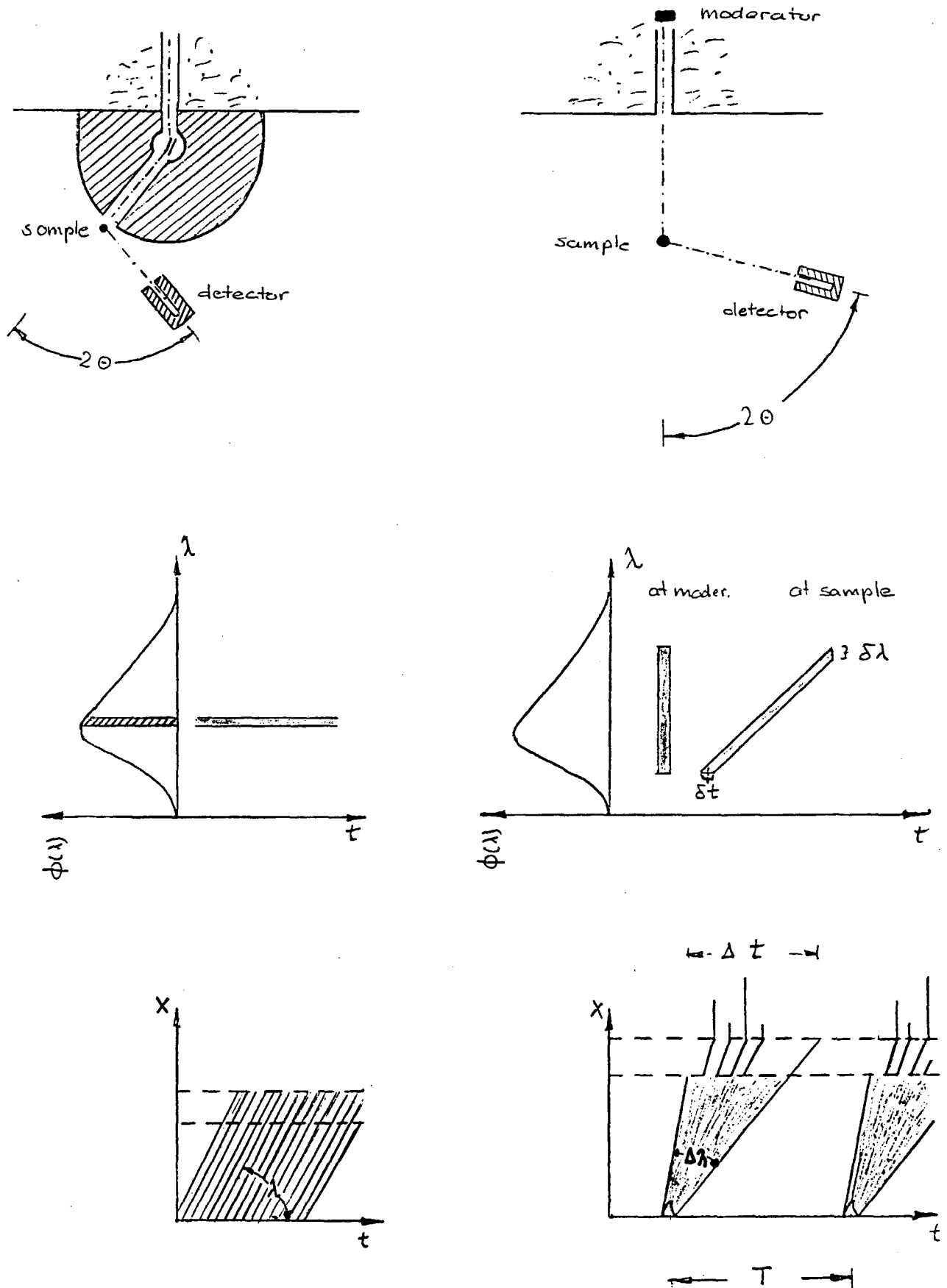
The papers collected in this issue comply more or less with these strikt rules. Dachs and Jauch /4/ for example, present a very thorough treatment of a TOF-Laue instrument designed for structure analysis with single crystalline samples, and compare it with a four circle crystal diffractometer with the same multidetector at a reactor. For specific samples, they present very detailed intensity estimates. Instruments of a similar design are already in operation at the pulsed spallation sources in ANL and Los Alamos.

Then we find novel ideas like a multi silicon analyzer backscattering instrument /5/, still in a design state, which, however, allows already to clearly evaluate its merits. On the other extreme new experimental components have been discussed like e.g. the time dependent "Drabkin" monochromator /6/ time focussing arrangements /7/, the acceleration of neutrons suffering Doppler shift from moving crystals /8/ or a triple axis spectrometer, whose analyzer, collimator (and even sample???) oscillate with 100 Hz /9/. The papers collected in this issue span the wide range from well established fields like diffraction or time of flight spectrometry, where sound and realistic intensity comparisons are presented, to a collection of new speculative ideas. It is just the upspringing of these "wild ideas" in the workshops, which demonstrated the liveliness of the community and gives great hope for future development.

Instead of reviewing all papers collected in this volume, we rather offer here a simplified access to the problem "time of flight versus steady state".

We take as an example powder diffraction, where one has to measure many Debye-Scherrer peaks with very good resolution, i.e. one scans the diffracted intensity  $I(Q)$  as a function of the momentum transfer

$$Q = \frac{4\pi}{\lambda} \cdot \sin \theta.$$



drawing: Thomas S.

Fig. 1 DC and TOF method, illustrated with the example of powder diffraction.

At a reactor the wavelength  $\lambda$  is fixed through a crystal monochromator, the intensity scattered in various angles  $2\theta$  is registered. At a pulsed source the principle is inverted. A white but shortly pulsed beam starts at the moderator or chopper, after some distance  $L$  different wavelengths impinge consecutively upon the sample. Neutrons scattered into a fixed angle  $2\theta$  are now registered as a function of time, where the flight time

$$t = \frac{h}{m} \frac{\lambda}{L} \quad \text{with } h/m = 3.96 \frac{\text{ms} \cdot \text{m}}{\text{\AA}}$$

now serves to identify their wavelength  $\lambda$ , hence  $Q$ . Both methods fixe  $\lambda$  and scan  $\theta$  at the reactor or the white beam technique fixe  $\theta$ , scan  $\lambda$  at a pulsed source give the same information and are principally equivalent.

They are illustrated in figure 1 in a  $\lambda$ - $t$  diagram as well as in the "neutron time table" distance versus time. The reactor instrument shines a narrow wavelength band  $\delta\lambda$  permanently on the sample, while at the pulsed machine the wavelength at the sample varies with time, the instantaneous resolution  $\delta\lambda$  being the same. This procedure is then repeated after  $T = 10$  ms. Very often the wavelength band  $\Delta\lambda$ , which can usefully be scanned, is restricted either by experimental reasons, like the need to suppress frame overlap or fast neutrons background, or only a limited  $\Delta\lambda$  contains interesting i.e. relevant data. Therefore the time  $\Delta t_{Z.P.}$ , during which the detector collects "publishable" information, is generally shorter than the pulse distance  $T$ , a fact, which we respect by defining the time utilization factor:

$$TUF = \frac{\Delta t_{Z.P.}}{T}$$

We now closer inspect the pulse form emerging from the moderators. G. Bauer /10/ describes the pulse by a single time constant  $\tau_m$ , the time a neutron stays within the moderator, by the expression

$$\begin{aligned} \phi(t) &= \frac{T}{t_p} \bar{\phi} (1 - e^{-t/\tau_m}) & t \leq t_p \\ \phi(t) &= \frac{T}{t_p} \underbrace{\bar{\phi} (1 - e^{-t_p/\tau_m})}_{\hat{\phi}} \cdot e^{-(t-t_p)/\tau_m} & t \geq t_p \end{aligned}$$



This pulse form is shown in figure 2 for both the thermal  $H_2O$  and a cold  $D_2$  moderator within a graphite reflector.

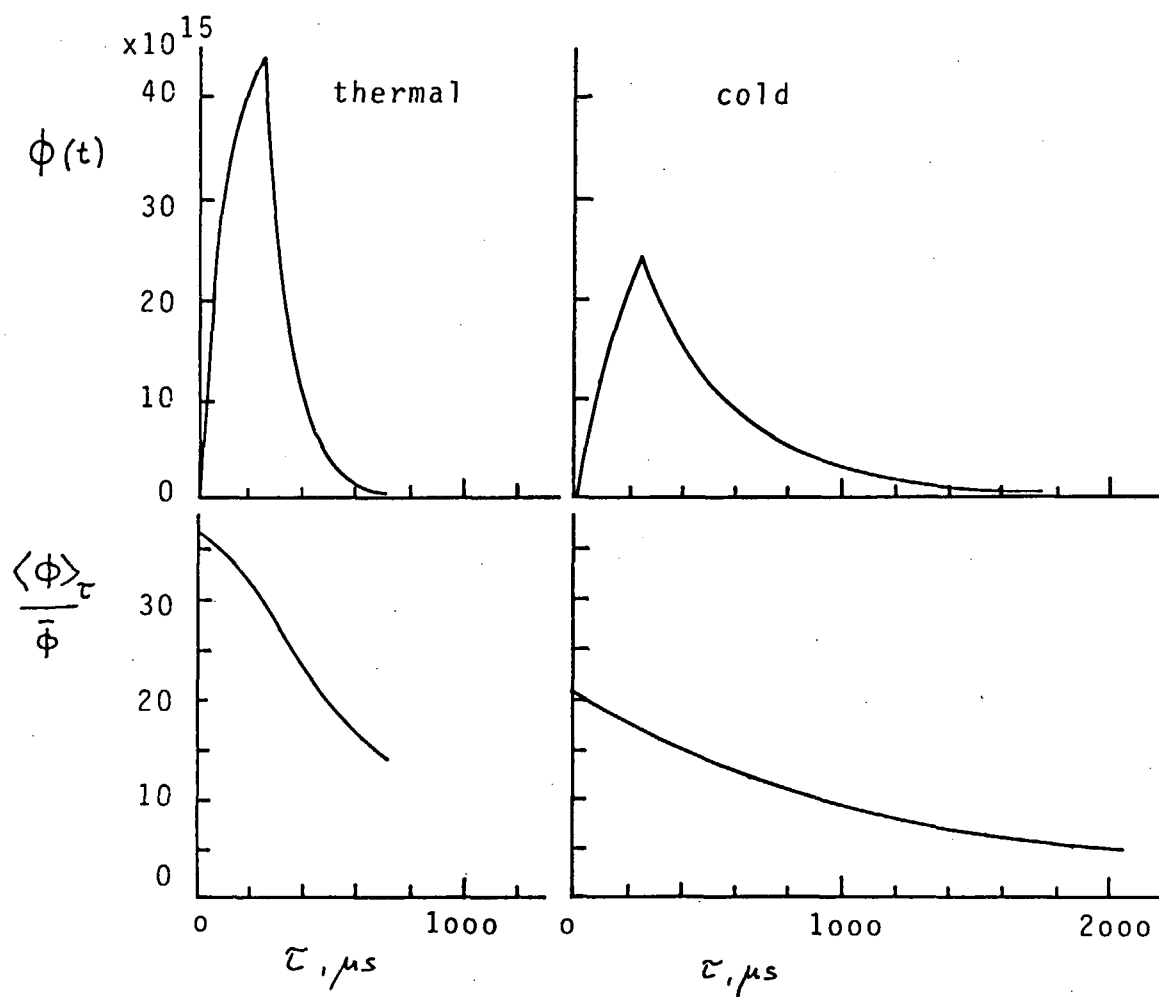


Fig. 2 : Pulse form  $\phi(t)$  from the thermal  $H_2O$  moderator and the cold  $D_2$  moderator in C reflector. Flux enhancement  $\langle \phi \rangle_\tau / \bar{\phi}$  as averaged over a pulse length  $\tau$  cut out from the moderator pulse.

|          | $\tau_M$    | $\tau_{1/2}$ (FWHM) | $\langle \phi \rangle_{\tau_{1/2}} / \bar{\phi}$ | $\hat{\phi} / \bar{\phi}$ |
|----------|-------------|---------------------|--------------------------------------------------|---------------------------|
| thermal: | 100 $\mu s$ | 260 $\mu s$         | 28                                               | 37                        |
| cold :   | 350 $\mu s$ | 390 $\mu s$         | 16                                               | 20                        |

Various instruments tend to use different pulse length  $\tau$ . Whereas a few use the complete moderator pulse, others, like high resolution time of flight arrangements, cut only 10  $\mu$ s from the pulse. In an intermediate case the half or even quarter width is used, while a simple auxiliary disk chopper cuts off the tail. It is convenient to replace the peak flux  $\hat{\phi}$  by the appropriately averaged quantity

$$\langle \phi \rangle_{\tau} = \frac{1}{\tau} \int_{t_1}^{t_1+\tau} \phi(t) dt \quad \rightarrow \quad \hat{\phi} \quad \text{for } \tau \rightarrow 0$$

The flux enhancement  $\langle \phi \rangle_{\tau} / \hat{\phi}$ , averaged over a pulse length  $\tau$  is illustrated in figure 2.

The useful intensity on the sample at the DC prototype instrument will be proportional to

$$I_{DC} \propto \hat{\phi} \delta\lambda \delta\Omega \cdot TUF_{DC}$$

The same quantity for a pulsed instrument, using a pulse  $\tau$ , is

$$I_p \propto \langle \phi \rangle_{\tau} \delta\lambda \delta\Omega \cdot \frac{\Delta t_{Z.P.}}{T}$$

Requiring identical resolution  $\delta\lambda$  for both cases we define a coarse gain factor  $G$

$$G = I_{DC} / I_p = \frac{\langle \phi \rangle_{\tau}}{\hat{\phi}} \cdot \frac{\Delta t_{Z.P.}}{T}$$

Here we assume, that a DC instrument can be utilized very economically with  $TUF = 1$  and set  $\delta\Omega_{DC} = \delta\Omega_p$ . Often the latter assumption is not quite justified. The moderator size of 10x10 cm<sup>2</sup> as well as the substantial 6 m shield of the target block allow larger solid angles at a reactor. A crystal instrument can further increase the intensity if a curved monochromator focusses a large beam onto a small sample. Intensity losses due to crystal reflectivity or chopper transmission are as well neglected. Nevertheless, we believe that the above defined gain factor is a simple, but reasonable measure to understand the basic virtues of different types of instruments. If we compare e.g. a TOF-Laue instrument to a four circle steady state machine we get a gain factor which agrees within a factor of 2 with Jauch's very detailed calculations /4/.

To better understand, how to make the best out of the pulse, we classify instruments according to their time of flight mode:

- Notorious DC instruments, carried over from the reactor, simply use the time average flux  $\bar{\phi}$ . Because of their well established merit to scan a specific excitation in single crystals we expect them to survive even at a pulsed source /9/. With the gain factor = 1, SNQ is just equal to a high flux reactor. But even here the pulse structure offers certain advantages. Gating the detector suppresses the background, distinguishes different order reflections from monochromator and analyzer (a main reason for one of the most frequent excitations, the "spurion") and allows even to deliberately use the  $\lambda/2$ .
- Real time of flight instruments, where the energy is defined by a short chopper pulse, fully profit from the peak flux  $\hat{\phi}$ . For classical inelastic instruments there is just one draw back: The SNQ repetition rate will not always coincide with the ideal chopper frequency.

Thus the gain factor becomes

$$G = \frac{\hat{\phi}}{\bar{\phi}} \cdot \frac{100 \text{ Hz}}{\nu_{\text{ideal}}}$$

which takes the following typical values

| neutron energy:           | thermal | cold   |
|---------------------------|---------|--------|
| $\hat{\phi} / \bar{\phi}$ | 35      | 25     |
| ideal $\nu$               | 250     | 100 Hz |
| typical global gain:      | 14      | 20     |

- Multiplexing: This is a completely new class of instruments which specifically use the long nearly uncut SNQ pulse. Consider a steady state instrument like e.g. a backscattering high resolution spectrometer. After the pulse has passed, the detector goes idle. Arranging n similar instrument components in such a way, that the neutron pulses enter or leave them at different times, allows to consecutively perform n nearly similar experiments. The flight time serves here to label the different results. As an example we refer to the multi silicon backscattering MUSIC /5/. The gain = 12 in table 3 is calculated for n = 11 monochromators, which each use the cold pulse over 450  $\mu\text{s}$  with  $\langle \phi \rangle / \phi = 15$ . Ideally such a multiplexing instrument uses nearly the peak flux continuously.

# TYPES OF SNQ NEUTRON SCATTERING INSTRUMENTS

$$\tau = \begin{cases} 120-300\mu s & \square \\ 440\mu s & \blacksquare \end{cases}$$

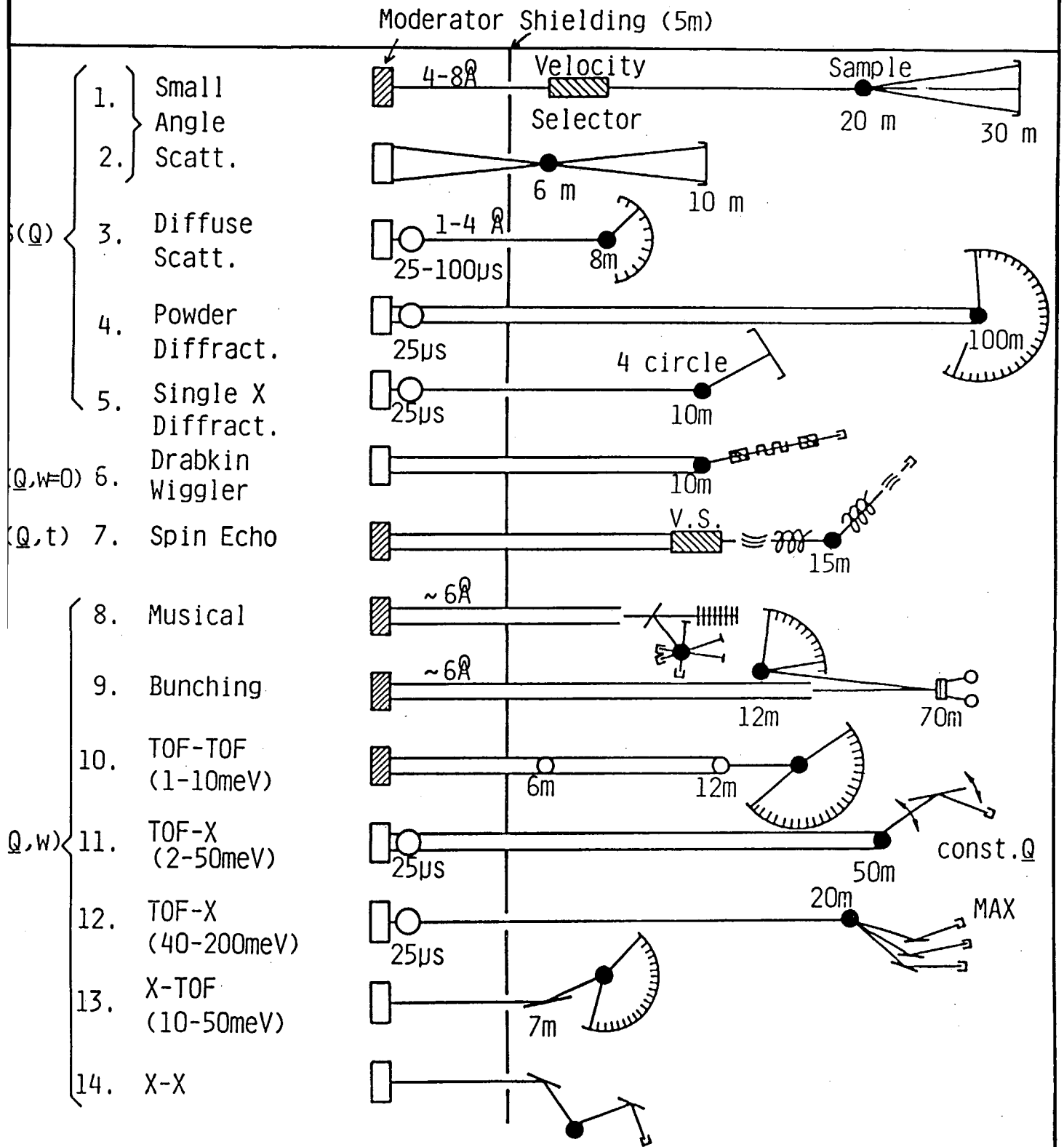


Fig. 3 Various types of Instruments proposed for the SNQ after C. Windsor /21/



At the last session the working groups in Maria Laach selected a number of instruments or instrumental principles to be proposed for the SNQ. These instruments are schematically shown in fig. 3 which has been compiled by C. Windsor. The summary of gain factors is given in table 3. They are partly taken from the corresponding papers in this volume partly based on arguments as given in this introductory article. Employing it with the restrictions mentioned before we believe that these numbers give a fairly honest picture of the possible advantages of properly designed instruments at the SNQ. We deliberately resist the seduction to average these gain factor. In most cases it is possible to make great benefit from the enhanced flux during the pulse and to achieve intensities which would be impossible even on a next generation high flux reactor.

How quantity turns into quality. A few words might be allowed to discuss the question whether more neutrons will produce new physics and not just "more of the same". In the introduction it has been laid out that neutron scattering principally measures the spin dependent scattering functions

$$S_{\alpha\beta}(\underline{Q},\omega) .$$

In view of the fact that it is not the performance of instruments, but only the notorious lack of neutron flux which prevents better measurements, we can firmly state that an improved pulsed source combined with an appropriate adapted instrumentation will allow to further push the limits of the experimental art. In particular we expect considerable progress on the following topics:

- better resolution in momentum  $\frac{dQ}{d\omega}$   
in energy
- polarization analysis  $S_{\alpha\beta}(\underline{Q},\omega)$
- space resolved  $S(Q,\omega;r) :$  surfaces, topography  
tomography
- time resolved  $S(Q,\omega;t) :$  kinetics of phase transitions  
and chemical reactions, off  
equilibrium system

- sample parameters temperature, pressure, magnetic or electric field
- smaller samples
- rare samples
- dilute systems
- more complex systems

It seems obvious that progress along these lines will unveil phenomena which nowadays are not yet or only very hardly accessible. In particular we expect from the SNQ, which has the chance to become the worlds best neutron source, an enormous impact on the development of new experimental methods. Therefore I dare say to state:

Intensities at SNQ will surpass today's standards by an order of magnitude. Combined with new experimental techniques this increase in intensity definitely will open the way to new science.

Acknowledgement: It is a great pleasure to express our gratitude to all those who made possible and participated in the workshops in Bad Kissingen and Maria Laach.

The KFA Jülich generously sponsored the workshops and many individual gatherings. The staff of the SNQ project group organized the meetings thus preparing a creative ambiance. Especially we want to thank all participants, who showed such great engagement and enthusiasm.

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## Introduction

Reflections on the SNQ Workshop, September 1984

Colin G Windsor, Bldg 418, MP & M Division, AERE, Harwell, OX11 0RA, UK

My thoughts are based around the list of 13 key instruments shown in figure 1. The list is perhaps best made by a visitor as any official list has dangers of promoting premature crystallisation. The list of instruments actually to be built should wait as long as possible. Nevertheless this list, based on the science we could do with SNQ if it existed today, has a value in our proposal for the project. It shows in concrete terms a possible configuration of the SNQ. I believe this is important if we wish to quote "gain figures" of SNQ with respect to a  $10^{15}$  flux reactor. These have almost no meaning for spectrometers working over unspecified energy ranges at unspecified resolutions. What is needed is a "typical experiment" for each proposed instrument. Sensible energy and resolution parameters can then be chosen from our experience, often on the  $10^{15}$  reactor. We can then calculate how each SNQ instrument would perform with the same parameters. The ratio of computed count rates for the two sources then defines the gain factor for that experiment on those instruments.

The instruments in the list refer to the source in its initial mode, say SNQ-L, for long pulses (and long wavelengths). In my opinion the compressor ring mode, say SNQ-S, with short pulses (and short wavelengths) requires a quite new instrument list drawn up on a clean sheet of paper at another workshop. Any instrument optimised for 300  $\mu$ s pulses can hardly be optimised for the  $2/\sqrt{E}$   $\mu$ s epithermal pulse widths possible with the compressor ring. The science possible in the two modes is different and complementary. I believe that when the time comes to build the compressor ring, the long wavelength users will demand that their experiments continue uninterrupted. Given that new instruments and moderators need to be built, the answer is to build two target stations sharing the protons on a 50-50 basis. Both modes would enjoy the reduction in frequency from 100 to 50 cycles.

The list shows only one instrument, the triple axis spectrometer, which uses the source in a continuous mode. The gain factor must be close to unity and one must question whether it deserves a precious SNQ beam hole. A TOF-X spectrometer can always be operated in a conventional constant Q mode by picking from the incident spectrum just that energy window which would have been chosen by the monochromator. The frequency scan is made sequentially. Nothing is lost compared to a triple axis. The time-of-flight scan is a bonus which can be used again to define the background and remove higher order analyser peaks. There are no higher order monochromator peaks to remove. This "triple axis analyser" would in

practice be an option on a MAX "host spectrometer".

The list shows only four of the remaining 12 instruments which use the natural SNQL pulse length without modification by choppers or velocity selectors. Have we chosen too long a pulse length? Possibly, but an alternative question is "have we chosen to concentrate on too high a neutron energy range?".

SNQ-L pulse length consequences

|                                       |                       |     |     |      |
|---------------------------------------|-----------------------|-----|-----|------|
| Incident energy                       | $E_o$ (meV)           | 50  | 5   | 0.5  |
| Wavelength                            | $\lambda_o$ (Å)       | 1.3 | 4.0 | 12.8 |
| Pulse length                          | $\Delta t$ ( $\mu$ s) | 300 | 440 | 440  |
| Moderator thickness                   | $\delta_m$ (cm)       | 93  | 43  | 14   |
| Resolution at $L_o = 20$ m            | R(%)                  | 9.3 | 4.3 | 1.4  |
| Flight path at $\Delta E_o/E_o = 2\%$ | L(m)                  | 93  | 43  | 14   |

The table shows the pulse time converted into an equivalent moderator thickness  $\delta_m = 0.3956 \Delta t/\lambda$ . The incident energy resolution is then given by  $\Delta E_o/E_o = 2\Delta t/t_o = 2\delta_m/L_o$ . The epithermal pulse widths  $\Delta t \approx 2/\sqrt{E}$   $\mu$ s possible with SNQ-S give a  $\delta_m$  of order 3 cm independent of energy leading to excellent 0.3% energy resolution at say 20 m flight paths. The nearly constant  $\Delta t$  of SNQ-L means that  $\delta_m$  decreases with energy to say 14 cm at 0.5 meV. Resolution at 20 m improves with decreasing energy reaching a useful 1.4% at 0.5 meV. The higher the energy at which we wish to work with given resolution, the more we must retreat in flight path, or the more of the available pulse needs to be chopped out by a rotor.

Some four of the 13 instruments demand a rotor close to the source which will chop the pulse without limiting the energy window. Ideally the chopper needs to give 2 to 100  $\mu$ s pulses over a 100 cm<sup>2</sup> beam area, with good transmission and without collimating the beam. This will be a crucial technical problem.

The majority of the SNQ-L instruments have gain factors greater than 5 over a 10<sup>15</sup> reactor and so represent world-beating performance even over Oak Ridge's proposed 5.10<sup>15</sup> reactor. The performance possible from the compressor ring SNQ-S source can be calculated by all from the figures in this years Physical Review Letters from IPNS. Simply multiply by 500 to extrapolate from 10  $\mu$ amp to 5mamp proton current! Sharing protons between the long and short pulse modes makes the SNQ the world's most powerful source over the complete neutron energy range.

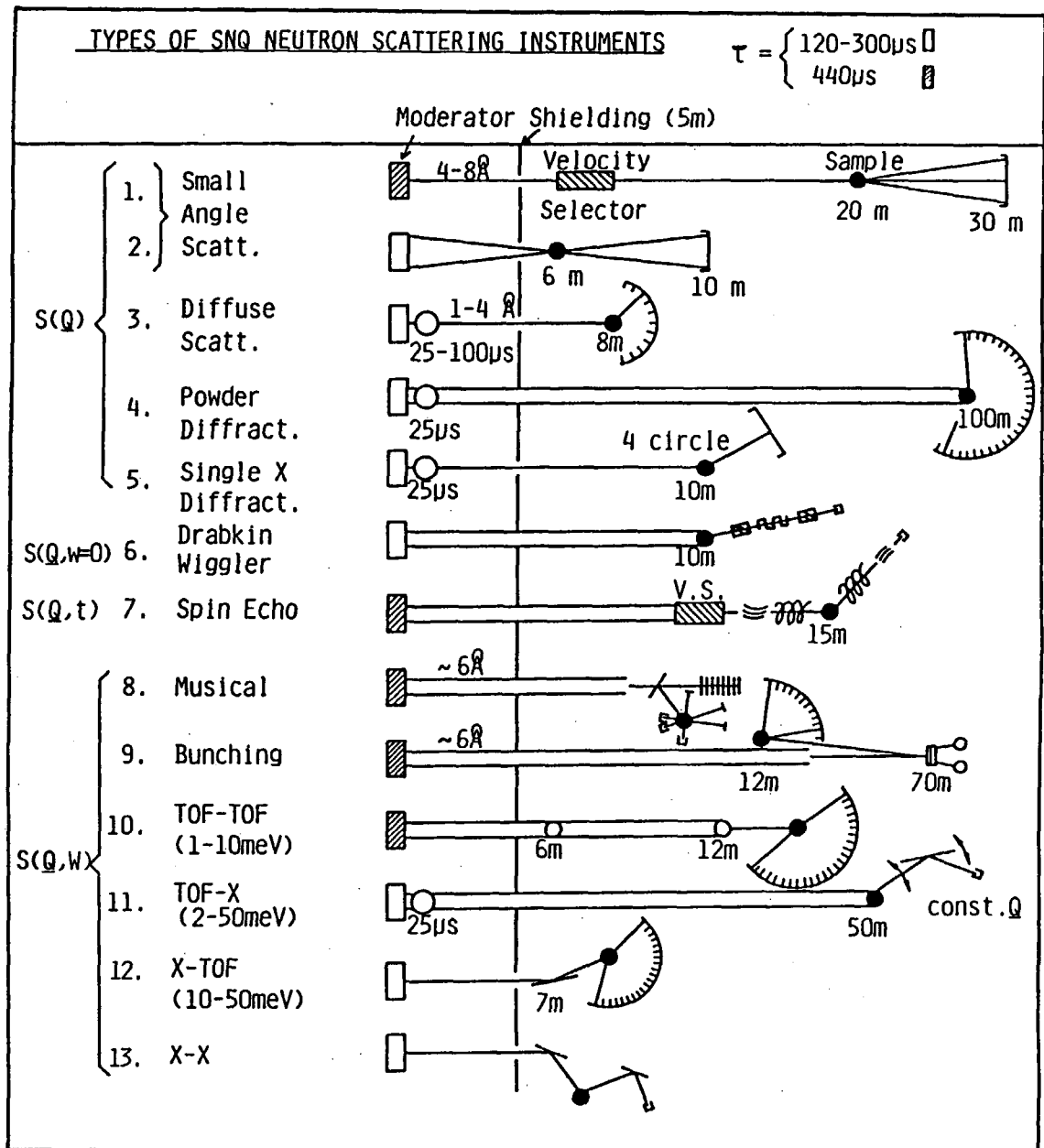


Fig. 1

This figure was hand-drawn by Collin Windsor at the Maria Laach workshop. The symbols mean:  $\square$  = moderator at ambient temperature;  $\blacksquare$  = cold moderator; — = beam without neutron guide;  $\equiv$  = beam with neutron guide;  $\circ$  = chopper;  $\bullet$  = sample;  $\square$  = two-dimensional detector;  $\square$  = detector;  $\diagup$  = reflecting crystal;  $\text{K}$  = drive for Doppler motion. Distances are counted from the moderator.

### Diffraction



# Some Aspects for the Design of a High Resolution TOF Powder Diffractometer at SNQ

by

E. Steichele, Fakultät für Physik, E 21  
Technische Universität München, 8046 Garching

## Introduction:

The development of high resolution neutron diffractometers and the advances in data analysis have widened the application of neutron powder diffraction during the last decade. Ferroelectrics, ionic conductors, hydrogen bonds, catalysts, minerals, magnetic materials and structural phase transitions as well as problems in practical material testing have been studied by neutron powder diffraction. There is no doubt, that this domain in the wide field of neutron scattering will have its importance by the time, when SNQ neutrons could be available. Moreover one could imagine that neutron diffraction could then be a routine method like NMR spectroscopy for many chemists.

Powder diffractometers can be subdivided into two classes: Intensity optimized machines, which give very high intensity with a moderate resolution of  $\Delta d/d \sim 10^{-2}$ , and resolution optimized instruments with very high resolution and still reasonable intensity. Only the latter type of a powder diffractometer will be dealt with in this chapter. The design is based on a resolution of  $\Delta d/d \sim 2 \times 10^{-4}$  for  $d$ -values of  $d \sim 1 \text{ \AA}$ . This resolution seems reasonable with respect to particle size broadening, it allows structure refinement in complex molecules with unit cells of about  $3000\text{-}4000 \text{ \AA}^3$  and/or about 500 structural parameters and ab initio structure determination by direct methods. A present-day design should moreover contain some reserves to guarantee the eager interest of users in 10-15 years from now.

### The Time-of-Flight (TOF)-Method of Neutron Diffraction.

Diffraction experiments are governed by Bragg's law  $\lambda = 2d_{hkl} \sin \theta$  and one is interested in a set of intensities  $I(d_{hkl})$ . In the classical diffraction technique one uses monochromatic neutrons and measures the count rate as a function of the scattering angle. To make use of the high peak flux at a pulsed source diffraction measurements have to be done with the TOF method. Here one uses neutrons of all wavelengths, produced during the burst, and measures the wavelength  $\lambda_{hkl}$  of the neutrons scattered by the lattice planes  $d_{hkl}$  under a fixed scattering angle  $2\theta$ . By normalization of the scattered neutron intensity to the spectrum of the incoming neutrons one gets an equivalent data set  $I(d_{hkl})$  as with the classical double-axis spectrometer (DAS).

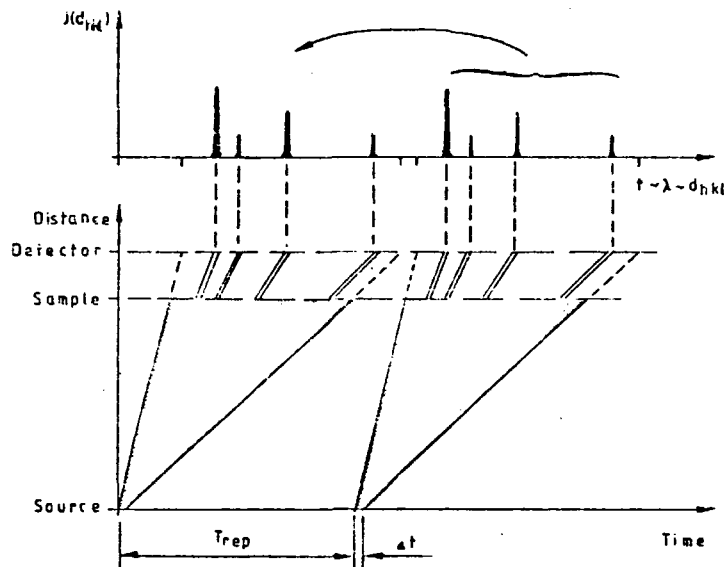


Fig. 1

The wavelength of Bragg reflected neutrons is measured via the time-of-flight over the distance source-detector using a multichannel time-analyser. Within the repetition time of the source ( $\sim 10$  msec) one scan over the whole diffraction pattern is being performed, which of course by intensity reasons has to be repeated many thousand times (Fig.1). Looking onto the scope of a multichannel time-analyser one can see the lines simultaneously growing up. This may be the origin of the misleading statement, that there is a big advantage and possibly a big gain of the TOF method due to the "simultaneous" measurement of many Bragg lines. This is not true. The time channels are scanned one after the other in a sequential and not in a parallel manner. The only way to measure many Bragg lines simultaneously is to use several detectors, each one looking

to a different range of reflections. The difference between the classical way and the TOF method would not seem so drastic if one could imagine to perform an angular scan in a classical powder instrument within 10 msec and to repeat these sweeps one million times per day. One therefore should be always aware, that there is no principal intensity gain by the TOF method over the classical method by "simultaneous" measurement of many Bragg lines.

The SNQ project is characterized by an average flux similar to the flux of a high flux reactor and a peak flux, which might be a factor 35 higher than the mean flux of the ILL reactor. Therefore intensity advantages can be obtained only, if the peak flux and not the mean flux of SNQ is relevant for a powder diffractometer. This is indeed the case as may be realized from fig. 2.

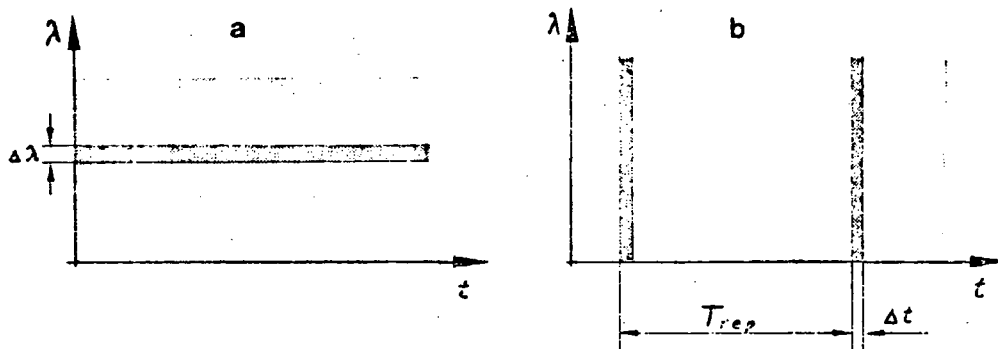


Fig.2

A steady state reactor produces neutrons of all wavelengths all the time. For a classical diffractometer one selects out a narrow wavelength band (fig.2a), the width  $\Delta\lambda$  of which contributes to the resolution of the instrument:

$$\Delta d/d = \left\{ (\Delta\lambda/\lambda)^2 + (\cot\theta\Delta\theta)^2 \right\}^{0.5}$$

For  $\Delta d/d = 1\%$  and  $\lambda$  about  $1 \text{ \AA}$  one gets  $\Delta\lambda \sim 0.01 \text{ \AA}$ . As the Maxwell spectrum can be roughly approximated by a rectangular distribution about  $1 \text{ \AA}$  wide one uses about 1 % of the neutrons at the exit of the beam tube.

For a TOF diffractometer one cuts out short pulses  $\Delta t$  wide, containing all wavelengths (fig.2b). The efficiency of the use of neutrons at the exit of a beam tube is again of the order of the instruments resolution, as  $\Delta d/d \sim \Delta\lambda/\lambda \sim \Delta t/T_{Tof}$ , and  $T_{Tof}$  is about  $T_{Rep}$ , the pulse repetition time. This means, that at a reactor the diffractometer intensities for a TOF and a double-

axis instrument are of the same order of magnitude. If however the peak flux at a pulsed source is higher than the constant flux at a reactor, the SNQ instrument gains by a factor  $\hat{\Phi}/\bar{\Phi}$  in first order.

Although there is no principal intensity gain in the TOF method, there may be practical intensity advantages due to experimental details. In a quantitative detailed analysis one has to compare the following parameters in the TOF and the classical method:

- ° Collimation in the primary and secondary beam, available and useful horizontal and vertical divergence
- ° Fraction of Debye-Scherrer ring, which can be accepted by stationary Soller collimators in a TOF arrangement and by movable collimators in a double axis instrument
- ° Peak reflectivity of a monochromator crystal and chopper transmission in a TOF instrument
- ° Transmission losses in guide tubes and Soller collimators
- °  $\lambda^4$  enhancement by the Lorentz factor in the white beam method

A complete intensity comparison between a TOF and a classical instrument at the same source (reactor), for the same sample volume and the same number of detectors has been performed some years ago (ILL report ISR 3/73 by Fender, Fuess, Steichele) and showed intensity gains within a factor 4 - 15 in favour of the TOF method. This may be partially compensated by the use of vertically bent monochromators in a modern classical diffractometer. A large vertical divergence, however, automatically limits the use of a large fraction of the whole Debye-Scherrer cone, which has been realized already in early TOF diffractometer set-ups. It is just the fact, that no detectors and collimators have to be moved, which opens the way to high intensity high resolution instruments by the TOF diffraction method. For low resolution instruments the beam divergence of a neutron guide is smaller than what one needs for an optimized design. For high resolution instruments, however, the collimation of a guide tube, which one needs for the long flight-path, fits quite well to the optimum primary divergence in

a backscattering arrangement and is, especially with conical trumpets (with super - mirror coating), larger than that of the 5' Soller collimators in modern double axis instruments. We therefore can make the statement: Given a steady state source a high resolution TOF diffractometer allows higher intensities than a classical instrument, mainly by the fact, that stationary detectors can accept a larger fraction of the scattered neutrons.

### Design of a High Resolution Diffractometer for SNQ

A prototype of a High Resolution TOF diffractometer has been built at the FRM in Garching. The primary pulses are produced by a drum rotor with  $\Delta t \sim 120 \mu\text{sec}$  time width. The resolution of the order  $\Delta d/d \sim 2 \times 10^{-3}$  for  $d_{hkl} \sim 1 \text{ \AA}$  was obtained by the use of a 150 m long neutron guide as a flight path and a backscattering arrangement of detectors near  $2\theta \sim 178^\circ$ . Two tail cutter choppers allow to select band widths of the incoming spectra between 0.1 and 2  $\text{\AA}$ .

A similar instrument with a guide tube and tail cutter choppers is being built at the SNS at Rutherford Appleton Laboratory, the pulse width being  $t \approx 10 \mu\text{sec}$ . In comparison with this value the pulse width of about 300  $\mu\text{sec}$  at SNQ is much too long for a high resolution instrument in the direct geometry. The only possibility is to use an additional chopper to reduce the pulsewidth. As shown in fig.3 the bandwidth of the transmitted neutrons is then determined only by the primary pulsewidth and the distance between the source and the chopper, when the chopper pulse is

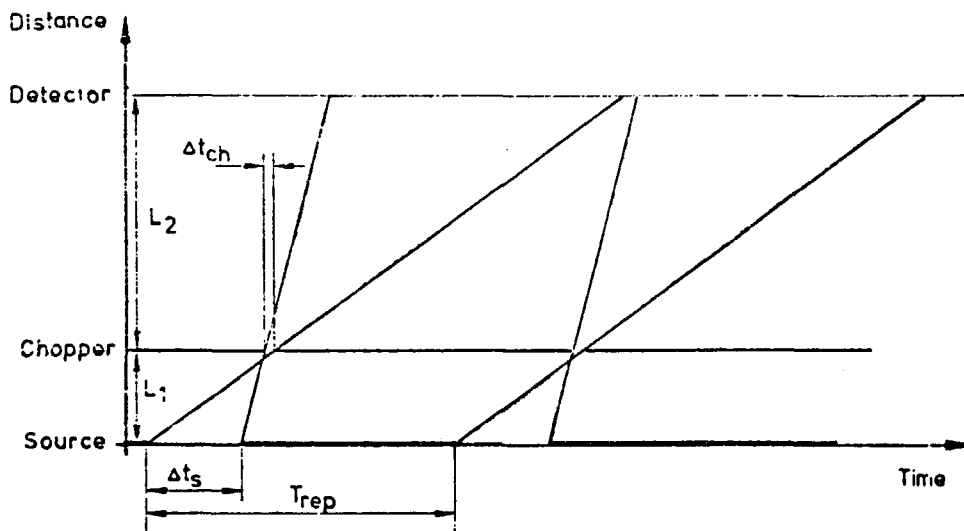


Fig.3

much shorter than the primary source pulse. In the optimum situation one uses each pulse and chooses a flight path length so that there are always useful neutrons at the sample. With a primary pulsewidth of 300  $\mu\text{sec}$  and a pulse repetition time of 10 msec one gets:

$$L_1/L_2 = 300 \mu\text{sec} / 10 \text{ msec} = 3/100$$

The length of the flight path is determined by the resolution wanted and the pulsewidth of the chopper. With  $\Delta t/t = 2 \times 10^{-4}$  for neutrons of 2 Å ( $v = 2000 \text{ m/sec}$ ) and  $\Delta t = 10 \mu\text{sec}$ , what is not too difficult to realize, one finds a flight path length of 200 m, which is quite realistic for a guide tube.

Reduction of the pulsewidth from 300  $\mu\text{sec}$  to 10  $\mu\text{sec}$  means, that only 3 % of the originally produced neutrons are used and there is no way around this. If one uses longer chopper pulses (20 - 30  $\mu\text{sec}$ ) the flightpath length has to be enlarged for a fixed resolution and consequently the wavelength spread in the beam has to be reduced to avoid frame overlap. The situation would improve, however, by use of a compressor ring: The instrument would profit from the higher peak flux and the efficiency factor (neutrons used/neutrons produced) would become higher. The low efficiency factor is the price to be paid for the high resolution. This efficiency factor is better in a source like SNS, but nevertheless an instrument at SNQ would be superior by the ratio of the peak fluxes.

The present design is characterized by a very narrow wavelength band  $\Delta\lambda = 0.2 \text{ Å}$  in the primary spectrum. With respect to a typical diffraction pattern as shown in fig.4 this means, that a complete pattern has to be taken in about 20 slightly overlapping frames or more. For each frame one has to take the primary spectrum and perform a normalization to the scattered spectrum. The high number of frames to be taken is a consequence of the high repetition rate of primary pulses. A reduction of the repetition rate, coupled with an increase of the peak intensity, would be a gain for powder diffraction at SNQ. A reduction of the pulse rate by an additional chopper would simplify somewhat the data evaluation without change of intensity. In this case, however, the "timing chopper" must be nearer to the source, i.e. inside the shielding. As the in-shield chopper cannot be nearer than 1.5 - 2m to the source the maximum bandwidth is

limited to about 0.6 - 0.8 Å. Instead of taking separate frames with fixed phase relations between the source and the chopper pulse one could think of a different way of data collection: Spallation source and chopper are running with different frequencies, so that for each source pulse the phase to the chopper pulse and hence the center of the wavelength band is different. This procedure might need a more complicated organisation in data storage, but would simplify the data collection.

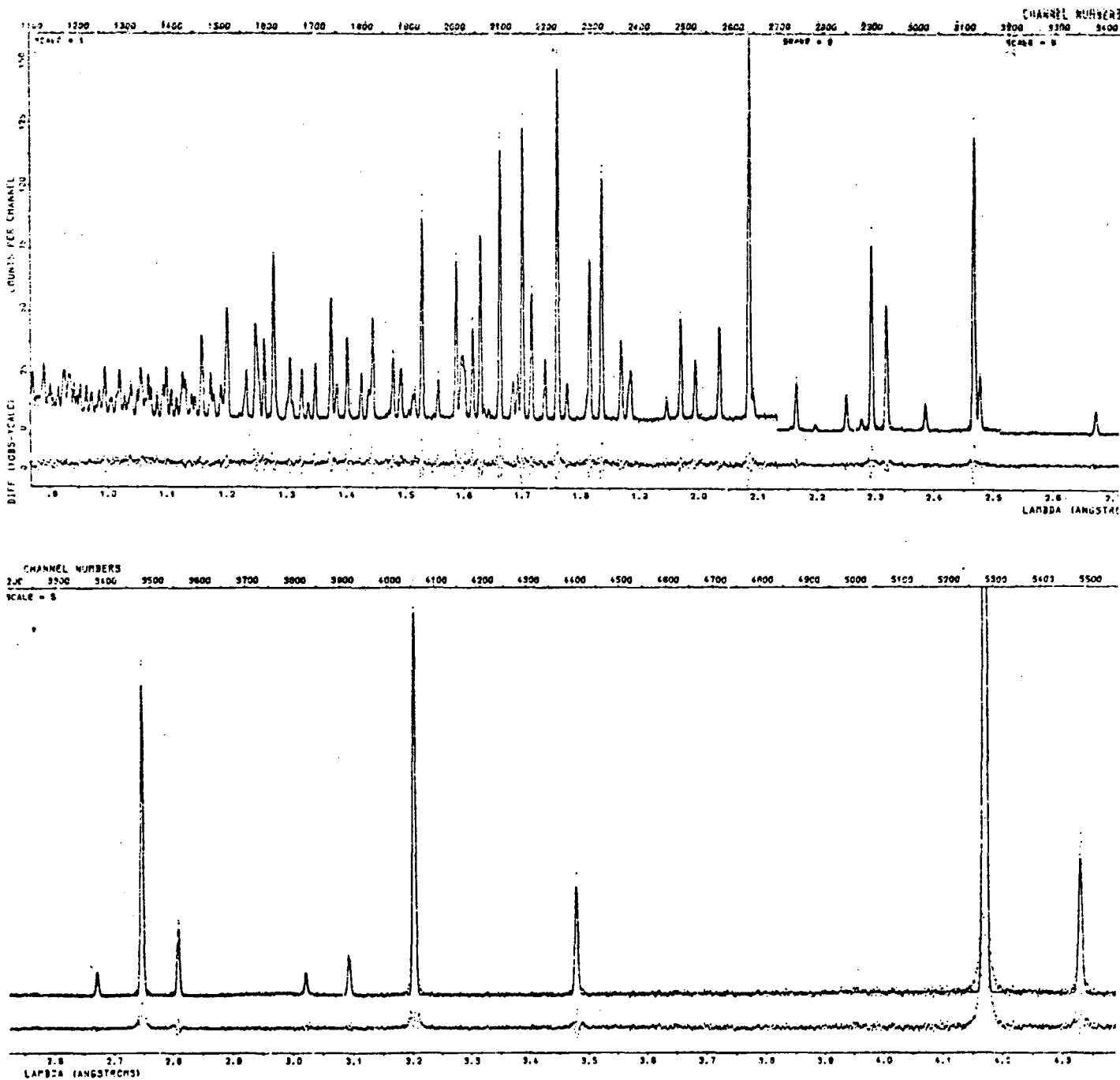


Fig. 4

According to formula (1) the best resolution is obtained with a back-scattering detector and in a good design the wavelength and angular part of the resolution function are about equal. With a flight path length of about 200 m resolution terms due to the thickness of the sample can always be neglected. For a fixed detector geometry the angular term is constant, the wavelength term is inversely proportional to  $d$  or  $\lambda$  ( $\Delta\lambda/\lambda \sim \Delta t/t$ ,  $\Delta t = \text{const.}$ ) and the total resolution function of the SNQ instrument with one detector is as shown in fig. 5a.

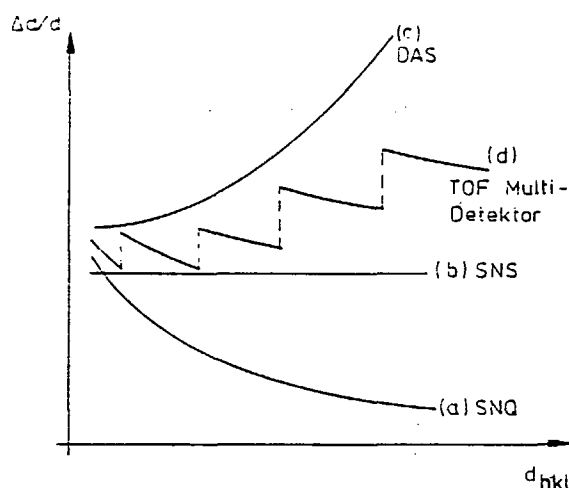


Fig. 5

This is slightly different from the resolution function of an instrument at SNS, where the pulselength  $t$  is nearly proportional to the wavelength, hence both terms of the resolution function are constant and the resolution function is independent of  $d_{hkl}$  as shown in fig. 5b. For a classical diffractometer the resolution function is mainly determined by the angular part and looks as shown in fig. 5c. It is sometimes argued that this is the ideal resolution function, as it corresponds to the line density in a powder pattern. This is true only, if with the relaxation of resolution an intensity gain and not only a smearing of intensity into a wider angular range is coupled. Moreover there are seldom intensity problems with low order reflections in TOF powder diffraction and the high resolution gives additional informations on particle size broadening, unit cell distortions etc. Therefore the resolution function as in fig. 5a is certainly an advantage and no handicap for the TOF diffraction.

An instrument with one backscattering detector allows all types of experiments, but it is not the most economic arrangement. The instrument can be much improved by installation of additional counters. The measure-



ment is then performed with one fixed wavelength band, say  $0.2 \text{ \AA}$ , and the different frames are collected by a corresponding number of detectors. The angular position of the detectors are chosen in a way, that the diffraction frames from two neighboured counters slightly overlap. With a primary spectrum from  $1.0$  to  $1.2 \text{ \AA}$  and an angular arrangement as shown in fig.6 one can collect all diffraction peaks within  $2 \text{ \AA} = 2d_{hkl} = 9 \text{ \AA}$ . Of course all detectors can be made as circular ring detectors and thus whole Debye-Scherrer rings can be accepted. The resolution function of the

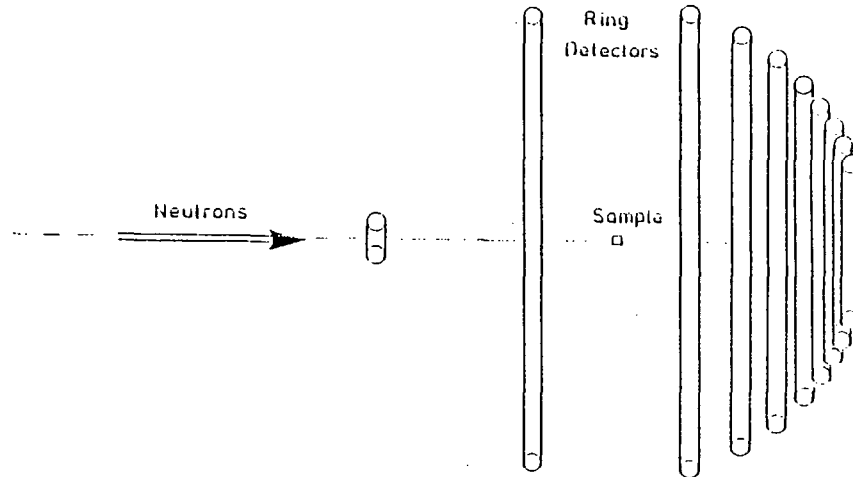


Fig. 6

instrument is then mainly determined by the angular terms and looks like fig. 5d, if the collimators for all detectors have the same divergence. By a different choice of collimator divergences the resolution function can be modified to any other shape. The data-set to be evaluated then looks as shown in fig.7. The linewidth within each frame is different from that in the neighboured frame. This situation is not unlike to that in conventional diffractometer data, where one has to work with angular dependent

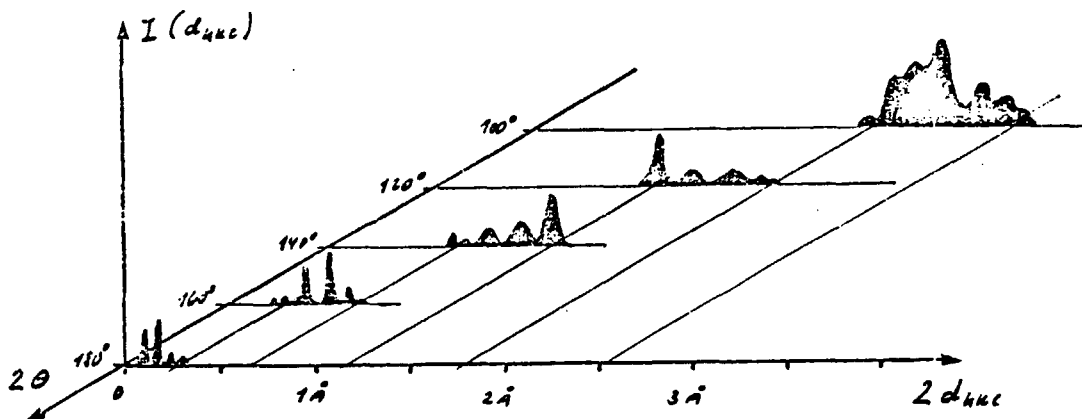


Fig. 7

line shapes in the Rietveld profile analysis. As all reflections are measured with neutrons within a very narrow wavelength band all problems with wavelength dependent absorption or extinction corrections, as sometimes typical for the TOF-diffraction method, disappear. Such an instrument would give very high intensity due to the many detector rings and it could be used very flexibly. The  $d_{hkl}$ -region with optimum resolution can simply be chosen by the phase between source and chopper pulse.

It might be interesting to compare such a multidetector TOF instrument with a multidetector classical double-axis-spectrometer as the D2B being installed at the ILL. Both instruments would have detectors at a large number of scattering angles. In the D2B instrument one has a fixed highly monochromated primary wavelength and makes high resolution (5' collimator divergence) angular scans over a small angular range ( $5^\circ$ ). In a multidetector TOF-instruments one has fixed well defined scattering angles and one performs high resolution wavelength scans over a small wavelength range ( $\Delta\lambda \approx 0.2 \text{ \AA}$ ). In a classical diffractometer high resolution powder data can be taken only on one side of the instrument because of the asymmetry of the phase space element behind the monochromator. Because of the vertical focussing from the monochromator to the sample only a small fraction of the Debye-Scherrer ring can be accepted. A TOF-instrument is axially completely symmetric and a complete Debye-Scherrer rings can be accepted.

#### Gain factor at SNQ

The TOF method with stationary detectors and axially symmetric geometry allows intensity gains compared to the classical double axis instrument, which in the final analysis depend on the number of detectors which can be installed. Of course, such a TOF-instrument could also be built at a reactor and the question is, what the effective gain factor at the SNQ would be. In first order the gain factor is the peak-flux/mean-flux ratio between SNQ and the reactor, which might be a factor 35 compared to the ILL reactor. This factor has to be, however, slightly reduced from the following reason. In a reactor instrument with a tail-cutter chopper for the wavelength band selection the transmission curve of this secondary chopper is nearly trapezoidal, which gives nearly full transmission for

all neutrons in the time-window. At the SNQ the primary pulse shape is more triangular than trapezoidal and that means a stronger attenuation of those neutrons near the ends of the selected wavelength band. This effect is wavelength dependent. Therefore we estimate a gain factor of 25 to 30 for a SNQ-TOF powder diffractometer compared to a similar instrument at the ILL reactor. This factor can be taken into account for normal powder diffraction. In all cases, where one is interested in the study of single lines, the pulse repetition time should be shorter and can be made shorter at a reactor instrument by a factor 3 to 4, the gain factor for the SNQ instrument should be correspondingly reduced for these special experiments.

Overview on Real Time Diffractometry at Wide Angles

and

Proposal for the Construction of an Intensity Optimized

Powder Diffractometer at the SNQ

C. Riekkel

Institut für Technische und Makromolekulare Chemie,  
Universität Hamburg, Martin-Luther-King Platz 6,  
D-2000 Hamburg 13, Fed. Rep. Germany

## I. Introduction

The dynamics of structural processes during phase transitions inclusive chemical reactions can be investigated by real time diffractometry<sup>1,2,3,4</sup> provided that the measuring time per spectrum -  $t_s$  - is sufficiently small.

In order to obtain a sufficient temporal resolution for a phase transformation - the extent of which can be quantified by a variable  $\alpha$  ( $0 < \alpha < 1$ ) - one has to demand that  $t_s \ll \tau$  where  $\alpha = 0.5$  at time  $\tau$  (Fig. 1).

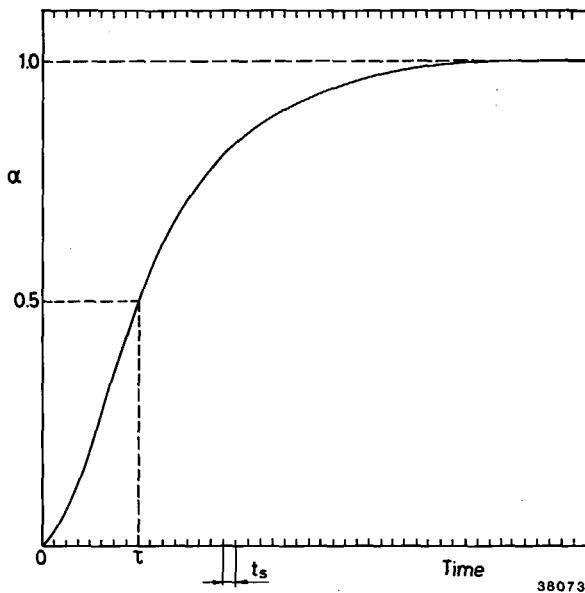


Fig. 1

The extent of a phase transformation can be characterized by a variable  $\alpha$ . The dynamics of the structural transformation can be investigated if the measuring time per spectrum -  $t_s$  - is considerably smaller than the half-time of the process  $\tau$ . In the hypothetical case shown one could have increased  $t_s$  with time in order to maintain the ratio  $\Delta\alpha/t_s$  constant. As  $\alpha(t)$  is generally unknown prior to the experiment this can only be done as part of the data reduction.

In contrast to macroscopic methods, real time diffractometry belongs to the class of microscopic methods. It differs, however, from other methods which probe the change in local order as it is sensitive to a change in long range order which is a cooperative process. Thus in the case of a nucleation controlled phase transformation one may try to learn not only something about the kinetics of the growth of the new phase but also on the size and the structure of the nuclei growing. New insight into the dynamics of phase transformations are therefore possible.

Real time diffractometry has been performed by neutron and X-ray scattering (especially synchrotron radiation scattering) both at wide- and small angles<sup>1-4</sup>. Although these methods are complimentary, mainly neutron wide-angle experiments will be discussed in the following with the aim of demonstrating the interest in an intensity optimized diffractometer for powders and highly disordered materials at the SNQ.

## II. Scope of Real Time Diffractometry

### II.1 General

The analysis of the temporal change of wide-angle spectra can be performed under different aspects:

- (i) The rate law of the formation (and/or) destruction of each crystalline phase can be determined. Only one reflection per phase has to be analyzed in principle.
- (ii) The mechanism of the phase transformation can be derived from the sequence of crystalline phases. Every reflection appearing in a given Q-range has to be analyzed although the sequence can again be determined from only one reflection per phase. The Q-range to be analyzed depends on the complexity of the phase mixture.
- (iii) Structural models for transient phases can be developed. This may imply only geometric models based on the positions of selected reflections or model calculations based on intensities. The accuracy of the model depends on the Q-range explored.

### II.2 Methodology

Both irreversible and reversible phase transformations can be investigated. In the case of irreversible transformations the temporal resolution is mainly determined by the flux of the source as all other parameters, i.e. the sample volume, the Q-range containing statistically significant reflections and often also the time necessary to perturb the sample depend on the available flux.

In the case of a reversible transformation, however, a repetitive perturbation and an accumulation of spectra allows a reduction of  $t_s$ . Thus if a reflection could be measured with a given intensity within 10 s, a reduction of  $t_s$

to  $10^{-3}$  s demands the accumulation of  $10^4$  spectra. Set aside a limitation by the sample on the number of repetitions - which is especially the case for biological material - a further limitation can be the repetition rate of the perturbation. Thus high power lasers operate at about 100 Hz which would increase the total measuring time in the case above already to 110 s. If the limiting temporal resolution of  $\sim 15$  ns shall be reached - given by the temporal width of the laser pulse - the flux of the source must be sufficiently intense.

In the case of a pulsed source the temporal width of the source -  $t_w$  - has also to be considered,  $t_w$  being the ultimate limit of the temporal resolution as the source pulse is used to trigger the perturbation.

Two limiting cases can be distinguished depending on the time scale of the structural transformation and the "temporal distance" between two pulses ( $t_p$ ):

- (i)  $\tau > t_p$ ; in this case the number of spectra which can be measured at different  $\alpha$ -values depends on the ratio  $\tau/t_p$ . With increasing ratio the condition of a steady state source approaches and it is no longer possible to trigger the perturbation by the source pulse.
- (ii)  $\tau < t_p$ ; spectra at different  $\alpha$ -values can only be recorded if the delay between the triggering pulse and the perturbation is varied (Fig. 2).

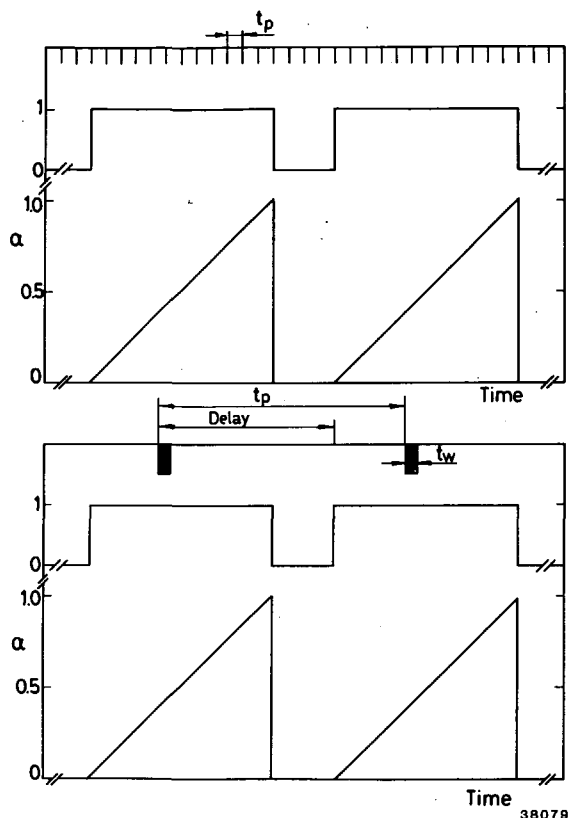
Such perturbations can be due to heating, electric or magnetic fields, pressure changes, changes in chemical potential etc..

Basically three different waveshapes for the perturbation have been used:

- (i) A sequence of short, ideally symmetric pulses is used to create transient states which relax to the initial state. Examples are laser flash initiated phase transformations<sup>5,6</sup>.
- (ii) Rectangular pulses are used in case the transient state builds up slowly after the perturbation. An example is the polarization reversal of  $\text{NaNO}_2$ <sup>7</sup>.
- (iii) Sine-wave perturbations are used as a stroboscopic technique in order to determine dispersion regions. An example is the reversible stretching of polymers like rubber<sup>8</sup>.

Fig. 2

$\alpha(t)$  for a hypothetical reversible phase transformation. At each cycle a perturbation pulse is switched on (0: no perturbation, 1: max. perturbation) and switched off at the end of a cycle. In the upper figure a process with  $\tau > t_p$  and in the lower figure with  $\tau < t_p$  are shown. Note that a homogeneous distribution of the perturbation across the sample is assumed within a negligible time.



### II.3 Neutron versus X-Ray Diffraction

The maximum flux available at the sample position on a powder diffractometer at the HFR-Grenoble is  $7 \times 10^7 \text{ n cm}^{-2} \text{ s}^{-1}$  ( $\lambda = 1.22 \text{ \AA}$ )<sup>9</sup> while the number of photons at the sample position for a diffractometer at the storage ring DORIS is  $\sim 10^{13} \text{ ph. cm}^{-2} \text{ s}^{-1}$ <sup>10</sup>. In the latter case a wavelength of  $1.45 \text{ \AA}$  is obtained from a bending magnet and monochromatized by a bent Ge(111) crystal.

Irreversible phase transitions of polymers can be studied at DORIS with  $t_s$ -values of a few seconds with an illuminated samples area of  $\sim 0.8 \text{ mm}^2$  and a sample thickness of  $10 - 20 \text{ }\mu\text{m}$ <sup>4</sup>. Comparable  $t_s$ -values can in principle be reached by a factor  $\sim 10^5$  larger sample volume in neutron diffraction which is not unusual<sup>1</sup>. Note, however, that the use of wigglers and detectors with a higher count rate capability will allow to reduce the  $t_s$ -values in synchrotron radiation scattering to the ms-range for irreversible processes.

Given the larger sample volumes necessary in real time neutron diffractometry a major problem will be to perturb the sample sufficiently quick. In practise, a compromise between the temporal resolution, the structural resolution,



and the sample volume has to be made. Any increase in intensity will therefore create new experimental possibilities.

Set aside this problem, the difference in scattering length makes neutron diffraction a complimentary technique especially in the case of heterogeneous chemical reactions involving a mixture of heavy and light atom reactants. Furthermore, special environment experiments are favoured in view of the low absorption cross sections of the neutrons. This is also a considerable advantage in solid/gas reactions as corrections due to the change in transmission can often be neglected in first order.

### III. Present Experimental Results and Possibilities

#### III.1 Irreversible Transformations

Neutron real time diffractometry has been performed almost exclusively at the I.L.L. using the position sensitive detector D1B<sup>9</sup> which has a flux of  $1.6 \times 10^6 \text{ n cm}^{-2} \text{ s}^{-1}$  at  $\lambda = 2.52 \text{ \AA}$ . If the analysis is limited to the strongest Bragg peaks,  $t_s$ -values of a few minutes are possible<sup>1</sup>.

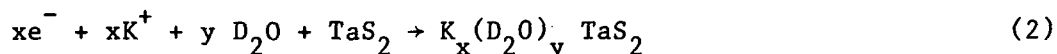
A typical example for a kinetic experiment is shown in Fig. 3 for the reaction of polyacetylene with gaseous  $\text{SbF}_5$  according to<sup>11</sup>



where the degree of reaction -  $\alpha$  - has been determined from the decrease of the  $[\text{CD}]_n$ -102 reflection ( $t_s = 300 \text{ s}$ ). These data were found to apply closely to a diffusion controlled process.

Although the transformation rate in diffusion controlled processes can be varied with the temperature, a detailed study of the kinetics is usually not possible in view of the restricted beam time. Other processes like Martensitic (diffusionless) transformations can occur on time scales of minutes or seconds and are not accessible.

The necessary large sample volumes complicate especially electrochemical reactions and demand slow transformations. Thus the reaction<sup>1</sup>



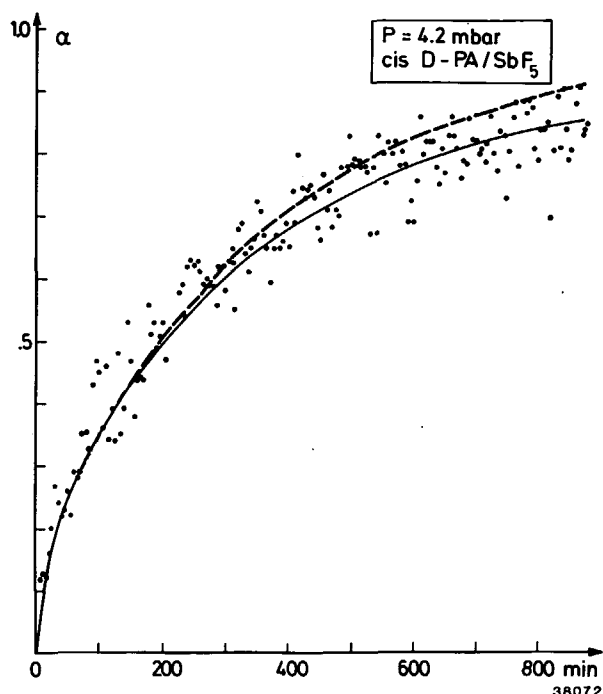


Fig. 3

Variation of the degree of reaction -  $\alpha$  - during the reaction of polyacetylene with gaseous  $\text{SbF}_5$ <sup>11</sup>. has been determined from the decrease of a polyacetylene reflection ( $t_s = 300$  s). The solid line is a guide line for the eye; the barred line has been calculated for a diffusion model with  $D = 10^{-9} \text{ cm}^2 \text{ s}^{-1}$ .

was performed galvanostatically with a 4 g (16 mmole)  $\text{TaS}_2$ -electrode ( $x_{\text{max}} = 0.33$ ;  $t_s = 900$  s). Small powder particles can complicate the homogeneity of solid/gas reactions.

The angular range of  $80^\circ$  ( $2\theta$ ) of the DIB-PSD is sufficient for most kinetic experiments but is already too small for a number of mechanistic and structural studies. Thus the analysis of the reaction of polyacetylene with  $\text{I}_2$  according to<sup>12</sup>



demands an analysis of reflections in the range  $0.6 \text{ \AA}^{-1} < Q < 6 \text{ \AA}^{-1}$  while the range available for one detector position will be  $0.6 \text{ \AA}^{-1} < Q < 3.6 \text{ \AA}^{-1}$  (Fig. 4). A maximum  $Q$ -value of  $\sim 4.8 \text{ \AA}^{-1}$  can be reached with this detector ( $\lambda = 2.52 \text{ \AA}$ ).

The temporal resolution decreases of course with increasing  $Q$ -value due to a reduction in reflection intensities. This limits also model calculations on transient phases as usually only few strong Bragg peaks can be evaluated<sup>1</sup>. Note, however, that data on intercalation reactions on transition metal dichalcogenides show rather disordered transient phases (Fig. 5) which suggest that the structural resolution could be more limited as compared to stable phases.

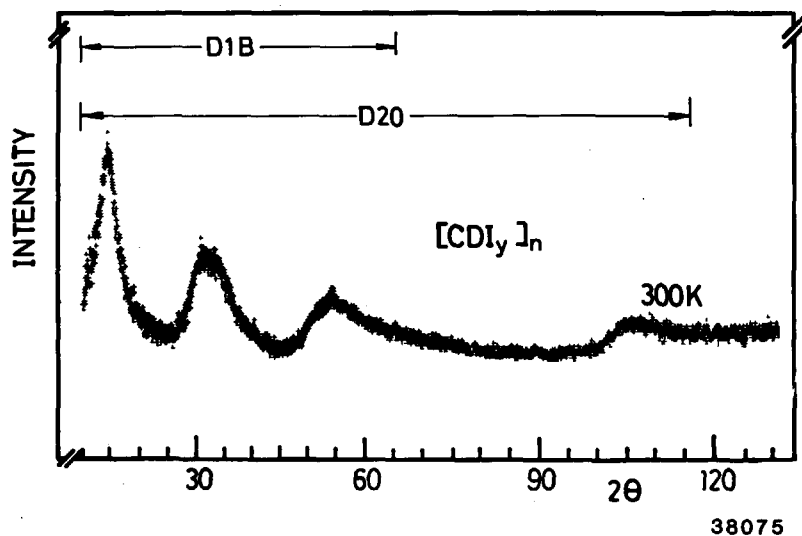


Fig. 4

Diffraction pattern of  $[\text{CDI}_y]_n$  ( $y = 0.17$ ) measured with  $\lambda = 1.909 \text{ \AA}$ . In order to study the formation of this phase from  $[\text{CD}]_n$ , reflections in the range  $0.6 \text{ \AA} < Q < 6 \text{ \AA}$  have to be studied. The angular ranges indicated for the D1B and D20 PSDs<sup>9</sup> correspond to one detector position.

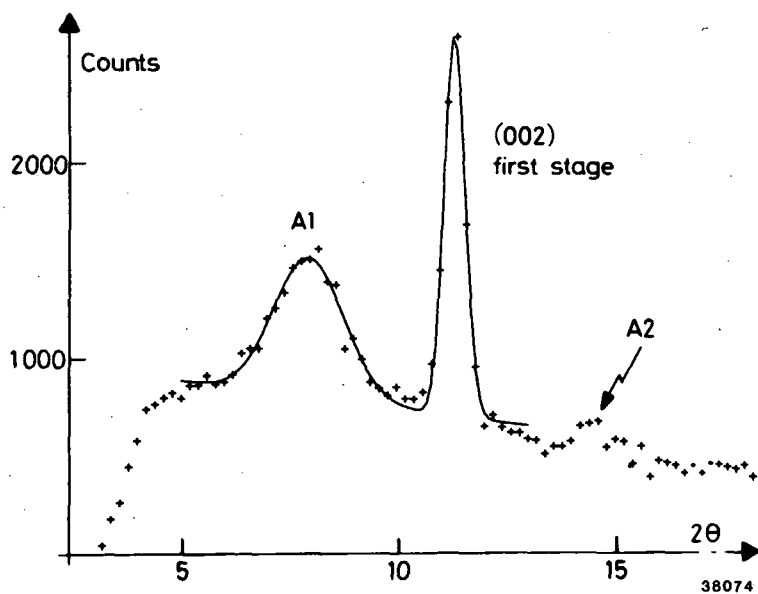


Fig. 5

The reaction of  $\text{TaS}_2$  with liquid  $\text{C}_5\text{D}_5\text{N}$  proceeds via an intermediate phase  $\text{TaS}_2(\text{C}_5\text{D}_5\text{N})_{0.25}$  to the first stage phase  $\text{TaS}_2(\text{C}_5\text{D}_5\text{N})_{0.5}$ .<sup>1</sup> The spectrum measured with the D1B-PSD shows the 002-reflection of the first stage phase and the reflections A1/A2 due to the intermediate phase ( $t_s = 300 \text{ s}$ ).

Some progress is expected from the  $64^\circ$  D20-PSD at the HFR-Grenoble<sup>9</sup> in terms of flux at the sample and  $Q$ -range. In the case of  $[\text{CDI}_y]_n$  (Fig. 4) at  $\lambda = 1.22 \text{ \AA}$  (max. flux) a range of  $0.6 \text{ \AA}^{-1} < Q < 5.6 \text{ \AA}^{-1}$  and a factor  $\sim 50$  higher flux at the sample as compared to the D1B-PSD will be possible.

### III.2 Reversible Transformations

Apparently no reversible phase transformations at wide angles have been studied on a steady state neutron source although the reversible stretching of polymers could well be studied similar to a neutron small angle scattering experiment<sup>13</sup>.

A pulsed source has been used to study the polarization reversal of  $\text{NaNO}_2$  in an electric field using rectangular perturbation-pulses and  $t_s = 10 \text{ ms}$ . Note that the sample size may also be important for such experiments as programmable voltage generators can be obtained only up to  $\sim 1 \text{ kV}$  which necessitates a reduction of the sample thickness for high field strength.

## IV. An Intensity Optimized Powder-Diffractometer at the SNQ

### IV.1 General

The previous examples suggest that neutron real time diffraction experiments would particularly profit from an increased flux. Furthermore, a "single line" instrument to be used only for kinetic studies of the formation or destruction of a single phase would not be useful. As the expected average thermal flux at the SNQ ( $\Phi$ ) is about  $7 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$  in comparison with the thermal flux at the I.L.L. of  $\sim 10^{15} \text{ n cm}^{-2} \text{ s}^{-1}$ <sup>9,14</sup>, a gain for an experiment on irreversible processes with  $\tau > t_p$  (p. 2, "Methodology") by using a monochromator/PSD instrument would not be expected (however, see p. 12 for reversible processes). Progress may only be expected from a time-of-flight instrument which uses a broader wavelength band. The use of such an instrument poses in many cases no problem as for kinetic, mechanistic, and part of the structural studies the raw data can be used directly without correction.

An ideal time-of-flight instrument should have the following features:

- (i) Optimization for flux at the sample which favours not only real time diffraction experiments but also experiments on surfaces and small samples.
- (ii)  $0.3 \text{ \AA}^{-1} < Q < 15 \text{ \AA}^{-1}$  in order to overlap with the range of a small-angle-scattering instrument and to make the instrument also of interest

- for classical powder-diffractometry which does not demand an exceptional resolution and liquid scattering.
- (iii) Possibility to study preferred orientation effects and textures by an appropriate detector geometry, e.g. rings of detector segments as proposed for the SNS or a spherical PSD.
  - (iv) A resolution  $\Delta d/d$  of at least 1 %.
  - (v) A data collection system adequate for real time diffractometry including specifically a mass-storage device with rapid access for storing and accumulating spectra; possibility to use the proton pulse for triggering repetitive perturbations, and an on-line data reduction plus data analysis independent of a central computer.

#### IV.2 Instrumentation

The optimization for intensity demands that the sample should be placed as close as possible to the source, the distance sample/detector should be  $\sim 1$  m and every pulse should be used. As the shielding terminates at  $\sim 7$  m from the source<sup>14</sup>, the sample could be placed at 8 m and the detector at 9 m which would allow to use detectors in backscattering geometry. Fig. 6 shows the corresponding space time diagram assuming a pulse frequency of

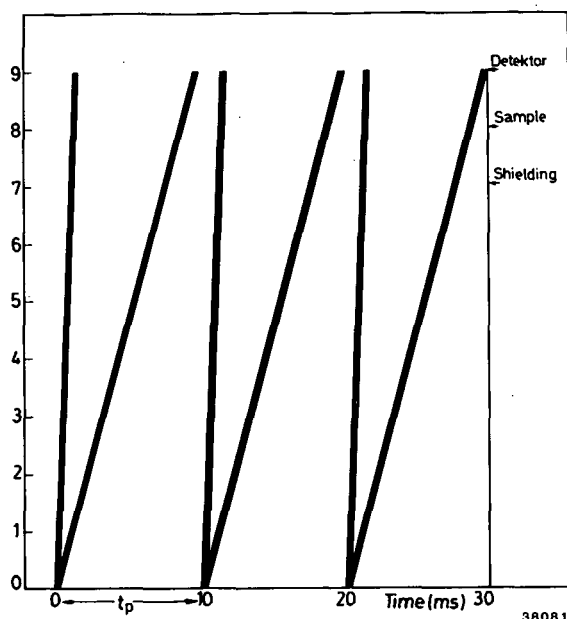


Fig. 6

Space-time diagram. Only neutrons with wavelengths  $0.7 \text{ \AA} < \lambda < 4.4 \text{ \AA}$  are registered by the detector. The ordinate corresponds to the flight path (m).

100 Hz and a temporal width of the neutron pulse of  $350 \mu\text{s}$ <sup>15</sup>. Assuming that the background level is reached 1.5 ms after the proton pulse, the utilizable wavelength band at 9 m from the source is  $0.7 \text{ \AA} < \lambda < 4.4 \text{ \AA}$  which implies the use of thermalized neutrons. Note that considerable intensity will be available even at  $\lambda = 5 \text{ \AA}$  although the peak intensity is about a factor 50 less at  $5 \text{ \AA}$  than at  $1.5 \text{ \AA}$ <sup>14</sup>. The intensity is weighted, however, by a factor  $\lambda^4$ .<sup>16</sup> In order to reach a Q-value of  $\sim 0.3 \text{ \AA}^{-1}$  the first detector has to be placed at  $2\theta = 15^\circ$ . Assuming an upper  $2\theta$ -value of  $150^\circ$ , the Q-range for the two limiting wavelengths is shown in Fig. 7.

In order to cut a wavelength band out of the spectrum a chopper has to be placed as close as possible to the source, i.e. into the shielding at  $\sim 2 \text{ m}$  from the source. This would result in a wavelength band  $\Delta\lambda$  of  $\sim 0.8 \text{ \AA}$ . In order to use the whole wavelength band between  $0.7 \text{ \AA}$  and  $4.4 \text{ \AA}$  the chopper would have to run at a different frequency as the source pulse which implies that the whole range is swept within 5 consecutive pulses. This has also been proposed for a high resolution TOF powder diffractometer at the SNQ<sup>16</sup>.

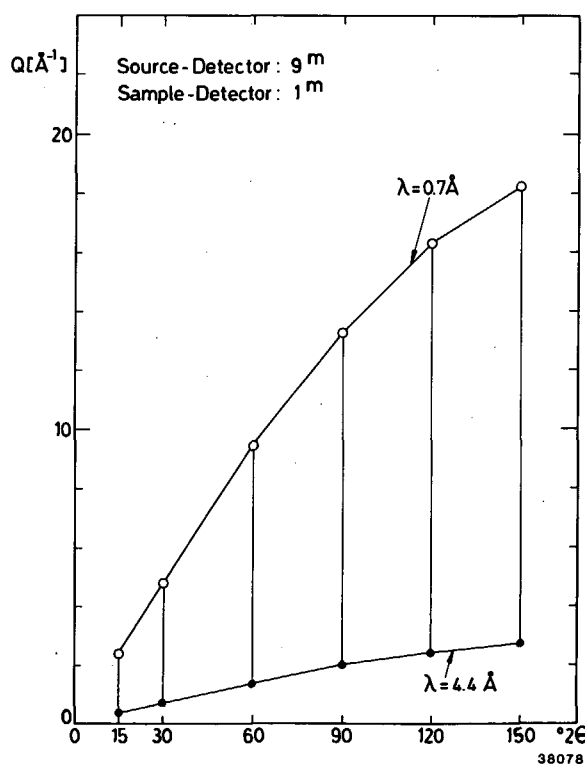


Fig. 7

Q-range covered by detectors between  $2\theta = 15^\circ$  and  $2\theta = 150^\circ$  for the wavelength-band  $0.7 \text{ \AA} < \lambda < 4.4 \text{ \AA}$ . Note that the whole Q-range could be covered by detectors at  $2\theta = 15^\circ$  and  $2\theta = 150^\circ$ .

Consider now the resolution which can be calculated approximately according to

$$\frac{\Delta\lambda}{\lambda} = \frac{t_w}{t} = t_w / (L/v) \quad (4)$$

where  $t$  is the flight time to the detector,  $L$  the flight path, and  $v$  the neutron velocity. Note that  $\Delta d/d \sim \Delta\lambda/\lambda$ .

In the case of pulses with a temporal width ( $t_w$ ) of 350  $\mu$ s, the resolution decreases rapidly with the decreasing wavelength (Fig. 8a). The best attainable resolution is still about a factor 3 worse than the corresponding resolution of the DIB-PSD<sup>9</sup>. Furthermore, in order to obtain this resolution, the wavelength band must be small which implies that there is no particular advantage as compared to a monochromator/PSD instrument, at least for irreversible transformations (see below).

In order to increase the resolution  $t_w$  has to be reduced by the chopper close to the source. Thus Fig. 8b shows the resolution for  $t_w \sim 10 \mu$ s. Effectively the whole wavelength band could be used while retaining a  $\Delta\lambda/\lambda < 1\%$ . Unfortunately this would have to be paid with a loss of intensity as only  $\sim 3\%$  of the neutrons are used. In this respect the construction of a compressor ring (IKOR) together with a pulsed source (GINA)<sup>14</sup> would be of considerable interest.

Consider finally the collimations  $\alpha_1$  and  $\alpha_2$ . Based on a distance source/sample of 8 m and a source dimension of 10 cm  $\alpha_1$  is calculated to be 42'. For smaller  $\Theta$ -values  $\alpha_1$  has to be matched with  $\alpha_2$  which is given by  $R/\cot\Theta$  where  $R$  is the  $\Delta d/d$  resolution<sup>18</sup>. Thus for  $2\Theta = 15^\circ$  and  $R = 0.01$   $\alpha_2$  is calculated to be  $\sim 5'$  which is the lower limit on collimation but still feasible. For a multiwire detector it can be written<sup>20</sup>:

$$\alpha_2 = \frac{3/4 d + p}{2 L_2} \quad (5)$$

where  $p$  is the distance of two wires,  $d$  the diameter of a circular sample and  $L_2$  the distance sample/detector. In this case, small sample diameters ( $< 5$  mm) and wire-wire distances of 1-2 mm have to be used to get  $\alpha_2$ -values close to 5'. Note that the incoming beam has to be collimated in order to reduce  $\alpha_1$ . The actual collimation conditions will depend, however, on the

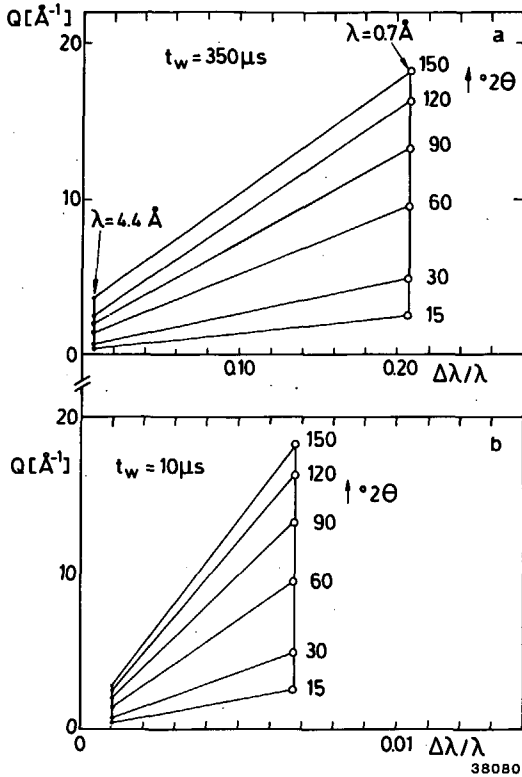


Fig. 8a

Calculation of the  $\Delta\lambda/\lambda$  resolution for a pulsed source with  $t_w = 350 \mu s$  and for various  $2\theta$ -values. The resolution has been calculated for the wavelengths 0.7 Å and 4.4 Å. The solid lines are guide-lines to the eye. This figure shows that in order to reach  $\Delta\lambda/\lambda \sim 0.03$  a small wavelength band has to be used. This implies a considerable restriction on the Q-range covered (Fig. 7).

Fig. 8b

Same as in Fig. 8a but for  $t_w = 10 \mu s$ . This figure shows that the whole Q-range available for detectors placed between  $2\theta = 15^\circ$  and  $2\theta = 150^\circ$  could be used (Fig. 7) while maintaining  $\Delta\lambda/\lambda < 0.01$ .

case considered. For larger  $2\theta$ -values, several wires can be grouped together as  $\alpha_2$  increases<sup>18</sup>.

A detailed calculation of the gain expected has not been performed as it depends on the type of experiment performed, i.e. resolution demanded, wavelength band,  $t_w$ . A general comparison of a reactor based monochromator-instrument with a pulsed-source time-of-flight instrument favours, however, the latter<sup>19</sup>.

The advantage will be demonstrated for the extreme case of a reversible phase transformation where  $t_s = t_w$ , i.e.  $\tau < t_p$  (p. 2, "Methodology"). In this case, the peak flux is exploited. Assuming a peak flux of  $1.2 \times 10^{16} \text{ ncm}^{-2} \text{ s}^{-1}$  a gain of a factor  $\sim 12$  can be obtained over a steady state instrument at the I.L.L.. The necessity to cut a small  $\Delta\lambda$ -band due to the small  $t_s$ -value will restrict in this case the Q-range per detector which suggests to use a PSD covering the range  $15^\circ < 2\theta < 150^\circ$  instead of individual detectors. For  $t_s > t_w$  a further increase in gain could be obtained by using a broader  $\Delta\lambda$ -band.



### Acknowledgement

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Single Crystal Diffractometry at the SNQ  
and at a Steady State Reactor

W. Jauch and H. Dachs

Hahn-Meitner-Institut Berlin

Introduction

In the following study a detailed comparison is made between the performance of single crystal diffractometry at the projected Spallation Neutron Source SNQ and at a steady state <sup>high flux</sup> reactor ( $\phi_0 = 1.2 \cdot 10^{15} \text{ ns}^{-1} \text{ cm}^{-2}$ ). The comparison is based on the total counting time necessary to obtain a sufficiently accurate data set for a meaningful determination of particular structural parameters.

It is supposed that the measurements are made with an area position-sensitive detector. At the reactor the diffraction experiment is performed with monochromatic radiation and the crystal is rotated around the vertical axis. At the SNQ a polychromatic pulsed beam impinges on the sample and the Laue method is used. Separation of the Laue spots into the individual component orders is achieved with a time-of-flight analysis. With a position-sensitive area detector a three-dimensional portion of reciprocal space is sampled during each pulse (Fig. 1).

In order to prevent overlap between neighbouring reflections a certain resolution in reciprocal space is required. The important problem-dependent parameters which determine the necessary resolution are the unit cell lengths and the minimum d-spacing at which information is required. With an instrument at a reactor resolution is achieved by suitably chosen beam divergence and wavelength spread. For TOF diffraction at a pulsed source the pulse length has to be cut down by a chopper under certain circumstances.

Our approach in the study is as follows:

- (i) the objective of the diffraction experiment is specified in

terms of the variance of the crystal-structural property of interest, (e.g. Fourier synthesis of scattering density, least-squares parameter refinement);

- (ii) experimental requirements and conditions such as luminosity, resolution, background and absorption are considered;
- (iii) a priori formulae for the counting time are derived from statistical considerations.

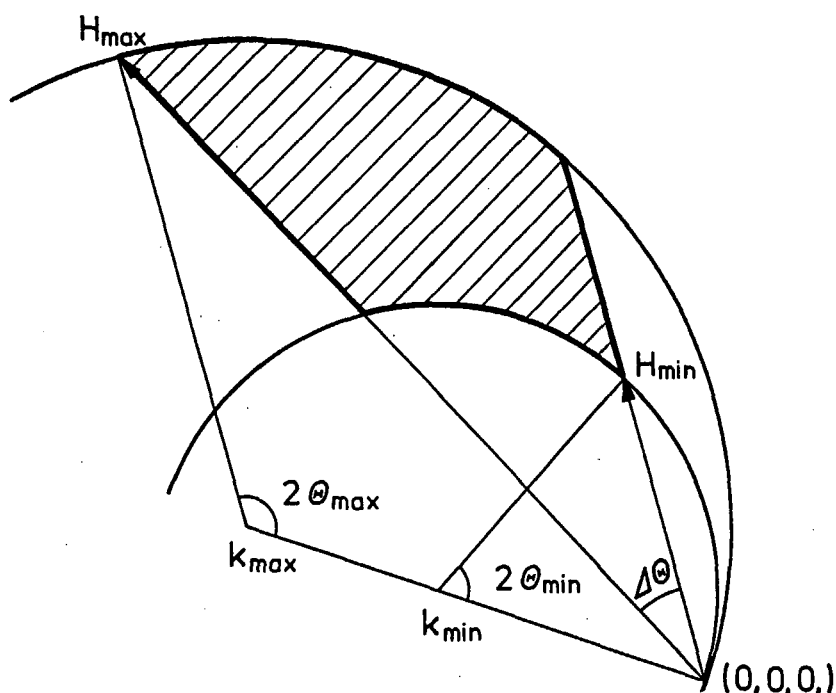


Fig. 1

We will treat two structural problems which at present lie on the border of feasibility:

- (i) protein crystallographic investigations at very high resolution ( $d_{\min} = 1.2 \text{ \AA}$ ) in order to localize H atoms;
- (ii) determination of positional parameters from small unit cells with very high precision ( $\sigma(x) = 5 \cdot 10^{-4}$ ).

Regarding (ii) there is the difficulty that it is too early to draw any conclusions about the accuracy limit attainable with area detectors. We have, therefore, considered the two possibilities of either collecting relatively few data of very high precision or measuring very many

reflections of relaxed precision.

We should emphasize here that the potential gain factors of the SNQ are not simply determined by the ratio of peak flux to time averaged flux since a number of conditions and requirements are different for the two techniques. The use of a position-sensitive detector implies that the instrumental resolution has to be matched to the most unfavourable case from the point of view of peak separation. In addition it is often not possible to use the optimum wavelength and the collimation cannot be relaxed beyond certain limits dictated by physical dimensions and distances. For these reasons the comparison of the SNQ with a reactor is rather complicated.

We arrive at the conclusion that in complex structural work the SNQ provides a substantial gain compared with a reactor.

## The Time-of-Flight Method

### Resolution

The resolution characteristics of a TOF diffractometer are given in /4/. For negligible sample dimensions the second moments of the resolution function depend in the following way on the time and angular uncertainties:

$$\sigma^2(H_x) = H^2 \left( \frac{\sigma_t^2}{T^2} + \cot^2 \theta \frac{\sigma^2(\gamma_i) + \sigma^2(\gamma_f)}{4} \right)$$

$$\sigma^2(H_y) = H^2 \left( \frac{\sigma^2(\gamma_i) + \sigma^2(\gamma_f)}{4} + \eta_s^2 \right)$$

$$\sigma^2(H_z) = H^2 \left( \frac{\sigma^2(\delta_i) + \sigma^2(\delta_f)}{4 \sin^2 \theta} + \eta_s^2 \right)$$

with  $H = 2 \sin \theta / \lambda =$  reciprocal lattice vector;  $\gamma_i$ ,  $\gamma_f$  and  $\delta_i$ ,  $\delta_f$  are the angular deviations in the scattering plane and vertical plane, respectively;  $\eta_s$  is the mosaic width (standard deviation) of the sample crystal.  $\sigma_t^2$  is the time dispersion of the neutron puls,  $T$  is the nominal time of flight.

We postulate that the integrated intensity is contained within  $\pm 2.5 \sigma$  of the profile. Then for the reflections of a cubic lattice with unit cell length  $a$  to be separated at least the following conditions must be satisfied:

$$\sigma(H_i) < \frac{1}{5a}, \quad (i = x, y, z).$$

The critical region for overlap corresponds to the largest value of  $H$  where the dominant contribution to  $\sigma(H_x)$  is the time uncertainty. From  $H_{\max} \sigma_t / T < 1/5a$  one obtains a condition for the maximum tolerable puls length:

$$\Delta t_{\text{puls}} (\text{FWHM}) < 238.1 \frac{\mu\text{s}}{\text{\AA m}} L \frac{d_{\text{min}}^2}{a} \sin\theta$$

with  $d_{\text{min}} (= H_0^{-1})$  = minimum interplanar distance for the data set,  $L$  = flight path. The conditions for angular separation impose limits on the tolerable sample mosaicity.

### Integrated Intensities

The integrated counts of a diffracted order of a Laue spot are given by

$$I = \phi(\lambda) \frac{V_s}{V^2} |F_H|^2 \frac{\lambda^4}{2\sin^2\theta} A(\lambda) t$$

where  $\phi(\lambda)$  = incident (time-average) differential flux at the sample,  $V_s$  = volume of the sample,  $V$  = unit cell volume,  $t$  = counting time,  $A(\lambda)$  = transmission factor. Fig. 2 shows the effective flux  $\phi(\lambda)\lambda^4$  for the SNQ parameters. In biological structures reflections are detectable at relatively small values of  $H$  only, and one can use wavelengths around the maximum of the effective flux.

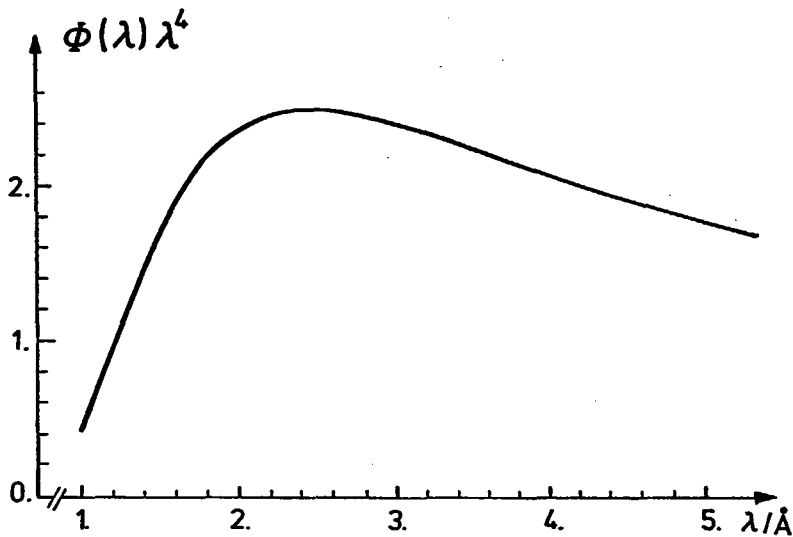


Fig. 2

### Background

Incoherent scattering from hydrogen is the principal source of background in biological structure work. The expected incoherent background counts under a Bragg reflection are:

$$I_{inc} = \phi(\lambda) \Delta\lambda \frac{V_s}{V} \sum \sigma_{inc} / 4\pi \Delta\Omega_C A(\lambda) t$$

where  $\Delta\lambda = 5\sigma_\lambda$  = wavelength interval contributing to a reflection,  
 $\Delta\Omega_C$  = angular dimension of a reflection at the detector.

$\Delta\lambda\Delta\Omega_C$  defines the reflection's volume which may be approximated by an ellipsoid

$$\Delta\lambda\Delta\Omega_C = \frac{4}{3} \pi \left(\frac{5}{2}\right)^3 \sigma_\lambda \sigma_\gamma \sigma_\delta \approx 5\sigma_\lambda \cdot 4\pi \sigma_\gamma \sigma_\delta.$$

with  $\sigma_\gamma^2, \sigma_\delta^2$  = angular dispersions of the diffracted beam.

From

$$\sigma_\gamma = 2\sigma(H_Y)/H, \quad \sigma_\delta = 2\sin\theta \sigma(H_Z)/H$$

and the resolution conditions follows

$$\Delta\lambda\Delta\Omega_C \leq H \lambda^2 \frac{d_{min}^3}{V}$$

### Variance of the Structure Factor Amplitude $|F_H|$

The counting statistical variance of the integrated intensity is the sum of the gross intensity (peak and background) and the variance of the background scaled to peak volume (detector pixels and time channels). We will assume that the background is determined from many more elements than the peak. Hence

$$\sigma^2(I) = I + I_{inc}$$

With

$$I = ct|F_H|^2$$

and

$$\sigma^2(|F_H|) = \frac{\sigma^2(|F_H|)}{4|F_H|^2}$$

the variance in the structure amplitude is obtained as

$$\sigma^2(|F_H|) = \frac{1}{4ct} \left(1 + \frac{I_{inc}}{I}\right).$$

From the conditions of peak separation it follows that

$$\frac{I_{inc}}{I} < \frac{1}{2} \frac{H^3}{H_0^3} \frac{\sum \sigma_{inc}/4\pi}{|F_H|^2}$$

#### Number of Simultaneous Reflections

An area position-sensitive detector permits a three-dimensional sampling of reciprocal space. For a detector centered at the Bragg angle  $\bar{\theta}$  and covering the solid angle  $\Delta(2\theta) = 2\Delta\delta$  the number of reflections measured simultaneously is given by

$$n = \frac{2}{3} V(\lambda_{min}^{-3} - \lambda_{max}^{-3}) (3\sin\bar{\theta} \sin\Delta\theta/2 - \frac{1}{3} \sin 3\bar{\theta} \sin 3\Delta\theta/2) \\ \times \left( \frac{3}{2} (-\Delta\delta + \sin 2\Delta\delta) + \frac{1}{8} \sin 4\Delta\delta \right)$$

#### Accuracy Factor and Counting Time

The precision of a data set may be assessed by the statistic

$$\epsilon^2 = \frac{\sum_{H_0} \sigma^2(|F_H|)}{\sum_{H_0} |F_H|^2} \quad (1)$$

$\epsilon$  is the lower limit of the crystallographic R-factor and is closely related to the variance of a Fourier synthesis. The local average of  $|F_H|^2$  is



$$\langle |F_H|^2 \rangle = T_H^2 \sum_{j=1}^N b_j^2$$

with  $T_H^2 = \exp(-B/2 H^2)$ .  $N$  is the number of atoms per primitive cell which participate in crystalline order. Hence, the sum over  $|F_H|^2$  can be obtained from

$$\sum_{H=0}^{H_0} |F_H|^2 = M \langle T_H^2 \rangle_{H_0} \sum_{j=1}^N b_j^2 \quad (2)$$

with  $M = 4/3\pi H_0^3 V$  = total number of reflections.  $\langle T_H^2 \rangle_{H_0}$  denotes the overall value of  $T_H^2$  for data with  $0 < H < H_0$  which can be evaluated from the spherical average

$$\langle T_H^2 \rangle = 3H_0^{-3} \int_0^{H_0} T_H^2 H^2 dH$$

The sum over  $\sigma^2(|F_H|)$  can be evaluated in a similar manner yielding

$$\sum_{H=0}^{H_0} \sigma^2(|F_H|) = \frac{MV^2 \langle \sin^2 \theta \rangle}{2t \phi(\lambda) \lambda^4 V_S A(\lambda)} \left[ 1 + \frac{3}{4} \frac{\sum \sigma_{inc}/4\pi}{\sum b_j^2} f\left(\frac{B}{2} H_0^2\right) \right] \quad (3)$$

with

$$f(x) = x^{-3} [(x^2 - 2x + 2)e^x - 2]$$

$\langle \sin^2 \theta \rangle$  is the average over the accessible domain in reciprocal space and depends on the position and dimensions of the detector. (3) implies that all reflections are counted for the same time  $t$  (area detector). With (1), (2), (3) one gets an a priori formula which can be used to predict the required counting time necessary to obtain a data set of a given statistical quality  $\epsilon$ :

$$t = \frac{V^2 \langle \sin^2 \theta \rangle}{\epsilon^2 \langle T_H^2 \rangle \sum b_j^2 \cdot 2\phi(\lambda) \lambda^4 V_S A(\lambda)} \left[ 1 + \frac{3}{4} \frac{\sum \sigma_{inc}/4\pi}{\sum b_j^2} f\left(\frac{B}{2} H_0^2\right) \right] \quad (4)$$

With an area detector  $t$  is the counting time per crystal setting. The total counting time depends on the number of settings that provide a coverage of a hemisphere in reciprocal space (or less, depending on crystal symmetry).

## Biological Macromolecules at High Resolution

We will consider the case that neutron diffraction is used to complete the structural information obtained by X-ray diffraction. With *single a few* exceptions high-resolution ( $d_{\min} \leq 2 \text{ \AA}$ ) X-ray studies of proteins and other biological macromolecules determine the coordinates of C, N, O and heavier atoms. The principal aim of the neutron experiment is the localization of the hydrogen atoms. Knowledge of the exact location of specific hydrogens is crucial for the understanding of many biological problems.

The quantity of interest is the Fourier synthesis of the scattering density

$$\rho(r) = V^{-1} \sum_{H_0} |F_H| \cos(2\pi H \cdot r - \phi_H)$$

where the amplitudes  $|F_H|$  are determined from the neutron experiment and initial estimates of the phases  $\phi_H$  are calculated using the X-ray atomic coordinates of the nonhydrogen atoms. The spatial resolution in the Fourier map is characterized by the smallest interplanar distance  $d_{\min}$ .

The contribution of the experimental errors to the mean variance of the Fourier map is related to the statistic  $\epsilon$

$$\sigma^2(\rho) = V^{-2} \sum_{H_0} \sigma^2(|F_H|) = V^{-2} \epsilon^2 \sum_{H_0} |F_H|^2$$

In addition, there is the intrinsic contribution due to the use of approximate phases. A priori estimates of both the expected peak heights and the error level can be derived from probability considerations /2/.

The following table shows results for a neutron protein map (fractional partial structure contribution to the scattering power  $\sigma_1^2 = 0.7$ ,  $B = 10 \text{ \AA}^2$ ,  $d_{\min} = 1.2 \text{ \AA}$ ):

| Mean positional error<br>of the phasing model<br>$\langle  \Delta r  \rangle$ (Å) | Average peak height<br>of hydrogen<br>$ \langle \rho(r) \rangle $ (f/Å <sup>3</sup> ) | Phase error<br>contribution<br>$\sigma_p(\rho)$ (f/Å <sup>3</sup> ) |
|-----------------------------------------------------------------------------------|---------------------------------------------------------------------------------------|---------------------------------------------------------------------|
| 0                                                                                 | 1.3                                                                                   | 0.34                                                                |
| 0.2                                                                               | 1.15                                                                                  | 0.49                                                                |

For  $\epsilon = 0.1$  the contribution of the amplitudes to the standard deviation is  $\sigma_A(\rho) = 0.13 \text{ f/Å}^3$ . Hence,  $\epsilon = 0.1$  will suffice to complete the structure. It is to be emphasized that we define  $\epsilon$  as an overall value taken over all observed reflections. When it is applied to a group of reflections in a narrow region of  $\sin\theta/\lambda$  it will show a progressive increase as a function of  $\sin\theta/\lambda$ .

#### Example

**S t r u c t u r a l p r o b l e m:** protein with cubic lattice constant  $a = 50 \text{ Å}$  at  $1.2 \text{ Å}$  resolution ( $d_{\min}$ ).

**I n s t r u m e n t:** flight path  $L = 8 \text{ m} \rightarrow \Delta t_{\text{puls}} < 55 \mu\text{s} \cdot \sin\theta_c$  ( $\sin\theta_c = \lambda_{\min}/2d_{\min}$ ). Hence, the thermal neutron pulses of the SNQ ( $300\mu\text{s}$ ) have to be shortened by a chopper near the moderator. A Fermi-chopper designed for a beam width of  $5 \cdot 10 \text{ cm}^2$  is described in the SNQ report. The minimum feasible distance moderator-chopper is assumed to be  $1.5 \text{ m}$  /7/. The transmitted bandwidth is then restricted to  $\Delta\lambda = h/m \cdot 300\mu\text{s}/1.5\text{m} = 0.8 \text{ Å}$ .

**W a v e l e n g t h s:**  $\lambda = 2 - 2.8 \text{ Å}$  (maximum effective flux)  
 $\rightarrow \Delta t_{\text{puls}} < 46 \mu\text{s}$ .

**D e t e c t o r:** area position sensitive detector covering  $\Delta\theta = 30^\circ$ ,  $\Delta\delta = 30^\circ$ . 100% efficiency will be assumed.

**N u m b e r o f s i m u l t a n e o u s r e f l e c t i o n s:**  
Two detector positions have to be used (Fig. 3).

| $\theta$   | $n$    | $d_{\min}/\text{Å}$ | $d_{\max}/\text{Å}$ | $\langle \sin^2\theta \rangle$ |                                                               |
|------------|--------|---------------------|---------------------|--------------------------------|---------------------------------------------------------------|
| $60^\circ$ | 6670*  | 1.2                 | 2.0                 | 0.771                          | * 7833 reflections are excited                                |
| $25^\circ$ | 1170** | 1.55                | 8.1                 | 0.287                          | ** 1011 reflections with<br>$2 \text{ Å} < d < 8.1 \text{ Å}$ |

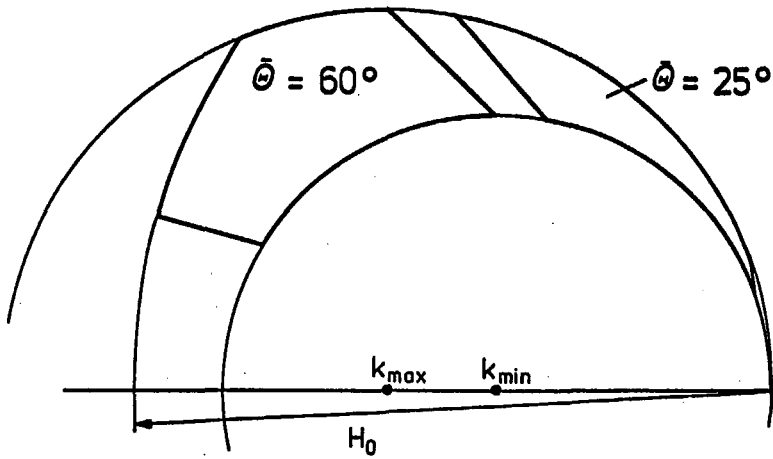


Fig. 3

Number of crystal settings:

| $d/\text{\AA}$ | Number<br>of reflections | Number of<br>crystal settings (M/n) |                             |
|----------------|--------------------------|-------------------------------------|-----------------------------|
| $> 1.2$        | $3.03 \cdot 10^5$        |                                     |                             |
| $1.2 - 2$      | $2.38 \cdot 10^5$        | 36                                  | $(\bar{\theta} = 60^\circ)$ |
| $2 - 8.1$      | $6.5 \cdot 10^4$         | 64                                  | $(\bar{\theta} = 25^\circ)$ |
| $> 8.1$        | 985                      |                                     |                             |

Effective flux  $\phi(\lambda)\lambda^4$  at the sample: Moderator area =  $10 \cdot 10 \text{ cm}^2$ , slit package area =  $5 \cdot 10 \text{ cm}^2$ ,  $l(\text{chopper-moderator}) = 1.5 \text{ m}$ ,  $l(\text{chopper-sample}) = 7.5 \text{ m}$ , solid angle (moderator-sample)  $\Omega_i = 10\text{cm}/900\text{cm} \cdot 5\text{cm}/750\text{cm} = 7.4 \cdot 10^{-5}$ . For  $\lambda = 2 - 2.8 \text{ \AA}$  the effective flux is practically constant. With  $\phi(\lambda) = 2\phi_0\lambda_T^4/\lambda^5 \cdot \exp(-\lambda_T^2/\lambda^2)$ ,  $\phi_0 = 1.2 \cdot 10^{15} \text{ ns}^{-1}\text{cm}^{-2}$ ,  $T = 322 \text{ K}$  ( $\lambda_T = 1.717 \text{ \AA}$ )  $/7/$ :  $\phi(\lambda)\lambda^4 = 4.1 \cdot 10^{14} \text{ s}^{-1}\text{cm}^{-2}\text{sterad}^{-1}\text{\AA}^3 \times 7.4 \cdot 10^{-5} \times 46\mu\text{s}/300\mu\text{s} \times 0.9 = 4.2 \cdot 10^9 \text{ s}^{-1}\text{cm}^{-2}\text{\AA}^3$  (chopper transmission = 0.9).

Crystal parameters:  $B = 10 \text{ \AA}^2$ ,  $\langle T_H^2 \rangle_{H_0} = 0.19$ ,

$V = 1.25 \cdot 10^5 \text{ \AA}^3$ ,  $\sum b_j^2 = 3400 \text{ barn}$ ,  $\sum \sigma_{\text{inc}}/4\pi = 17800 \text{ barn}$  (50% H/D-exchange,  $\sigma_{\text{inc}}(\text{H}) = 50 \text{ barn}$ ),  $V_S = 1 \text{ mm}^3$ ,  $A = 0.78$  ( $\mu = 2.5 \text{ cm}^{-1}$ ).  $\eta_S(\text{FWHM}) \leq 40'$ . For the atomic composition of the partially deuterated unit cell see /6/.

**Optimum division of counting time:** Since the two detector settings cover sections of reciprocal space with high and low d-spacings, respectively, the counting time should be varied between the two settings. One finds that with

$$t_l/t_h = (\Gamma_l/\Gamma_h)^{1/2}$$

$$\Gamma_l = M_l \langle \sin^2 \theta \rangle_l \left[ 1 + \frac{3}{4} \frac{\sum \sigma_{\text{inc}}/4\pi}{\sum b_j^2} f\left(\frac{B}{2} H_l^2\right) \right]$$

$$\Gamma_h = M_h \langle \sin^2 \theta \rangle_h \left[ 1 + \frac{3}{4} \frac{\sum \sigma_{\text{inc}}/4\pi}{\sum b_j^2} f\left(\frac{B}{2} H_h^2\right) \frac{M}{M_h} \right]$$

(l: low  $\theta$ , h: high  $\theta$ ) the prescribed precision  $\epsilon^2$  is achieved with minimum amount of counting time. To a good approximation  $t_h$  is given by (4).

**Total counting time:** With  $\epsilon = 0.1$  and the above parameters:

$$t_l = 16 \text{ min}, t_h = 115 \text{ min}.$$

Hence, the total counting time for data collection of a hemisphere in reciprocal space is

$$T = 1/2(36 \times 115 \text{ min} + 64 \times 16 \text{ min}) = 43 \text{ h}$$

#### Comparison with the Conventional Technique at a Steady State Reactor

A conventional diffractometer equipped with an area position-sensitive detector is considered (Fig. 4). Data are collected with crystal rotation perpendicular to the beam ( $\omega$ -scan). Integration of the Bragg peaks is performed over the rotation angle ( $\Delta\omega$ ) and the detector pixels.

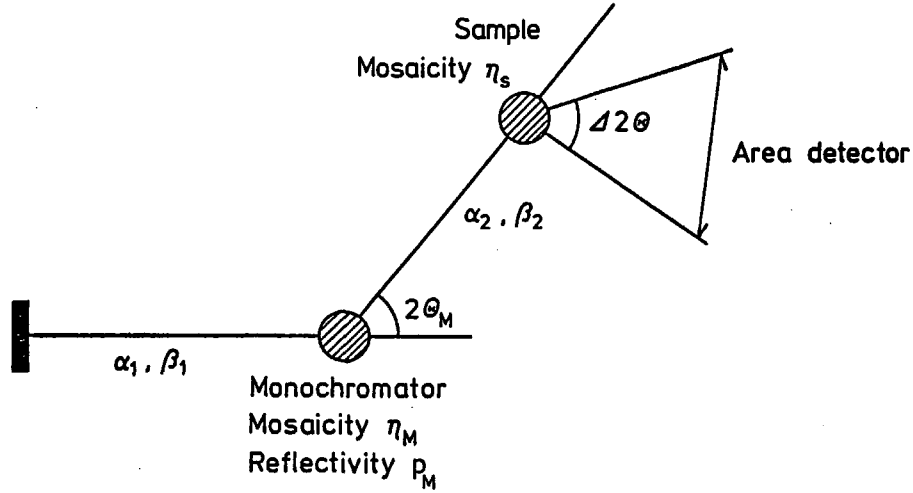


Fig. 4

### Luminosity and Resolution

If the horizontal and vertical collimations are described by Gaussians of standard deviation  $\alpha$  resp.  $\beta$  ( $\alpha_1 = \alpha_2 = \alpha$ ,  $\beta_1 = \beta_2 = \beta$ ) the neutron flux at the sample is calculated as

$$\phi = \left(\frac{\pi}{2}\right)^{1/2} \frac{\alpha^2}{(2\alpha^2 + 4\eta_M^2)^{1/2}} \frac{\beta^2}{(2\beta^2 + 4\sin^2\theta_M \eta_M^2)^{1/2}} p_M \eta_M \lambda \cot\theta_M \phi(\lambda)$$

with  $\phi(\lambda) = 2\phi_0 \lambda_T^4 \lambda^{-5} \exp(-\lambda_T^2/\lambda^2)$ .

With  $\alpha_1 = \alpha_2$  the wavelength spread is  $\sigma_\lambda/\lambda = \cot\theta_M \alpha/2^{1/2}$ . In the focusing region ( $\theta \approx \theta_M$ ) the width in the rotation angle is  $\sigma_\omega \approx \eta_s$  for large sample mosaicity (proteins) and the angular width of the diffracted beam in the horizontal direction is  $\sigma_\gamma \approx \alpha/8$ . The width in vertical direction depends essentially on the vertical divergence:  $\sigma_\delta \approx \beta$ . The focusing region will comprise the bulk of reflections if  $\theta_M$  is equal to the Bragg angle corresponding to the smallest d-spacing to be measured

$$\cot\theta_M = [(2d_{\min}/\lambda)^2 - 1]^{1/2}$$

The criterion for no-overlap of neighbouring reflections leads to the following conditions:

$$\alpha(\text{FWHM}) \text{ and } \beta(\text{FWHM}) \text{ and } \eta_s(\text{FWHM}) < \lambda/2a \quad (5)$$

$$\Delta\lambda/\lambda(\text{FWHM}) < d_{\min}/2a$$

The condition for the wavelength spread implies

$$\alpha(\text{FWHM}) < \tan\theta_M \frac{d_{\min}}{2^{1/2}a}$$

which, in general, imposes a more severe restriction for the horizontal divergence than (5). For reasons of simplicity we have considered equatorial reflections only.

#### Integrated Intensity

$$I = \phi \frac{V_s}{V^2} \lambda^3 L |F_H|^2 A/\dot{\omega} \quad (6)$$

( $L$  = Lorentz-factor,  $\dot{\omega}$  = angular scanning speed =  $\Delta\omega/t$ ,  $\Delta\omega = 5\sigma_\omega$ ).

#### Incoherent Background

$$I_{\text{inc}} = \phi \frac{V_s}{V} \sum \sigma_{\text{inc}}/4\pi \Delta\Omega_c A t \quad (7)$$

#### Variance of the Structure Factor

With  $\dot{\omega} = 2\pi/\tau$  it follows

$$\sigma^2(|F_H|) = \frac{\pi V^2 L^{-1}}{2\tau \phi \lambda^3 V_s A} \left(1 + \frac{I_{\text{inc}}}{I}\right)$$

$\tau$  denotes the time for a  $2\pi$ -rotation of the sample. The Lorentz-factor for the equatorial plane will be used:  $L = 1/\sin 2\theta$ . From (6), (7) and the conditions of peak separation it follows that

$$\frac{I_{inc}}{I} = \frac{VL^{-1} \Delta\omega \Delta\Omega_c \sum \sigma_{inc}/4\pi}{\lambda^3 |F_H|^2} \sim \frac{1}{2} L^{-1} \frac{\sum \sigma_{inc}/4\pi}{|F_H|^2}$$

### Precision of the Data Set

$$\sum \sigma^2(|F_H|) = \frac{\pi MV^2}{2\tau \phi \lambda^3 V_S A} [\langle L^{-1} \rangle + \frac{1}{2} \langle (LT_H)^{-2} \rangle \frac{\sum \sigma_{inc}/4\pi}{\sum b_j^2}]$$

$\langle L^{-1} \rangle$  and  $\langle (LT_H)^{-2} \rangle$  are average values over reciprocal space /3/.

### Counting Time

$$\tau = \frac{\pi V^2 [\langle L^{-1} \rangle + 1/2 \langle (LT_H)^{-2} \rangle (\sum \sigma_{inc}/4\pi / \sum b_j^2)]}{2\varepsilon^2 \phi \lambda^3 V_S A \sum b_j^2 \langle T_H^2 \rangle}$$

### Example

**Wavelength:** A detector covering the same solid angle ( $\Delta\theta = 30^\circ$ ,  $\Delta\delta = 30^\circ$ ) as in the previous example is assumed. In order to collect the data from a hemisphere with a single detector setting the wavelength is restricted to  $\lambda = 1.4 \text{ \AA}$  ( $\theta = 35.7^\circ$  for  $d_{min} = 1.2 \text{ \AA}$ ,  $\theta_{min} = 5.7^\circ$  corresponding to  $d_{max} = 7 \text{ \AA}$ ). 11% of the reflections lie in the blind region.

**Neutron flux at the sample:**

$$\alpha(\text{FWHM}) < 0.7^\circ, \beta(\text{FWHM}) < 0.8^\circ$$

$$\phi(\lambda) = 8.62 \cdot 10^{14} \text{ s}^{-1} \text{ cm}^{-2} \text{ \AA}^{-1} \quad (\phi_0 = 1.2 \cdot 10^{15} \text{ ns}^{-1} \text{ cm}^{-2}, \lambda_T = 1.717 \text{ \AA})$$

$$\eta_M(\text{FWHM}) = 20', p_m = 0.5 \rightarrow \phi = 3.1 \cdot 10^7 \text{ ns}^{-1} \text{ cm}^{-2}.$$

**Counting time:** For the same crystal parameters as in the previous example one obtains with  $\langle L^{-1} \rangle = 0.77$  and  $\langle (LT_H)^{-2} \rangle = 8.76$ :

$$\tau = 16 \text{ d}$$

→ gain factor SNQ versus reactor: 9.



### Instrument with Neutron Guide Tube

The resolution requirements for  $\sigma(H_X)$  are fulfilled without the need of shortening the pulses if the flight path is  $L \approx 50$  m. For such distances one has to use neutron guide tubes. For  $Ni^{58}$  the limiting angle of total reflection is  $\theta_c = 0.12^\circ/\text{\AA} \cdot \lambda$ . For a rectangular guide the acceptance solid angle is  $(2\theta_c)^2$ . With  $\lambda_{\min} = 2 \text{ \AA}$ :  $\Delta\Omega_i = 7 \cdot 10^{-5}$ . From the use of a neutron guide tube results a gain of

$$\frac{7 \cdot 10^{-5} \times 0.6}{46\mu\text{s}/300\mu\text{s} \times 0.9 \times 7.4 \cdot 10^{-5}} = 4.1$$

compared to the chopper instrument. A reflectivity loss of 1%/m has been assumed (pessimistic estimate).

Hence, a TOF-diffractometer at a guide tube would be a very attractive instrument for structure investigation of biological macromolecules.

### Conclusions (biological macromolecules)

- (1) With the SNQ it is possible to resort to samples considerably smaller than  $1 \text{ mm}^3$ . Crystals with edge lengths of  $\sim 0.5 \text{ mm}$  can be grown for many proteins. At present, the need for large crystals dictates the choice of a particular system to be studied.
- (2) The SNQ would allow high resolution studies of nondeuterated crystals. This would facilitate the interpretation of the Fourier maps considerably since there would be no uncertainty in the degree of H/D-replacement.
- (3) At present only few proteins have been studied with X-rays at such a very high resolution we considered in our example ( $d_{\min} = 1.2 \text{ \AA}$ ). Very high resolution, however, is of particular importance in the neutron case since the observation-to-parameter ratio has a value which is only about a half of the corresponding X-ray value. With the SNQ, data could be collected up to higher  $d$ -values than it is presently possible because of the large amount of beam time needed. It is to be noted that lowering the resolution  $d_{\min}$  will increase the gain factor of the SNQ as compared to a steady

state source.

- (4) It is not only the relative gain factor which is important but also the absolute magnitude of the expected counting time. It implies that with the SNQ the number of proteins to be studied per year could drastically increase. Neutron diffraction could become a common routine method to complete the structure of biological macromolecules obtained from X-ray diffraction. In view of the clear success of neutron protein crystallography during the last years /9/ interest in the applications of neutron diffraction to protein structure will certainly continue to increase.

#### Accurate Atomic Coordinates from Small Unit Cells

The position of the  $j$ th atom in the unit cell is  $r_j = (x_j, y_j, z_j)$ . The variance of a coordinate as determined from least-squares refinement with isotropic data is approximately given by

$$\sigma^2(x_j) \approx \left\{ \frac{1}{s} \sum_h \sigma^{-2}(|F_H|) (\partial |F_H| / \partial x_j)^2 \right\}^{-1}$$

( $s$  = symmetry factor).

Usually the estimates of  $\sigma(|F_H|)$  are based not merely on counting statistics but one has to include random experimental errors which affect the net intensity in proportion to its magnitude

$$\sigma^2(I) = P^2 I^2 + \sigma^2(|F_H|) = P^2 |F_H|^2 / 4$$

For neutron single counter diffractometers  $P$  is typically found to be in the range of 0.01-0.04. The need to take wavelength-dependent factors into account (incident effective spectrum, absorption, extinction, detector efficiency) exposes TOF-diffraction data to sources of error which are absent with the monochromatic technique.

Aiming at accurate structural parameters the best strategy of data collection depends on the characteristic value of  $P$ . If counting statistics is the only detectable error source high parameter precision can be achieved from few selected reflections of high precision. When

the errors independent of the amount of counting time are large measurement of very many reflections will compensate the low precision of the data set. In addition, the use of many reflections also allows to reduce the effect of systematic experimental errors.

For non-centrosymmetric space groups one obtains the following expression (see also /5/)

$$\sigma^{-2}(x_j) = \frac{2}{3} \pi^2 b_j^2 \frac{M}{s} \langle \sigma^{-2}(|F_H|) T_H^2 H^2 \rangle$$

If the counting statistical contribution to  $\sigma^2(|F_H|)$  is negligible

$$\sigma^{-2}(x_j) = 66.2 \frac{b_j^2 V H_0^5}{s P^2 \sum b_j^2}$$

The precision factor  $\epsilon$  is then given by  $\epsilon = P/2$ . It is now possible to determine the reciprocal radius  $H_0$  which ensures a certain standard deviation  $\sigma(x_j)$  for a given value of  $P$

$$H_0 = 0.43 \left( \frac{s P^2 \sum b_j^2}{b_j^2 V \sigma^2(x_j)} \right)^{1/5} \quad (8)$$

### Example

As an example we consider the crystal structure of  $\text{Li}_2\text{SO}_4 \cdot \text{H}_2\text{O}$  where recently an accurate neutron diffraction study has been performed in order to determine small atomic displacements as a function of temperature /1/.

Crystal parameters:  $\sum b_j^2 = 4.26 \text{ barn}$ ,  $V = 207 \text{ \AA}^3$ , largest lattice constant:  $8 \text{ \AA}$ ,  $B = 2 \text{ \AA}^2$ ,  $V_S = 4 \text{ mm}^3$ ,  $A = 0.76$  ( $\lambda = 1 \text{ \AA}$ ),  $s = 4$ .

Desired parameter precision:  $\sigma(x, \text{Li}) = 5 \cdot 10^{-4} \text{ \AA}$ .

### SNQ

Suppose  $P = 0.06$ . From (8) follows then:

$$d_{\min} = 0.3 \text{ \AA} \rightarrow \Delta t_{\text{puls}}(\text{FWHM}) \lesssim 20 \text{ \mu s} \quad (L = 8 \text{ m}, a = 8 \text{ \AA}).$$

The accuracy factor  $\epsilon^2 = (0.03)^2 + \epsilon_{CS}^2$  will not be deteriorated very much if the counting statistical contribution is  $\epsilon_{CS} = 0.02$ .

Effective flux  $\phi(\lambda)\lambda^4$  at the sample:

$$\phi(\lambda)\lambda^4 = 5.8 \cdot 10^{12} \text{ s}^{-1} \text{ cm}^{-2} \text{ \AA}^3 \text{ sterad}^{-1} \times 7.4 \cdot 10^{-5} \times (20\mu\text{s}/300\mu\text{s}) \times 0.9 \\ = 2.66 \cdot 10^7 \text{ s}^{-1} \text{ cm}^{-2} \text{ \AA}^3 \text{ (calculated with } \lambda = 0.7 \text{ \AA)}.$$

Wavelengths:  $\lambda = 0.6 - 1.4 \text{ \AA}$  (+ L(chopper-moderator) = 1.5 m)

Number of simultaneous reflections ( $\Delta\theta = \Delta\delta = 30^\circ$ ):

| $\theta$   | n   | $d_{\min}/\text{\AA}$ | $d_{\max}/\text{\AA}$ | $\langle \sin^2\theta \rangle$ |
|------------|-----|-----------------------|-----------------------|--------------------------------|
| $70^\circ$ | 871 | 0.3                   | 0.85                  | 0.88                           |

Number of crystal settings:

| $d/\text{\AA}$ | Number of reflections |     |
|----------------|-----------------------|-----|
|                | M                     | M/n |
| $> 0.3$        | 32,113                |     |
| $0.3 - 0.85$   | 30,701                | 36  |
| $> 0.85$       | 1,412                 |     |

Counting time per crystal setting (background counts assumed to be negligible):

$$t = \frac{V^2 \langle \sin^2\theta \rangle}{2\epsilon^2 \phi(\lambda)\lambda^4 V_S A \langle T_H^2 \rangle \sum b_j^2}$$

With  $\langle T_H^2 \rangle_{\Delta H} = 0.016$  ( $\Delta H = 1/0.3\text{\AA} - 1/0.85\text{\AA}$ ):  $t = 2.5 \text{ h}$

Total counting time for the complete shell in reciprocal space:

$$T = 36 \times 2.5\text{h} \approx 4 \text{ d}$$

### Steady State Source

- 1) A value of P smaller than in the TOF-case will be assumed (no wavelength-dependent uncertainties):  $P = 0.03 \rightarrow d_{\min} = 0.4 \text{ \AA}$  and  $\epsilon_{CS} = 0.01$ .

Wavelength  $\lambda = 0.6 \text{ \AA}$  at a hot source:  $\phi = 2 \cdot 10^7 \text{ ns}^{-1} \text{ cm}^{-2}$   
(expected flux at D9B).

Area detector:  $\Delta\theta = \Delta\delta = 30^\circ \rightarrow \theta_{\max} = 49^\circ, \theta_{\min} = 19^\circ$   
( $d_{\max} = 0.92 \text{ \AA}$ )

Number of reflections  $M(0.4 \text{ \AA} < d < 0.92 \text{ \AA}) = 10161 - 867$

$$\langle T_H^2 \rangle_{\Delta H} = 0.046 \quad (\Delta H = 1/0.4 \text{ \AA} - 1/0.92 \text{ \AA})$$

Counting time:

$$\tau = \frac{\pi V^2 \langle L^{-1} \rangle}{2\epsilon^2 \phi \lambda^3 V_S A \langle T_H^2 \rangle \sum b_j^2} = 3 \text{ d}$$

Total counting time for the complete shell:  $T = 6 \text{ d}$

2) Assume now the same value of  $P$  as in the TOF case:  $P = 0.06$ .

$\rightarrow d_{\min} = 0.3 \text{ \AA}$  and  $\epsilon_{CS} = 0.02$ .

A wavelength of  $\lambda = 0.4 \text{ \AA}$  is chosen in order to collect the data with one detector setting ( $\theta_{\max} = 42^\circ, \theta_{\min} = 12^\circ$  ( $d_{\max} = 0.96 \text{ \AA}$ )).  
 $\phi = 1 \cdot 10^7 \text{ ns}^{-1} \text{ cm}^2$  (expected flux at D9B),  $\langle T_H^2 \rangle_{\Delta H} = 0.02$ :

$$\tau = 12 \text{ d}, T = 24 \text{ d}$$

### Comparison based on Counting Statistics only

The following comparison is based on the required experiment time for collecting (almost) identical data sets of equal counting statistical precision. We refer to a data limit of  $d_{\min} = 0.5 \text{ \AA} / 1/$ .

SNQ:  $\lambda = 1 - 1.8 \text{ \AA}$

$$\Delta t_{\text{puls}} < 40 \mu\text{s} \quad (d_{\min} = 0.5 \text{ \AA}, a < 12 \text{ \AA}, L = 8 \text{ m})$$

$$\phi(\lambda)\lambda^4 = 1.3 \cdot 10^{14} \text{ s}^{-1} \text{ cm}^{-2} \text{ sterad}^{-1} \text{ \AA}^3 \times 7.4 \cdot 10^{-5} \times 40/300 \times$$

$$0.9 = 1.2 \cdot 10^9 \text{ s}^{-1} \text{ cm}^{-2} \text{ \AA}^3 \quad (\text{calculated with } \lambda = 1.1 \text{ \AA})$$

| $\bar{\theta}$ | $\Delta\theta$ | $n$ | $d_{\min}/\text{\AA}$ | $d_{\max}/\text{\AA}$ |
|----------------|----------------|-----|-----------------------|-----------------------|
| 70°            | 30°            | 169 | 0.5                   | 1.1                   |

$M(0.5 \text{ \AA} < d < 1.1 \text{ \AA}) = 6069 \rightarrow 36 \text{ crystal settings}$

$\langle T_H^2 \rangle_{\Delta H} = 0.12, \epsilon = 0.01: t = 100 \text{ s}$

Total counting time  $T = 36 \times 100\text{s} = 1 \text{ h}$

Steady State Source:  $\lambda = 0.6 \text{ \AA}$  ( $\theta_{\max} = 37^\circ, \theta_{\min} = 7^\circ$

( $d_{\max} = 1.2 \text{ \AA}$ ))

$\phi = 2 \cdot 10^7 \text{ ns}^{-1} \text{ cm}^{-2}$  (hot source)

$\epsilon = 0.1: \tau = 22 \text{ h}, T = 44 \text{ h}$

Of course,  $\lambda = 1 \text{ \AA}$  would be more favourable with respect to flux and reflectivity. With a detector of limited solid angle ( $\Delta\theta = \Delta\delta = 30^\circ$ ) it would necessitate, however, quite a number of different detector settings.

### Conclusion (precise structural parameters)

In biological structural work rather inaccurate structure factor amplitudes are tolerable and the final accuracy is determined primarily by counting statistics and not by other error sources. The situation is quite different when precise parameters of simple structures need to be obtained. From the very limited studies to date, it seems that the final R-factors resulting from structure refinements with TOF data are high in comparison to the common standard. The potential gain of the SNQ will depend on the magnitude of the non-counting statistical errors that can be achieved. To derive accurate structure factor amplitudes a number of wavelength-dependent factors must be known very precisely. A loss in accuracy due to these wavelength dependencies may offset any gain in intensity. Otherwise the gain factors of the SNQ are considerable.

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A diffractometer for  
low Q diffraction from large unit cells

M.S. LEHMANN  
Institut Laue-Langevin  
Avenue des Martyrs  
Grenoble

Abstract

A diffractometer for low resolution, single-crystal structure analysis of very large biological systems is discussed. It is shown that an instrument with a flight path of 30 m would be suitable for the limiting case of  $10 \text{ \AA}$  resolution.

A comparison with a similar instrument on a steady state reactor gives gain factors for the SNQ instrument of 6 assuming no frame overlap, a pulse rate of 100 Hz and a pulse length of 0.6 mS. Decreasing the pulse rate to 25 Hz, assuming the pulse length to be 1.2 mS, and including frame overlap gives a total gain factor of 26.



## Introduction

Determination at low resolution of biological crystalline structures with very large unit cells using neutron diffraction methods has already been shown to be feasible<sup>1</sup>. The basis of the method is the use of contrast variation<sup>2</sup>, where the scattering density of the solvent - and to a certain extent the scattering density of the macromolecules - are varied by varying the H/D content of the components involved. The advantage of the method is that in principle the molecular arrangement is only slightly influenced by the H/D exchange, while the changes in scattering densities are very considerable.

Using deuteration of part of a macromolecular agglomerate the intermolecular vectors can be determined by triangulation methods, and this kind of work is in progress using small angle scattering from solutions<sup>3</sup>. Similar techniques could be used where single crystals are available. It would equally be possible to use a small, deuterated part of the macromolecule or a deuterated substrate for structure solution, employing "heavy atom" techniques as used in X-ray diffraction<sup>4</sup>.

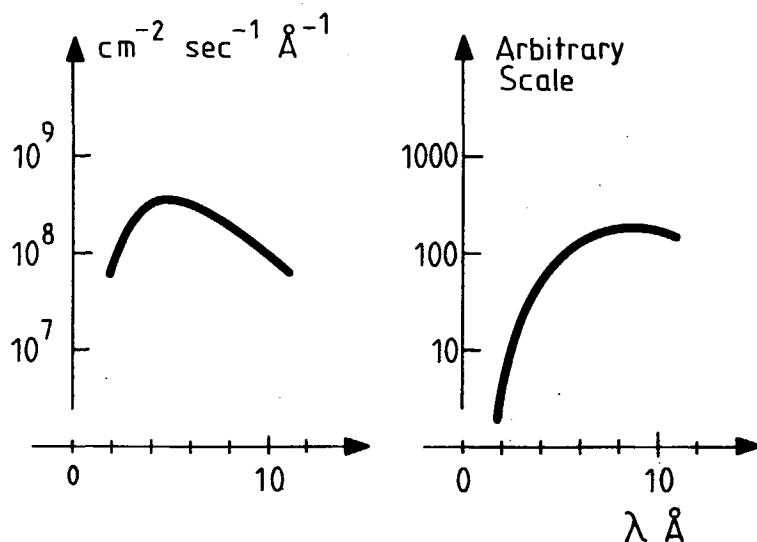
The neutron techniques outlined do not for very large cells have any parallels in other fields of crystallography, and they might therefore be helpful in untangling complex biological structures. Unfortunately single crystals, when available, are small and the neutron flux is relatively low. It is therefore necessary to use the highest flux source, and to seek to optimise the experimental conditions.

The present note discusses resolution conditions and possible geometries for a special diffractometer for this purpose, and compares

this to a similar diffractometer at a high flux steady-state reactor<sup>5</sup>.

### General considerations

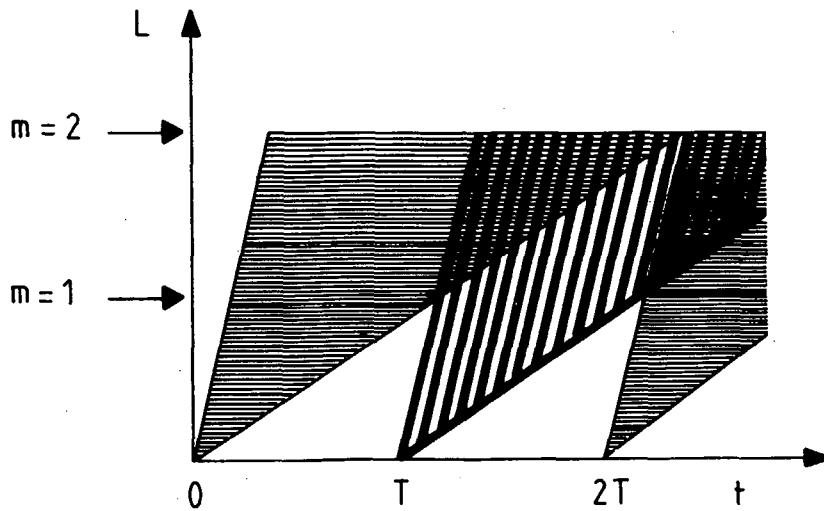
The unit cells in question will have axis length ( $a$ ) from  $300 \text{ \AA}$  and upwards. As area detectors will be used, this sets instantly a requirement on the wavelength  $\lambda$ . For perfect collimation the smallest angle between adjacent diffracted beams will be  $\lambda/a \cos \theta$ . If we require that the distance between spots on the detector is larger than 1 cm, and we take the sample-detector distance to be around 1 m, then the requirement on  $d$  is  $\lambda > a \cos \theta / 100 \approx 3 \text{ \AA}$ . A cold source is therefore needed. We shall assume the same source distribution as for the cold source of the Institut Laue-Langevin :



At left, the intensity as a function of  $\lambda$  for the cold source<sup>6</sup>. At right is given intensity times  $\lambda^2$ , which is proportional to counts in the detector.

### Length of instrument

For the pulsed source the length,  $L$  (in m), of the instrument is given from the time-length diagram



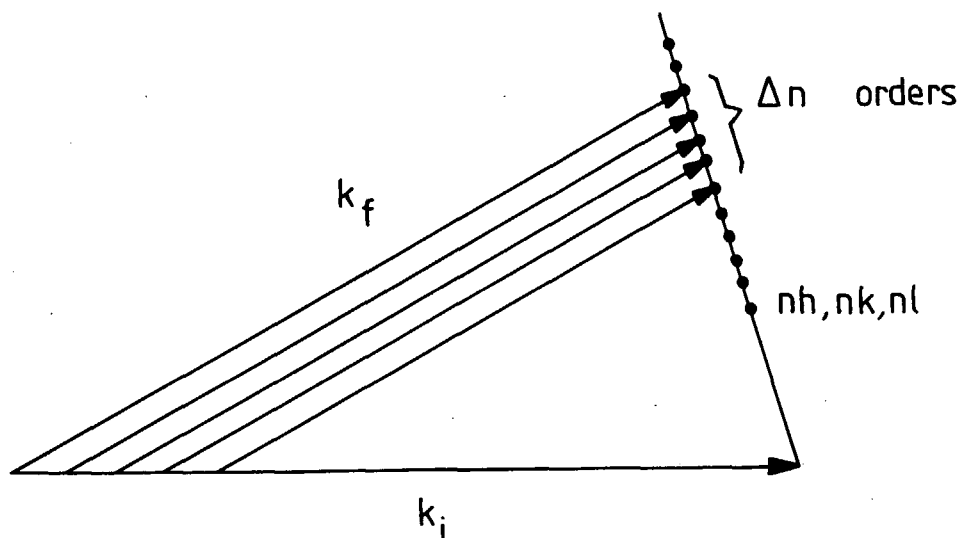
where  $T$  is time in seconds between pulses. We get for limiting wavelengths  $\lambda_1$  and  $\lambda_2$

$$L = m T 4000 / (\lambda_2 - \lambda_1) = m T 4000 / \Delta\lambda$$

If  $m$  is 1, there is no frame overlap, but the detector receives neutrons continuously. For  $m = 2$ , two frames overlap exactly, etc. The pulselength,  $\tau$ , at length  $L$  is  $mT$ .

#### Bragg reflection overlap along a lattice direction

From the scattering diagram



where  $k_i$  and  $k_f$  are incoming and scattered wavevectors, respectively, we can estimate the number of orders  $\Delta n$  of reflections with indices  $nh$ ,  $nk$ ,  $nl$  that can be illuminated for a given setting. From Braggs law ( $2a \sin\theta = n\lambda$  where  $a$  is the distance between planes  $\langle h, k, l \rangle$ ) we get

$$\Delta n = 2a \sin\theta \Delta\lambda/\lambda^2$$

For measurements at a resolution in real space of  $d$  ( $2d \sin\theta = \lambda$ ) we then obtain

$$\Delta n = a/d \cdot \Delta\lambda/\lambda$$

To resolve adjacent reflections in time we must require that there are  $R$  time-resolution elements,  $\Delta t$ , between each reflection, where  $\Delta t$  is the pulse length at  $t = 0$ . We then get

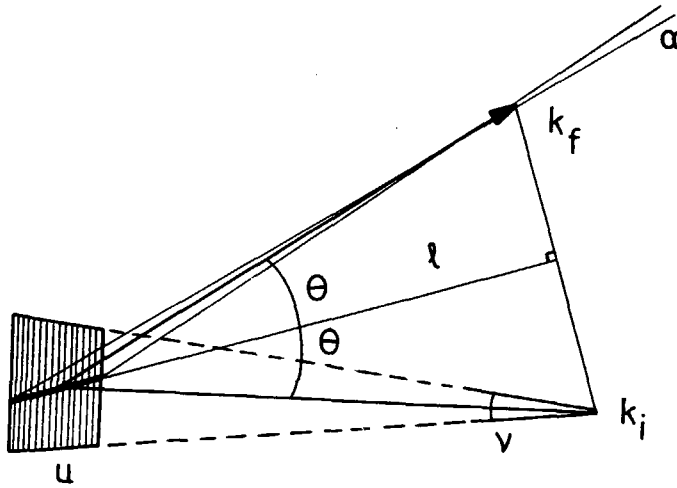
$$\Delta t = T/R\Delta n$$

and finally

$$\Delta\lambda/\lambda = T/\Delta t \cdot d/Ra$$

#### Bragg-spot size on detector

The linear spotsize on the detector can be estimated from the scattering diagram for a given wavevector  $|k| = |k_i| = |k_f| = 1/\lambda$



The length of the incoming wavevector distribution (shaded part) along  $k_i$  at a given time is defined by the time resolution and is

$$u = \Delta k \cdot \Delta t / T = kd / Ra$$

for  $m \geq 1$ . The width of the wavevector distribution is  $\nu \cdot k$ , where  $\nu$  is the beam divergence. The line  $l$  bisects the scattering angle, and its intersection with the distribution defines the allowed wavevectors  $k_f$ <sup>7</sup>. This is the thick part of  $l$ . The angle  $\alpha$  is then given by

$$\alpha = \tan \theta \cdot u / k = \tan \theta \cdot d / Ra$$

For large  $u$  or large  $\theta$  the limiting value for  $\alpha$  is  $\nu$ .

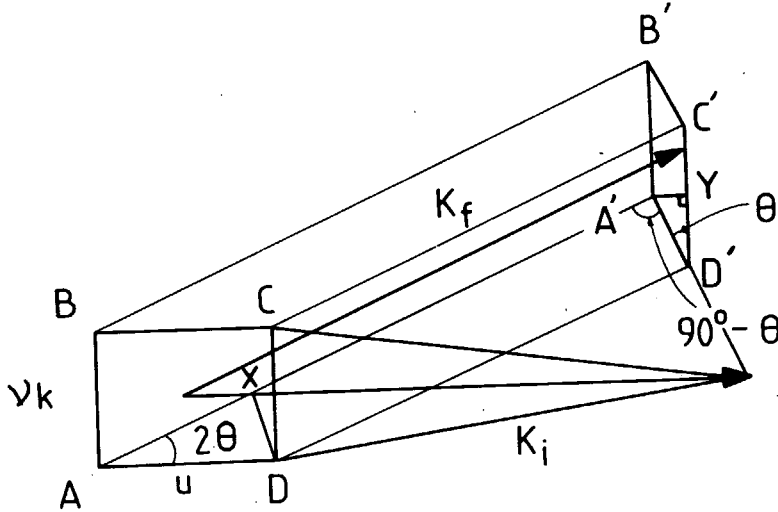
If we work in the range of  $\theta$  less than  $30^\circ$ , then  $\sin \theta \approx \tan \theta$  and we get

$$\alpha = \lambda / 2Ra$$

The linear spotsize is now  $\alpha$  multiplied by the distance from the sample to the detector.

### The resolution function

The condition for more reflections falling simultaneously on the same location of the detector - the overlap condition - is estimated from the following diagram.



Here we assume elastic scattering and parallel beams for  $k_f$ , i.e. all beams fall at the same detector location. Box ABCD is the wavevector distribution and as  $AA' = BB'$  and  $CC' = DD'$  the resolution element becomes a parallelogram. Taking XD parallel to  $A'D'$  we get  $XD = 2u \sin\theta = A'D'$ , and we then obtain for the longest direction

$$\gamma' = B'D' = ((vk + u \sin 2\theta)^2 + 4u^2 \sin^4 \theta)^{1/2}$$

For the width across the thinnest part, along the direction  $k_i$ , we have

$$\gamma'' = A'Y = 2u \sin^2 \theta$$

The important quantity is the ratio between distances between lattice points and the size of the resolution element, and we can define

$$r_{\gamma'} = 1/a\gamma' \quad \text{and} \quad r_{\gamma''} = 1/a\gamma''$$

For the out of plane direction the maximum size is  $\gamma''' = \nu k$ , assuming a spherical cross-section of the beam, and we get

$$r_{\gamma'''} = \lambda/\nu a$$

For  $r_{\gamma} > 1$  the Bragg spots do not overlap.

### Some numerical estimates and the diffractometer size

To get an idea of the size and geometry of the diffractometer we shall introduce a series of assumptions. The length of the whole instrument is assumed to be the longest obtainable without frame overlap, i.e.  $m = 1$ . We set the mean  $\lambda$  to  $10 \text{ \AA}$ , and we assume that the beam has a divergence of  $1^\circ$ , which is slightly less than the divergence of a beam coming from a neutron guide. We will assume, that the peaks are nearly overlapping along the most crowded direction, i.e. along the  $a$ -direction, and we choose  $R = 4$ . For the pulse height we use  $0.6 \text{ mS}$ , and the calculations are done for two possible pulse frequencies,  $100 \text{ Hertz}$  and  $25 \text{ Hertz}$ , and 3 resolutions expressed in  $d$ , the minimum spacing between planes. For  $25 \text{ Hertz}$  the pulse length is unknown, and we shall assume that it is twice the value given for  $100 \text{ Hertz}$ .

We then get :

| $d(\text{\AA})$                     | = | 15   | 10   | 5    | $\Delta t(\text{mS})$ |
|-------------------------------------|---|------|------|------|-----------------------|
| 100 Hz : $\Delta\lambda/\lambda$ =  |   | 0.21 | 0.14 | 0.07 | 0.6                   |
| 25 Hz : $\Delta\lambda/\lambda$ =   |   | 0.42 | 0.28 | 0.14 | 1.2 (assumed)         |
| 100 Hz : $L(\text{m})$ =            |   | 19   | 29   | 58   |                       |
| 25 Hz : $L(\text{m})$ =             |   | 10   | 14   | 29   |                       |
| max $2\theta(\text{deg})$ $\approx$ |   | 39   | 60   | 180  |                       |

For 5 Å resolution it is obviously necessary to reduce the wavelength a bit and we shall not further discuss this case. For the other two cases there is no serious resolution problem as  $r_{\gamma'} = 1.3$ ,  $r_{\gamma''} = 8$  and  $r_{\gamma'''} = 2$  for the worst case. For the direction approximately orthogonal to the outgoing beam we are at the limit of resolution, but overlap will here occur for reflections which lie next to each other on the detector surface and this will reduce the risk of non-detected overlap.

At the limiting resolution of 10 Å we therefore arrive at an instrument with a total length of around 30 m. For the sample-detector distance we can choose 1 m. This would give a maximum spot size on the detector surface of about 0.4 cm. As the source to sample distance is approximately equal to the total length,  $L$ , the sample will be continuously illuminated by neutrons, and we do therefore not require any very fast detector electronics. Altogether the requirements on the detector therefore corresponds to present day technology. The size will be defined by what is financially possible.

The beam distribution on the sample position could cause problems, as 10 Å neutrons fall about 6 cm over a distance of 30 m. The final distribution will be correlated with wavelength, and we must ensure by the construction of the guides that all the required wavelengths do fall on the sample.

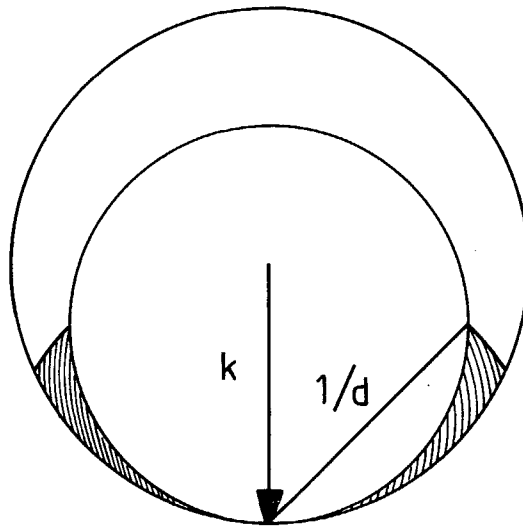


### Comparison with steady state instrument

An extensive analysis of the required resolution in  $\Delta\lambda/\lambda$  for steady state instruments has been carried out by Roth et al.<sup>8</sup> Here, we use the diagrams employed above, and for the steady state instrument we shall assume that the wavelength is fixed by a velocity selector. In this case the resolution functions for the two instruments are similar, and for the steady state diffractometer the length of the wavevector distribution,  $u$ , is now  $\Delta k$ . We keep the other values as  $\lambda = 10 \text{ \AA}$  and  $\nu = 1^\circ$ . We choose the size of  $\gamma''$  to be determining for the  $\Delta k/k$ , as this is along  $k_f$  and overlap is therefore not obvious for a given area detector recording, but only for a series of measurements. We shall choose  $r_{\gamma''} = 2$ , and we get  $\Delta k/k = 0.07$  for  $d = 15 \text{ \AA}$  and  $\Delta k/k = 0.03$  for  $d = 10 \text{ \AA}$ .  $r_{\gamma''}$  is the same as for the SNQ instrument, but  $r_{\gamma'}$  becomes 0.5 and 0.8 for  $d = 15$  and  $10 \text{ \AA}$ , respectively. As the first value is rather low, we shall finally choose  $\Delta k/k = 0.03$  for both  $d$  spacings.

For the comparison we shall only look at the case for  $d = 10 \text{ \AA}$ , as this corresponds to the instrument discussed above.

The data collection strategy, and the shape of the detector can strongly influence the time for data collection, and as no unified approach exists, we shall assume the style of data collection to be similar in the two cases. The time will then be proportional to the number of settings, and therefore inversely proportional to the volume of reciprocal space illuminated. This is shown in the figure



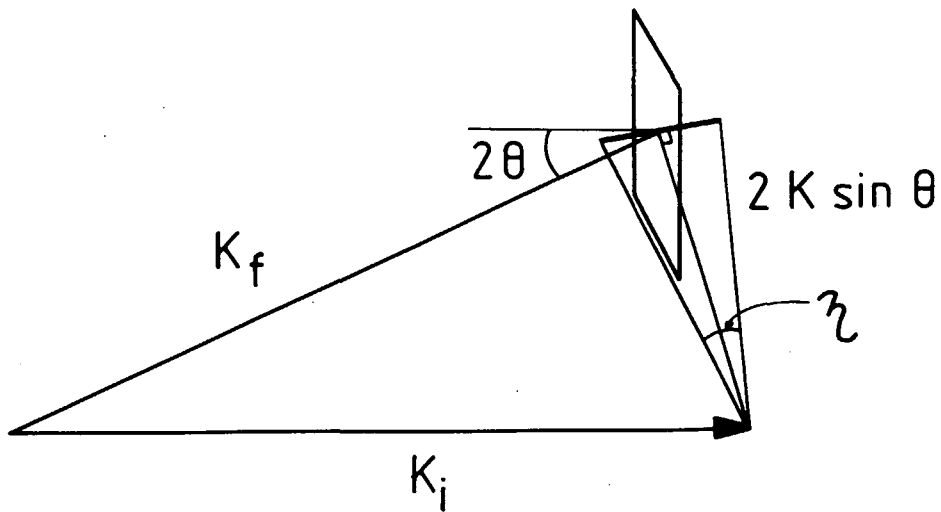
and is found to be  $\Delta V = \pi \Delta \lambda / 4d^4$ .

The detector efficiency can be assumed to be the same, but the neutron guide for the steady state instrument can be shorter than for the SNQ machine, giving the first a gain. This will be counter-balanced by the loss in the velocity selector of the steady state instrument. All together we then get, assuming the time average flux at the SNQ to be 1.4 times the flux at a high flux reactor, and comparing only  $\Delta \lambda$ , the following gain factors for an SNQ diffractometer

|                 |   |    |
|-----------------|---|----|
| $d(\text{\AA})$ | = | 10 |
| 100 Herz        | = | 6  |
| 25 Herz         | = | 13 |

#### Crystal mosaic spread

We have till now neglected the effects of crystal mosaic spread. This corresponds to the smearing of the lattice point in the directions orthogonal to the lattice vector, and is depicted in the figure



The projections of this distribution onto the directions of interest are, if we assume a mosaic with fullwidth  $\eta$  :

1. Along  $\gamma''$  :  $2k\eta \sin\theta \cos\theta = k\eta \sin 2\theta$
2. Orthogonal to  $\gamma''$ , in the plane :  $2k\eta \sin^2\theta$
3. Orthogonal to the scattering plane :  $2k\eta \sin\theta$

It is mainly 1 and 3 that are of interest. The limit to overlap is thus given by the condition that the sum of the size of the resolution in a given direction and the mosaic in the same direction should be less than  $1/a$ . For  $d = 10 \text{ \AA}$ , and the values employed above we get the following conditions on  $\eta$  :

|              | 1.          | 3.          |
|--------------|-------------|-------------|
| Steady state | $1.2^\circ$ | $0.9^\circ$ |
| SNQ          | $1.9^\circ$ | $0.9^\circ$ |

So the SNQ instrument can accept a larger mosaic along one direction than the steady state instrument. As however the problems seem mainly to occur in the "out-of-plane" direction on the detector, this does not

give any obvious advantage to the SNQ instrument. It is worth noting, though, that mosaic effects might become a serious obstacle to the employment of the maximum flux.

### Frame overlap

The estimates have been done assuming no frame overlap. For single crystal elastic measurement (i.e. we assume elastic scattering only) the scattering directions are both very well defined and limited. If we have two-frame overlap, for example, only two different wavelengths can contribute to a given observed scattering, and for each of these we can calculate the corresponding point in k-space. One of these must be a lattice point, and we can attribute a count to the corresponding Bragg-reflection if we know the crystal orientation. If both correspond to lattice points we must either rotate the crystal a bit, or find some algorithm for sharing the intensity. This goes beyond the scope of the paper. We can however estimate the probability of the incident, as this is the ratio between the volume of the resolution element and the volume of the reciprocal unit cell. We get for both d spacings

$$p = A'B'\gamma''\gamma'''.a^3 = 0.07$$

The probability of finding that a given neutron can originate from two different reflection processes is therefore limited, and the influence on the precision of the measurement is probably within the acceptable limit.

### Conclusion

A simple design study of an SNQ diffractometer for very large unit cell has shown this instrument to be feasible. The instrument would be around 30 m long, and would aim at studies of  $10^{\circ}$  resolution in crystals with  $300^{\circ}$  cell dimensions. A comparison with a steady state instrument indicates a gain factor of 6 assuming no frame overlap. With frame overlap, which is feasible, the gain can be doubled.

The analysis shows that for a 100 Hertz pulse rate with 0.6 mS pulse length the gain factor is limited. For a lower frequency pulse rate the gain factor increases. At 25 Hertz it will be around 13, if we assume the pulse to be 1.2 mS.

As the useful wavelength band  $\Delta\lambda/\lambda$ , which defines the flux and gain factor, is inversely proportional to  $\Delta t/T$ , a considerable gain could clearly as well be obtained by reducing the pulse length  $\Delta t$ .

Altogether it is therefore very likely that the proposed instrument would be of use in macromolecular crystallography.

### ACKNOWLEDGEMENTS

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## A PHOTOGRAPHIC LAUE CAMERA FOR PULSED NEUTRON BEAMS

W. Prandl, D. Hohlwein\*

Institut für Kristallographie der Universität Tübingen, Charlottenstr. 33,  
D-7400 Tübingen and \*guest at the Hahn-Meitner-Institut Berlin

### (1) Introduction

Single crystal diffraction work at pulsed neutron sources may be done, at least in principle, with a completely conventional diffractometer: use a monochromator in the beam line and forget about the time structure. An apparatus of this kind uses only the time averaged flux  $\bar{\phi}$ . We introduce a gain factor  $g = (\text{fraction of peak flux } \phi \text{ used})/\bar{\phi}$ . Then evidently we have  $g = 1$  for the conventional diffractometer. On the other hand single crystal time of flight (TOF) machines with two dimensional electronic counters having  $g > 1$  are operating at least at Argonne and Los Alamos.

We propose here as an alternative method a Laue camera with photographic registration. This device may be used at pulsed neutron sources as well as with existing TOF beam lines on steady state reactors. Due to the intrinsically small dimension of the camera and the large source-to-camera distance needed for reasonable resolution several such devices can be operated at a single beam hole.

The application of photographic films as two-dimensional neutron detectors has been discussed and demonstrated earlier [2c].

### (2) Layout of the instrument

The principle of operation is shown in fig. 1. A pulsed beam, tailored properly in time by one or more choppers, is Bragg reflected by the sample and the reflected neutrons are recorded on a rotating film after an overall flight path  $L$ . Let  $\Delta T$  be the time between consecutive neutron pulses.

We require that any lattice plane should generate just one spot on the film. Therefore the film must rotate with an angular velocity  $\dot{\psi} = 1/\Delta T$ . At the SNQ we have  $\Delta T = 10$  msec and  $\dot{\psi} = 6000$  r.p.m.. On the other hand there

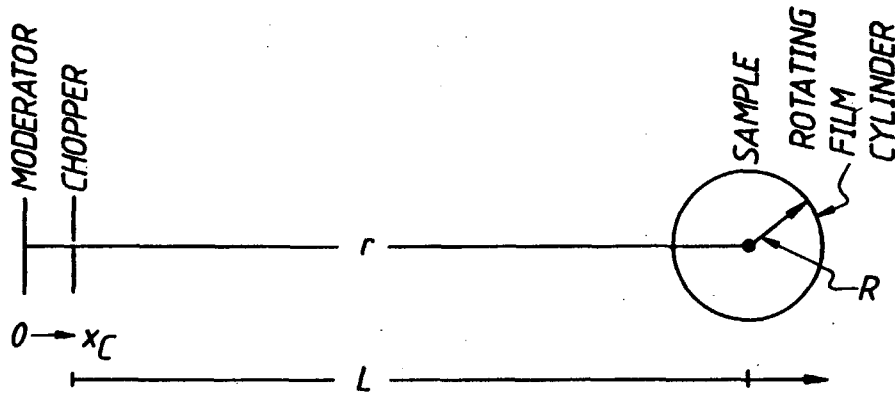


Fig. 1. Basic geometry of the instrument:  $x_C \cong 0.6$  m;  $L = 20$  m;  
 $6 \leq R \leq 12$  cm

should be no overlap of spots. This condition asks for a finite band width  $\Delta\lambda$ . If, finally, we require that all reflections excited within the band  $\lambda \leq \tilde{\lambda} \leq \lambda + \Delta\lambda$  should fill the film without a gap, we arrive at the usual relation between  $\Delta\lambda$  and  $L$ :

$$L = \frac{\Delta T}{\Delta\lambda} \cdot \frac{h}{m} = 3.956 \frac{\Delta T_{\text{msec}}}{\Delta\lambda_{\text{\AA}}} \text{ [m]} \quad (1)$$

So, for the SNQ we obtain  $L = 20$  m for a band width  $\Delta\lambda = 2\text{\AA}$ . We point out that there is no coupling between  $\Delta\lambda$  and  $\lambda$ : the wavelength  $\lambda$  is determined from the phase lag of the chopper or chopper system at  $x_C$  with respect to the SNQ fast neutron pulse. Actually an efficient chopper system for the photographic Laue camera must be designed under conditions which apply in the same way to diffractometers using electronic two-dimensional detectors. The moderator-chopper distance  $x_C$  for the camera parameters chosen is  $x_C = 0.6$  m (fig. 2). Such a

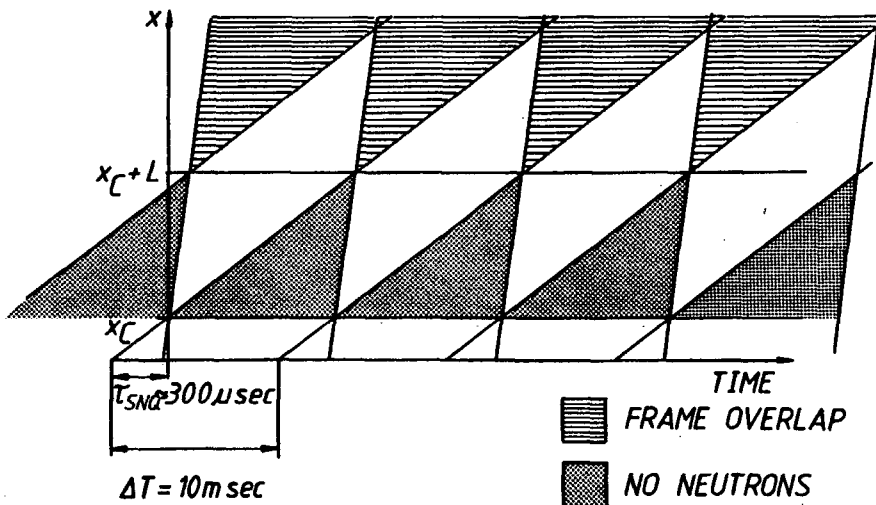


Fig. 2. Flight path vs. time diagram. The width of the thermal neutron pulse is assumed to be  $300 \mu\text{sec}$  ( $\tau_{\text{proton}} = 250 \mu\text{sec}$ )



short distance might be hard to realize. On the other hand choosing  $x_C = 1.2$  m we obtain  $\Delta\lambda = 1 \text{ \AA}$  and  $L = 40$  m. An instrument with these parameters would be better suited for larger unit cells. Data collection, however, must be done piecewise in radial shells: from 1 to 2  $\text{\AA}$ , from 2 to 3  $\text{\AA}$  etc., in this case.

We introduce a laboratory coordinate system as shown in fig. 3. The camera itself may either have only one "upper" cylinder (fig. 4a) or a split cylinder (fig. 4b) for letting the primary neutrons in and out. Spots on the film will be described by  $\alpha$  and  $\psi$  (fig. 4c).

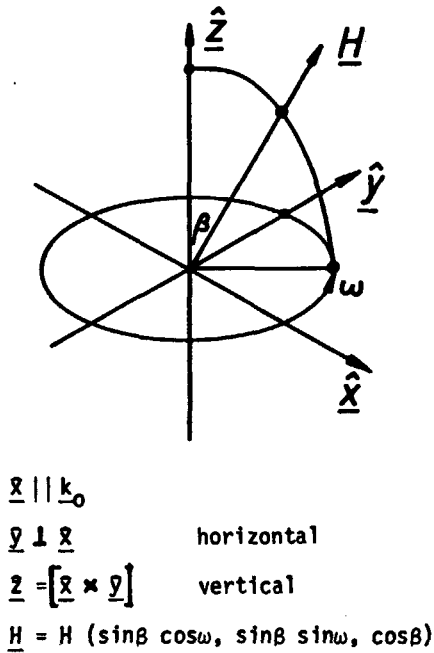


Fig. 3.  
Coordinate system used

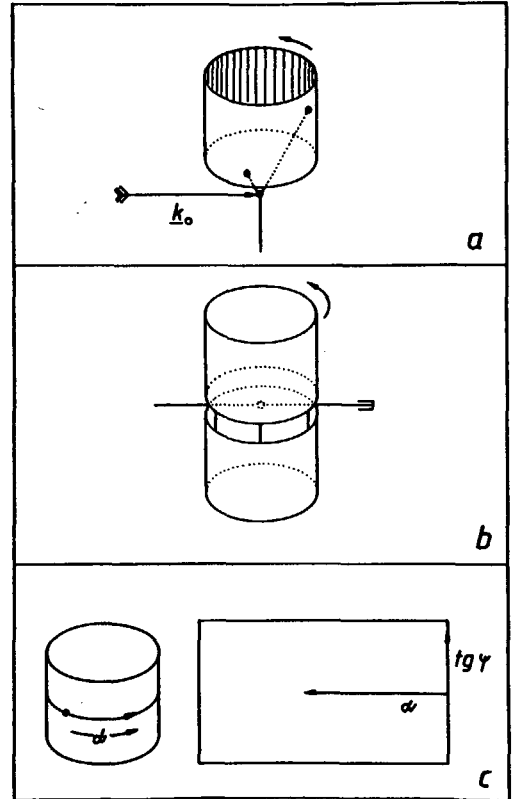


Fig. 4. Two different types of rotating film cylinders (a,b). Location of a spot on the film (c)

We define that the point  $\alpha = \psi = 0$  coincides with the positive  $\hat{x}$ -axis wherever the chopper is open. Then  $\alpha$  and  $\psi$  are given by

$$\alpha(H) = 2\omega(H) - \pi + K \frac{\sin\beta(H) \cos\omega(H)}{H} \quad (2)$$

$$\sin\psi(H) = -\sin 2\beta(H) \cdot \cos\omega(H) \quad (3)$$

$$K = 2 \frac{m}{h} L \dot{\psi} \left( = 363 \frac{\text{degrees}}{\text{\AA}} \text{ for } L = 20 \text{ m and } \dot{\psi} = 6000 \text{ rpm} \right).$$

### (3) Resolution

In the photographic Laue camera described so far the detector (film) is permanently open. We therefore have to discuss three different topics referring to resolution:

- (a) spot size
- (b) separation of spots belonging to neighbouring reflections
- (c) overall density of spots on the film.

The spot size is given by the TOF resolution functions calculated, e.g., by Stoica [1]: the width in arrival time  $\sigma_T$  has to be converted into an angular width  $\sigma_\alpha$ . In order to find out whether the SNQ thermal pulse width is adequate to a photographic camera we simplify the resolution calculation radically and consider only the angular width  $\Delta\alpha(\tau)$  due to either the thermal neutron pulse width of the source or else of the chopper. As a worst case study we investigate the radial and transverse resolution of reflections which are neighbours of an  $\underline{H} = (n00) \cdot \underline{a}^*$  reflection (fig. 5,6).

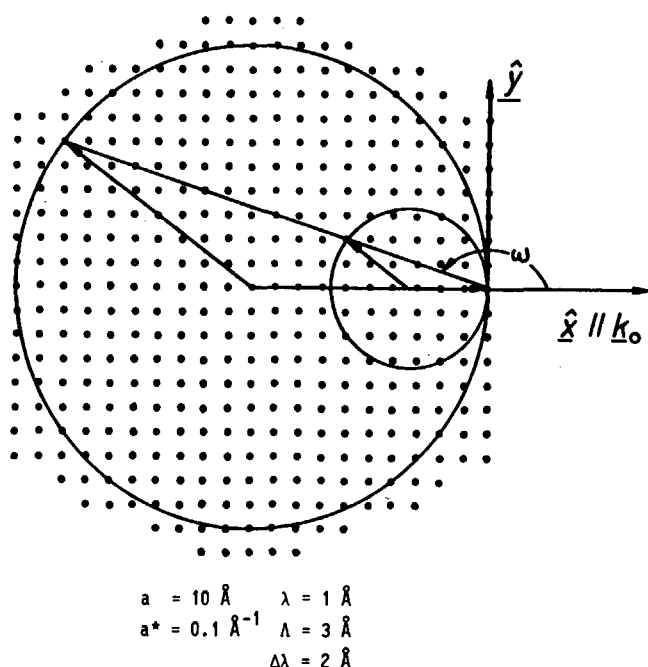


Fig. 5. Typical scattering geometry

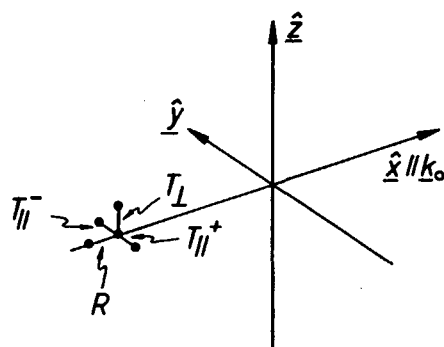


Fig. 6.  
The meaning of radial (R)  
and transverse ( $T_{\parallel}^{\pm}$ ,  $T_{\perp}$ )  
resolution

A cubic crystal with  $a = 10 \text{ \AA}$  and no extinctions is assumed throughout. In all cases a pulse with  $300 (\equiv \tau_{\text{SNQ}})$ , 60, 30  $\mu\text{sec}$  duration is assumed. During such pulses the film cylinder would move by  $\Delta\alpha = 10.8^\circ$ ,  $2.2^\circ$  and  $1.1^\circ$ , respectively. If we require radial reflections  $(30, 0, 0)$  and  $(31, 0, 0)$  to be separated at  $\beta = 90^\circ$  (horizontal plane) as well as at  $\beta = 60^\circ$  then we need a 30  $\mu\text{sec}$  chopper pulse (fig. 7). The transverse in-plane-resolution  $T_{\parallel}^{\pm}$  is more complicated (fig. 8). At  $\omega = 225^\circ$  a 60  $\mu\text{sec}$  pulse is sufficient for a

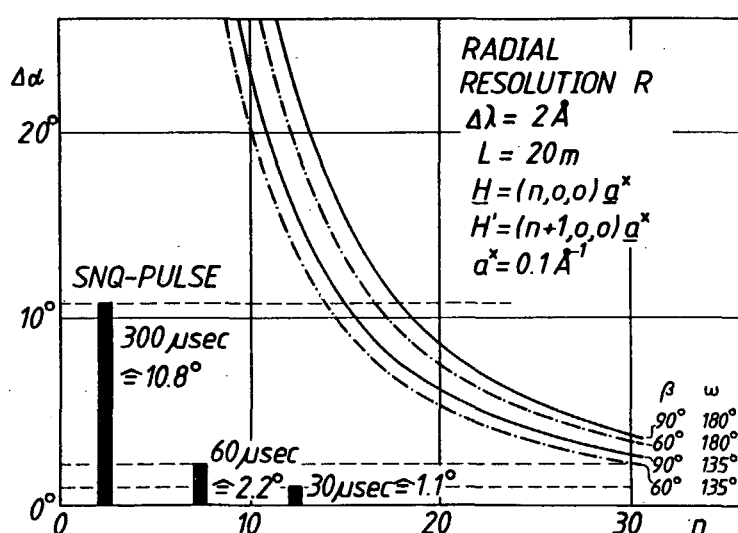
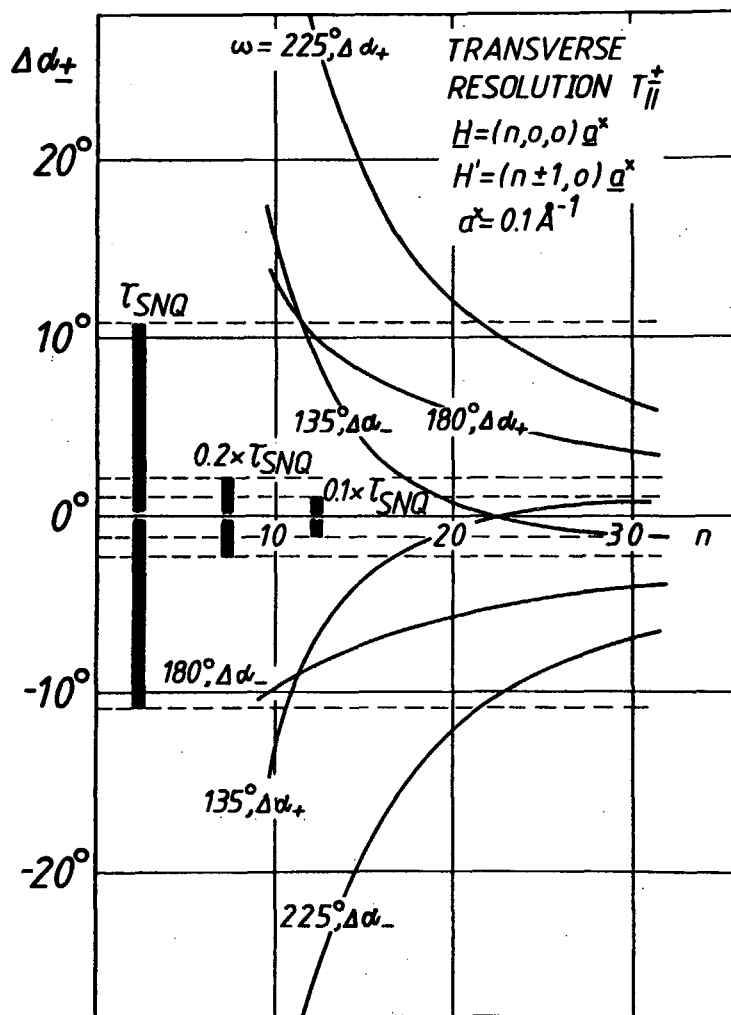
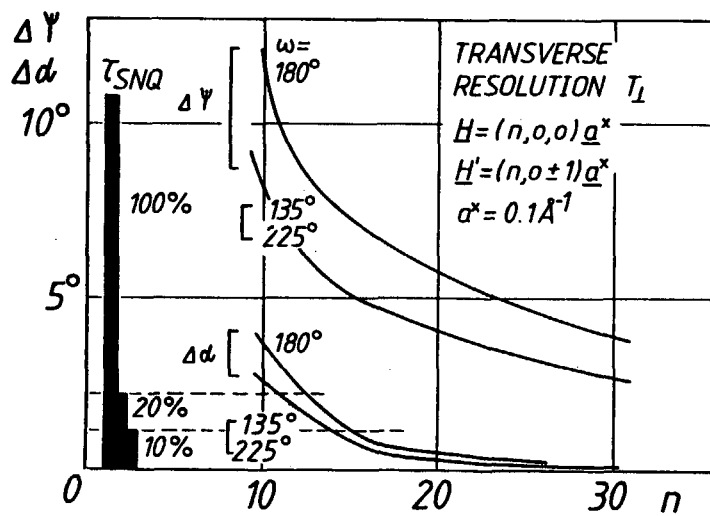


Fig. 7. Radial resolution vs. order  $n$  of a reflection.  
For  $\beta = 90^\circ$ ,  $60^\circ$  we have  $\psi = 0^\circ$ ,  $37.8^\circ$ ,  
respectively

separation of reflections up to the 30<sup>th</sup> order. At  $\omega = 135^\circ$  however reflections  $(22, \pm 1, 0)$  will coincide. It is a principal limitation of the method proposed here, due to the open detector, that in some specific orientations reflections on non-radial lattice lines have the same image on the film (comp. the text explaining figs. 10-12). This accidental coincidence is however removed when the crystal orientation is changed. In order to scan the reciprocal lattice completely several orientations are needed and therefore these accidental coincidences are no real problem.

For completeness sake the out-of-plane transverse resolution is given in fig. 9. In this case reflections  $(n \ 0 \ 0) \underline{a}^*$  will occur on the equatorial line of the film whereas  $(n \ 0 \ \pm 1) \underline{a}^*$  is shifted by  $\pm \Delta\psi$ . Even with the small  $\Delta\alpha$ 's shown in fig. 9 these reflections are separated as long as the vertical

Fig. 8. Transverse resolution  $T_{II}^{\pm}$ Fig. 9. Transverse resolution  $T_I$

divergence of the primary beam is smaller than  $\approx 3^\circ$ . This condition is definitely obeyed since with a distance  $L = 20$  m or larger neutron guides must be used which provide for a smaller beam divergence even with  $3 \text{ \AA}$  neutrons.

The coincidence problem (fig. 8) we arrived at and possibilities circumventing it are demonstrated in fig. 10. The curves given are locations in reciprocal

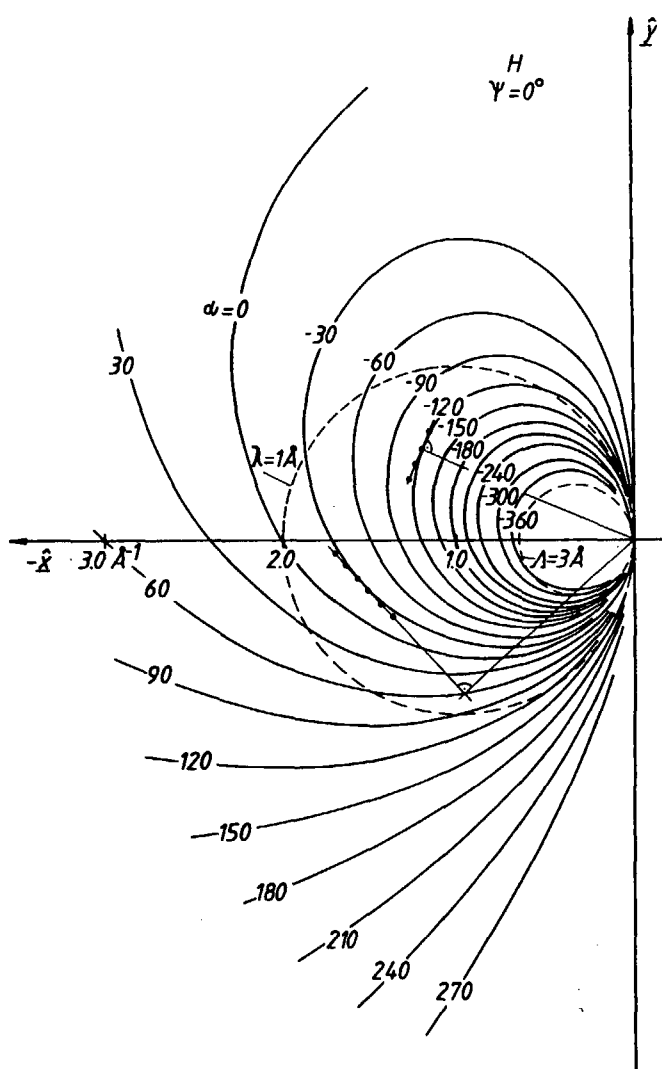


Fig. 10.

Coincidence curves in the  $\hat{x} - \hat{y}$  plane ( $\psi = 0$ )

space which map onto one film spot with the coordinates  $\alpha$  ( $\alpha$  is the curve parameter) and  $\psi = 0$ . It is immediately clear that reflections on lattice lines which for a particular crystal orientation are tangential to one of the curves will coincide on the film. In any case, however, this unfavorable condition can be removed by rotating the crystal. This is demonstra-

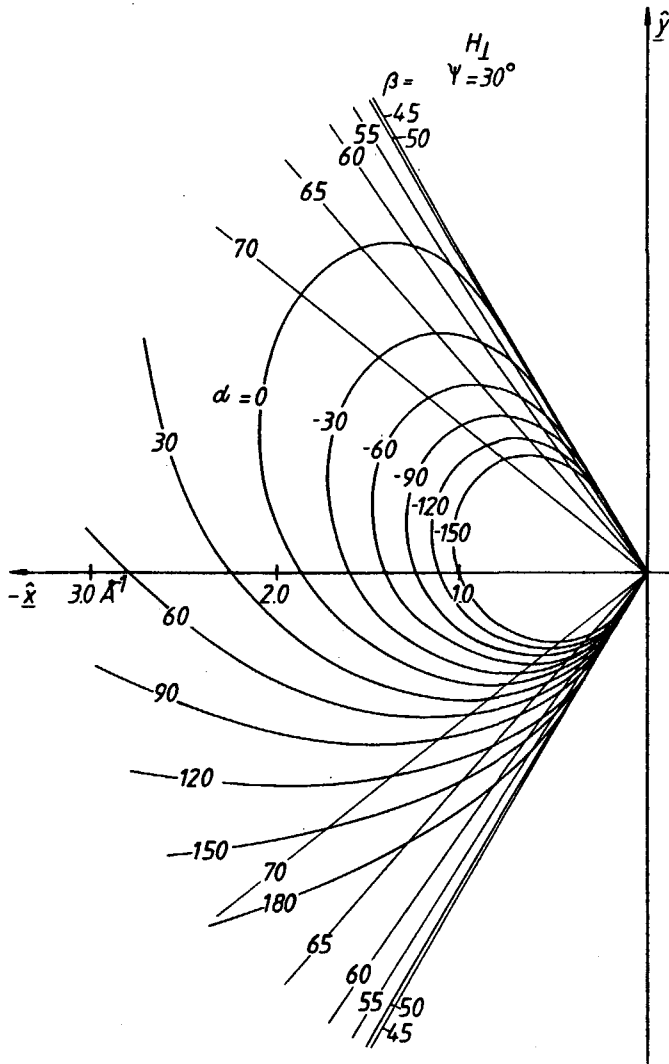


Fig. 11. Out-of-plane coincidence curves ( $\psi = 30^\circ$ )

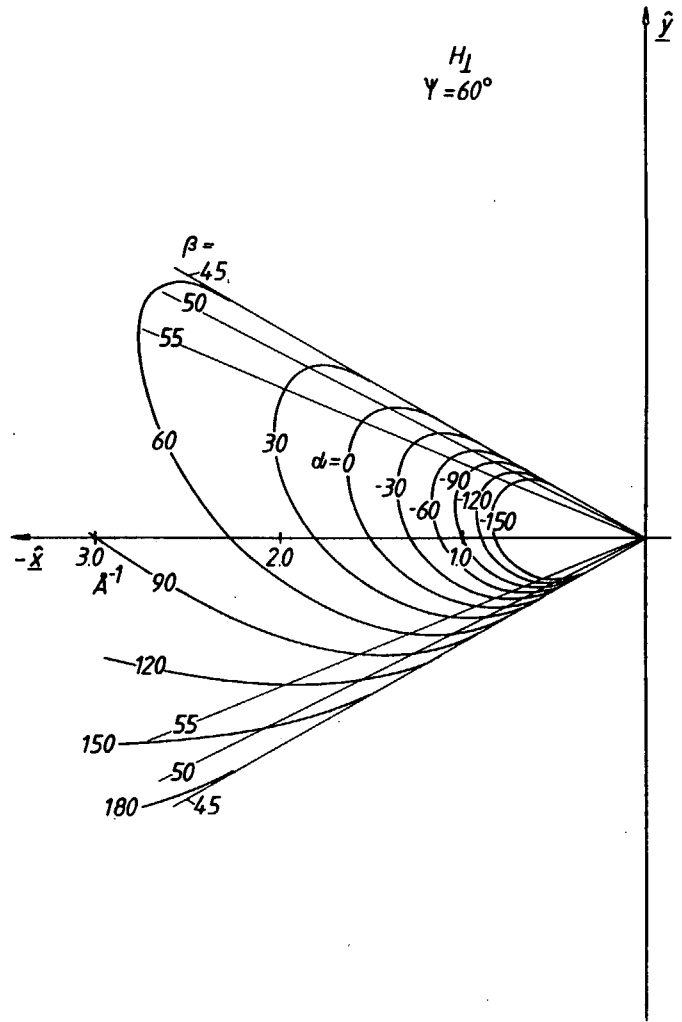


Fig. 12. Out-of-plane coincidence curves ( $\psi = 60^\circ$ )

ted in fig. 10 for one lattice line in two different orientations. All curves in fig. 10 are flat. This is due to the inherent horizontal symmetry plane of the instrument. In figs. 11 and 12 we show the projection of curves in reciprocal space the points of which coincide on the film at  $\alpha$  as given by the curve parameter and at  $\psi = 30^\circ$  (fig. 11), and  $\psi = 60^\circ$  (fig. 12), respectively. For  $\psi \neq 0$  the coincidence curves are not flat any more as indicated by the rays with constant polar angle  $\beta$ . Due to this double curvature of the coincidence curves for  $\psi \neq 0$  the problem of accidental coincidences is relaxed for reflections in such a region. Strictly speaking the worst case situation mentioned earlier does not occur actually because observations in the equatorial plane ( $\psi = 0$ ) are excluded due to the design of the camera: fig. 4. It is advantageous for the proposed instrument to orient the crystal unsymmetrically with

respect to the horizontal mirror plane of the instrument.

In order to estimate the number of measurable reflections we assume that only reflections in back scattering with  $120^\circ \leq 2\theta \leq 180^\circ$  ( $150^\circ \leq |\omega| \leq 180^\circ$ ) are registered. This restriction can be easily achieved by a screen. With  $\lambda = 1 \text{ \AA}$ ,  $\lambda + \Delta\lambda = 3 \text{ \AA}$  and a cubic crystal with  $a = 10 \text{ \AA}$  we find 2000 reflections for one crystal orientation. We allow for a 50 % reduction of this number due to blind spots of the instrument or accidental coincidences, and assume two exposures with 10 and 80 min for each orientation. Under these conditions we would expect that a Laue camera with photographic registration could measure 1000 reflections in 90 min or 16000 per day. With the current photographic neutron technique the standard deviation of intensities was found to be 7 % in a study of L-histidine  $\cdot$  HCl  $\cdot$  H<sub>2</sub>O [2c].

With a camera radius  $R = 5.73 \text{ cm}$  and a vertical divergence  $\psi = \pm 60^\circ$  (fig. 4) the total film area is  $71000 \text{ mm}^2$ . 2000 reflections, each with an estimated area of  $9 \text{ mm}^2$ , would cover 25 % of the film area. If the camera radius is doubled the coverage reduces to roughly 6.25 % as long as the spot size is given by the crystal size rather than by the divergence of the reflected beam.

#### (4) Gain

The problem of the gain factor is the standard instrument at a high flux steady state reactor to compare the proposed machine with. We first compare two identical Laue cameras with photographic registration, one operating at the SNQ ( $\hat{\phi}$ ), the other at the ILL high flux reactor ( $\bar{\phi}$ ). For the SNQ we assume  $\tau_{\text{proton}} = 250 \text{ } \mu\text{sec}$  and  $\tau_{\text{moderator}} = 120 \text{ } \mu\text{sec}$ , from which we find  $\hat{\phi}/\bar{\phi} = 35$ . For chopper pulse widths of 30, 60, 300  $\mu\text{sec}$  the effective peak flux is 34, 33,  $25 \times \bar{\phi}$ , respectively if the chopper window can be placed symmetrically with respect to the SNQ thermal neutron pulse maximum. At the HFR the effective peak flux is  $\bar{\phi}$  in all cases. We therefore have relative gain factors of 34, 33 and 25.

We arrive at roughly the same gain factors when we compare the ILL instrument using the modified Laue method with our proposed machine. In the modified Laue method [2] a curved monochromator is used to focus a wavelength band onto the sample, the important feature being that the wavelength of the neutrons arriving and the direction of incidence are correlated. Due to this correlation neutrons reflected from  $(n \ 0 \ 0) \underline{a}^*$  and  $(n \pm 1, 0, 0) \cdot \underline{a}^*$  planes travel in different directions and hit the film at different spots. The crystal is stationary and the integration over the mosaic distribution is done by the divergence of the primary beam ( $3^\circ$ ). Typical exposure times at the ILL HFR are 30 min and this is the exposure time to be expected for a rotating film

device at the SNQ also since it is the average flux  $\bar{\phi}$  which counts for a single reflection. The gain factor then will be given as the ratio between the numbers of reflections registered simultaneously in both cases with some correction for the reflectivity of the monochromator used in the modified Laue method. With some arbitrary but reasonable assumptions we arrive at  $g \approx 20$  in this case.

In these estimates we have not taken into account differences in the thermal spectrum of the two sources.

#### (5) Measuring strategy

The best one can do in measuring Bragg reflections is to measure all of them, and to measure as many as possible under the conditions of optimum resolution and highest intensity: this means close to back scattering and in the wavelength band  $1\text{\AA} \leq \lambda \leq 3\text{\AA}$ . A Laue photographic camera should therefore scan the reciprocal space in equal pieces of solid angle, and the scanning strategy can be made completely independent of the actual crystal under investigation. The most regular tessellation of the unit sphere is given by the regular Platonic polytopes. Let us consider, e.g., an icosahedron fixed rigidly with the crystal under investigation. Then all of the vectors  $\underline{H} = h\underline{a}^* + k\underline{b}^* + l\underline{c}^*$  can be classified into 20 sets, each set containing those rays  $\lambda\underline{H}$  which penetrate the icosahedron within one particular triangle. In order to record all reflections of a crystal we need 20 orientations. In an actual camera the region of reciprocal space which is mapped simultaneously in one orientation of the sample will not have the shape of a solid triangle. Rather, there will be some overlap of the recorded regions which allows for a correct scaling of data common to different films.

It might be that with the 20 orientations given by the icosahedron there remain some blind spots due to a limitation of the horizontal or vertical angle of acceptance of the camera. In such a case other tessellations based e.g. on a general  $\{hkl\}$  polyhedron with cubic symmetry (48 faces) can be found. In any case there is a small number of orientations characterised by a set of Eulerian angles  $\{\alpha_i, \beta_i, \gamma_i\}$  which is sufficient for any crystal. This means that the software for the crystal orientation is very simple.

In order to speed up measurements and to minimize service a dozen (e.g.) of film cylinders with their drives could be mounted on a kind of merry-go-round (fig. 13). For every new orientation of the crystal one would use one or more



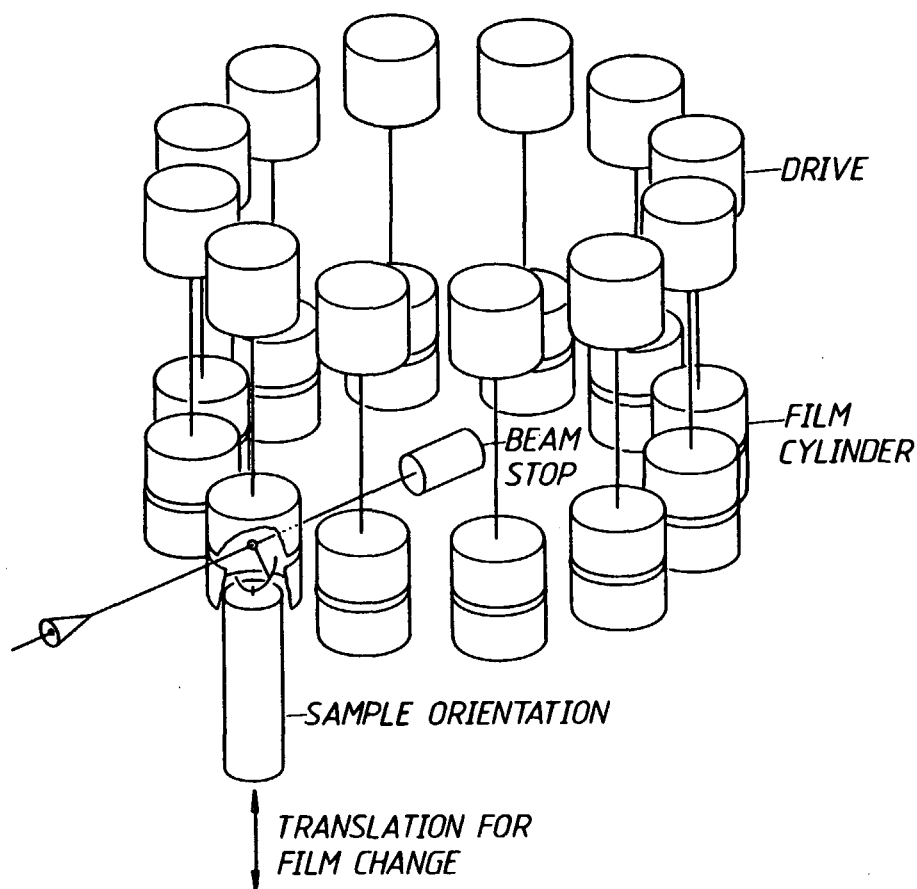


Fig. 13. Automatic film changer

films (with different exposure times). Processing of the films should be done by an automatic batch processor operating under well defined standard conditions.

#### (6) Efficient use of beams and chopper systems

The average sample size for diffraction experiments is estimated to be less than 3 mm in diameter. This means that a neutron beam diameter of 5 mm is sufficient. Resolution on the other hand requires a distance of 20 m between the choppers and the samples, and for such a long distance neutron guides must be used. Narrow guides (5 mm inner diam.) need only a small curvature in order to avoid the direct sight from one end to the other. Several guides starting from the same chopper system could be used to conduct neutrons to several instruments of this kind.

### (7) Advantages, disadvantages, open questions

A Laue-TOF-diffractometer would be of a very simple mechanical design. It would use only reliable and long proven technology. Phase jitter in the film drive is not very critical because small phase errors are averaged. Diffractometers of this kind could be built easily and at low costs (if costs should play an important role in a project of this size). Several diffractometers could share one beam. A properly designed instrument would need to be serviced only once a day, and all films could be processed automatically. Fast film scanners are available today, and so the conversion of the data to digital form is a simple routine. There are some apparent disadvantages the importance of which must be investigated. Since the detector records all neutrons hitting one spot on the film the actual local exposure is due to scattering events which originate from an extended region of the reciprocal space (cf. figs. 10,11,12). The situation in our case is comparable to an X-ray or neutron rotation / oscillation camera which uses monochromatic radiation: in the latter case the Bragg spots are superimposed on a background due to scattering from circles/circular arcs in  $R^*$ , and usually the background separation is no problem. X-ray cameras without layer line screens are standard in macromolecular crystallography [3].

Unfavorable for the Laue-TOF-photographic camera as well as for an oscillation neutron camera are samples with a high amount of chemical disorder and/or incoherent scattering. We emphasize that, for the reason just mentioned, the camera proposed here is restricted to the measurement of Bragg intensities. - Accidental coincidences of Bragg reflections may occur. Their frequency depends on the lattice geometry of the sample under investigation and on the orientation. These coincidences can be removed, however.

There are open questions. First of all it is not clear whether a Fermi chopper is the best choice for the selection of  $\lambda$  and  $\Delta\lambda$ . The quantum efficiency of the neutron-to-photon converters used in photographic work is not known very accurately. It is very likely that this efficiency could be improved with new materials or new techniques.

For a camera designed for high momentum transfer  $Q = 2\pi H \approx 20 \text{ \AA}^{-1}$  neutrons with a wavelength of  $0.6 \text{ \AA}$  are needed. Because of the  $\lambda^4$ -dependence of the Lorentz factor for the Laue method such an instrument should operate at a hot rather than at a thermal source.

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Small Angle Scattering

S M A L L - A N G L E   S C A T T E R I N G  
A N D   D I F F U S E   S C A T T E R I N G

Heinrich B. Stuhmann

Institut für Physikalische Chemie der Johannes-Gutenberg-Universität, 65 Mainz

These techniques investigate a wide variety of materials, which are mostly non-crystalline. Most of the material we encounter in daily life will give rise a diffuse scattering pattern if it is irradiated by thermal neutrons. Steel knives and plastic spoons will scatter thermal neutrons in a similar way as the meat you can cut with it and the soup you can eat. There will be relatively strong forward scattering, called small-angle scattering and diffuse scattering at wider angles eventually decorated with some diffraction peaks. The molecular structure of these materials is too complicated as to be determined by one method only. The share of neutron scattering techniques however is considerable and in many cases the contributions of neutron scattering have to be regarded as unique.

This is so, as the dependance of the scattering on the isotope and the nuclear and electronic spin state, opens quite new ways of structure research. Specific isotopic replacement provides the scenario we need to understand the scattering pattern. Spin dependent change of the scattering length by external fields is a non-destructive method of contrast variation.

It is therefore important to know, how these techniques would develop, if a pulsed neutron source were available. In most cases, gain factors due to the puls structure are achieved by time-of-flight techniques. The advantage of a pulsed source with respect to a steady state neutron reactor strongly depends on the specific problem. In small angle scattering, for instance it may assume any value between 1 and approximately 15. A similarly wide spread is encountered in other scattering methods of structure determination.

# LOW - Q - SCATTERING

## ( Small - Angle Scattering )

Heinrich B. Stuhrmann

Institut für Physikalische Chemie der Johannes-Gutenberg-Universität, 65 Mainz

Konrad Ibel

Institut Max von Laue - Paul Langevin, 38042 Grenoble, France

Jean-Pierre Cotton

Laboratoire Léon Brillouin, C.E.N. Saclay, 91191 Gif-sur-Yvette, France

Knud H. Nierhaus

Max-Planck-Institut für Molekulare Genetik, Ihnestraße 63-73, 1000 Berlin 33

### Abstract

Low-Q scattering or neutron small-angle scattering has been developed to a high performance at reactor neutron sources. The pulsed neutron beam from the spallation neutron source offers additional advantages. Using time-of-flight methods the gain factor at an SNQ operated at 100 Hz frequency may assume values up to 8. At lower frequency, say 25 Hz, an additional gain by a factor 2 to 4 can be obtained. Cold neutrons remain superior to neutrons thermalized at room temperature.

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## INTRODUCTION

Low-Q scattering of neutrons is mainly an elastic scattering process. The momentum transfer  $Q$  therefore is simply related to the scattering angle  $2\theta$  and the (unchanged) wavelength by

$$Q = 4\pi / \lambda \sin \theta \quad (1)$$

Low-Q scattering originates from heterogeneities of any kind in the colloidal range. Among them free molecules represent a particularly simple case. In forward direction, i.e.  $Q = 0$ , the scattering intensity then is proportional to the square of the sum of amplitude of the atoms each molecule is made of. A molecule ten times bigger in molecular weight will scatter the incident neutrons at a probability which is a hundred times higher. Dilute solutions of macromolecules give rise to strong low-Q scattering.

The halfwidth of the central peak of low-Q scattering is inversely proportional to the mean dimensions of the macromolecule. To a good approximation it is related to the radius of gyration  $R$  of the scattering particle by

$$Q_{1/2} = 1.4 / R \quad (2)$$

Scattering of particles characterized by  $R = 70 \text{ \AA}$  (e.g. ribosomes, see Nierhaus (1984)) will have dropped to half the value of forward scattering at  $Q = 1.4/70 = 0.02 \text{ 1/\AA}$ . Using thermal neutrons with wavelengths around  $6 \text{ \AA}$  this is going to happen at a scattering angle  $2\theta = Q \lambda / 2\pi = 0.02 * 6 / 6.28 = 0.02 \text{ radians} = 3^\circ$ . For  $1.54 \text{ \AA}$  X-rays emitted by ordinary X-ray tubes, the halfwidth of the central peak would decrease to less than  $1^\circ$ . Therefore low-Q scattering may be observed as small-angle scattering of thermal neutrons or X-rays. We just mention that the much more widely used technique of light scattering using wavelengths between  $4000$  and  $8000 \text{ \AA}$  would extend the angular width to the backscattering region.

The other way to measure low-Q scattering consists in monitoring the scattering intensity as a function of the wavelength at constant scattering angle. This is a most attractive alternative both with X-ray synchrotron radiation and pulsed neutron sources. The energy analysis is achieved by a energy-dispersive X-ray detectors and time of flight methods respectively. It is the applicability of this method which makes a pulsed neutron source and the SNQ in particular superior to a continuously operating research reactor.

The objective of this article is twofold. It will describe the performance of existing small-angle instruments at a pulsed source and it will try to show which modifications are necessary, in order to make full use of the new features of a pulsed source. To simplify the discussion we shall refer to the total small-angle scattering of macromolecules in dilute solutions and use this as a standard for comparison of different techniques. Given an experimental set up the question then is: what is the total number of neutrons scattered at low  $Q$  per second ?

## PRINCIPLES OF LOW- $Q$ -SCATTERING

(the still most comprehensive treatment of small-angle scattering is given by A. Guinier in his famous book "Small-Angle scattering of X-rays", Wiley, 1955. For a more recent review see G. Porod, in "Small-Angle X-ray Scattering", O. Glatter, O. Kratky eds. Academic Press 1982)

The total low- $Q$  scattering is intimately related to the

$$\text{correlation length } l_c = 2 \int_0^D \gamma(r) dr \quad (3)$$

which is the mean width of the correlation function  $\gamma(r)$ . If lines are drawn through all points of a particle  $\rho(\vec{r})$  in all directions the number average of chords so obtained is equal to  $l_c$ .

To find  $l_c$  from the intensity function  $I(Q)$  we consider the integral

$$\int_0^\infty I(Q) Q dQ = \bar{\rho}^2 V 4\pi \int \gamma(r) dr = \bar{\rho}^2 V 2\pi l_c \quad (4)$$

In dilute systems  $V$  is the particle volume and  $\bar{\rho}$  its contrast with respect to the solvent density. This integral can be expressed in terms of the total energy  $P$  scattered in all the low energy region. On a film placed in a distance  $a$  from the sample, the area that receives the rays scattered through the small angles contained between  $2\theta$  and  $2\theta + d(2\theta)$  can be written to a first approximation as

$$dS = 2 a^2 2\theta d(2\theta) = (\lambda^2 / 2\pi) a^2 Q dQ \quad (5)$$

The ratio between the incident intensity ( $P$ ) and the total small-angle scattering ( $S$ ) then is



$$S/P = \frac{1}{P} \int I(Q) d\Omega = (\lambda^2 / 2\pi) \int I(Q) Q dQ = \lambda^2 \bar{\rho}^2 v l_c \quad (6)$$

If a dilute solution contains  $c$  grams of the solute with a specific volume of  $v \text{ cm}^3/\text{g}$  then

$$S/P = \lambda^2 \bar{\rho}^2 c v l_c d e^{-\mu d} \quad (7)$$

where  $d$  is the thickness of the sample and  $\mu$  the absorption coefficient. The scattering density increases with the thickness of the sample and it reaches its maximum when the incident intensity has dropped to about  $1/e = .37$  where  $d = 1/\mu$ .

$$S/P = 0.37 \lambda^2 \bar{\rho}^2 c v l_c d_{\text{opt}} \quad (8)$$

For spherical structures  $l_c = 1.5 R$ . The radius of gyration  $R$  amounts to 0.8 of the radius of the sphere. With  $\lambda$  and  $R$  in  $\text{\AA}$ ,  $\bar{\rho}$  in  $10^{10} \text{ cm}^{-2}$ ,  $v$  in  $\text{cm}^3/\text{g}$  and  $d_{\text{opt}}$  in cm we obtain the probability of scattering into small angles by spheres

$$S/P_{(\text{spheres})} = 5.5 \cdot 10^{-5} \bar{\rho}^2 \lambda^2 c R v d_{\text{opt}} \quad (9)$$

Example: neutron scattering from 1% solution ( $c = 0.01 \text{ g/cm}^3$ ) of ribosomes.  $R = 70 \text{ \AA}$ ,  $\bar{\rho} = 3 \cdot 10^{10} \text{ cm}^{-2}$ ,  $\lambda = 5 \text{ \AA}$ ,  $d_{\text{opt}} = 1 \text{ cm}$ ,  $v = 0.6 \text{ cm}^3/\text{g}$

$$S/P = 5.5 \cdot 10^{-5} \cdot 25 \cdot 9 \cdot 0.02 \cdot 70 \cdot 0.6 \cdot 1 = 0.005 = 0.5\%$$

In biological systems neutron enter the channel of small-angle scattering at a much lower rate than those of other, usually incoherent scattering processes. The total small-angle scattering would decrease by 10 if we would use  $\lambda = 1.5 \text{ \AA}$ . The optimal thickness of heavy water solutions varies only slightly with the wavelength.

#### THE THERMAL NEUTRON SPECTRUM

The velocity distribution of neutrons, which are in thermal equilibrium with a moderator of temperature  $T$ , is given by

$$f(u) = \frac{1}{(\sqrt{\pi} u_T)^3} \exp\left(-\frac{u}{u_T}\right)^2 \quad (10)$$

$u_T$  is the most probable velocity:  $u_T = (2kT/m)^{1/2}$  and  $\int_{-\infty}^{\infty} f(u) du = 1$ . The phase space density can be written as  $N = N_0 f(u)$ .  $N_0$  is the number of neutrons per  $\text{cm}^3$ . With  $\Phi = 2 N_0 u_T / \sqrt{\pi}$  and  $\Delta z = v \Delta t$  the number of neutrons within the velocity space volume  $\Delta v_x \Delta v_y \Delta v_z$  passing through an area  $\Delta x \Delta y$  per second is given by

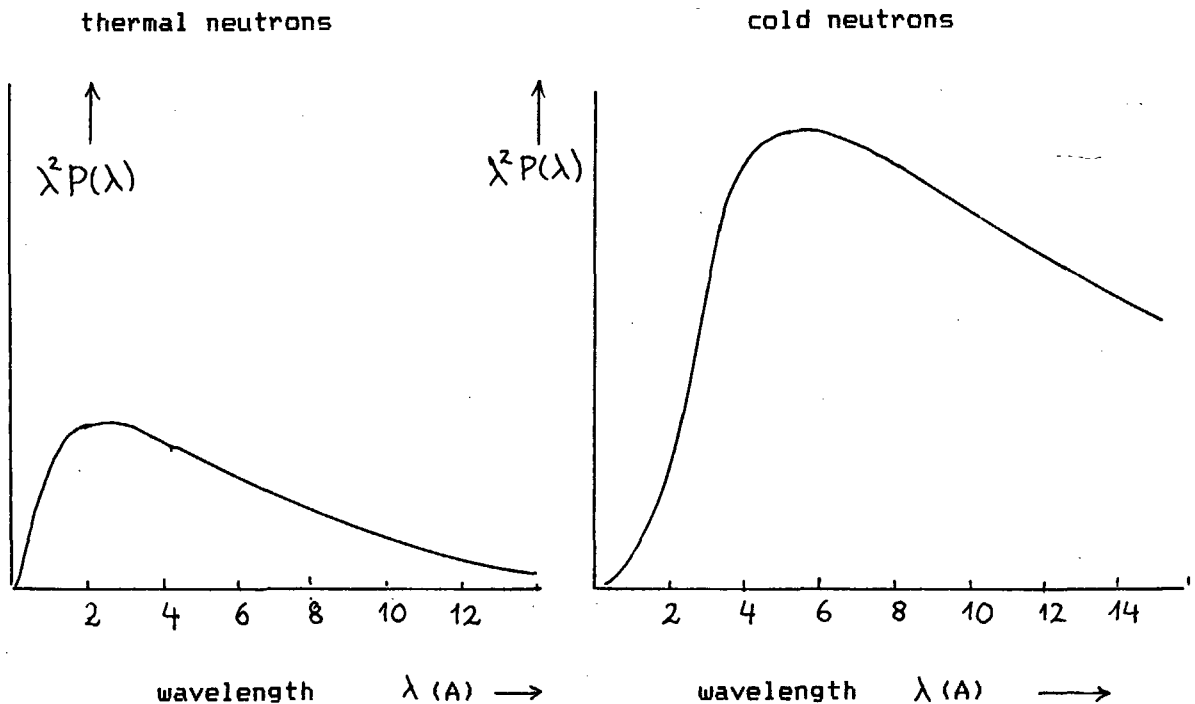
$$\Delta^5 I = \frac{\Phi}{2\pi} \frac{u}{u_T} \exp \left( - \frac{u}{u_T} \right)^2 \Delta x \Delta y \Delta v_x \Delta v_y \Delta v_z \quad (11)$$

This function has a maximum for  $u = u_T / \sqrt{2}$ . Transformation to the wavelength scale according to the relation  $\lambda = h / (mu)$  yields:

$$\Phi(\lambda) d\lambda = \frac{\lambda^3}{\lambda_T^4} \exp \left( - \frac{\lambda_T}{\lambda} \right)^2 d\lambda \quad (12)$$

where  $\lambda_T$  is the most probable wavelength. With  $\lambda_T = 3.6 \text{ \AA}$  this type of distribution fits fairly well the wavelength spectrum of cold neutrons used by D11 of the ILL. For neutrons emerging from a heavy water moderator  $\lambda_T$  was assumed to be  $1.35 \text{ \AA}$ .

The scattering intensity has been shown to increase with the square of the wavelength. The advantage of using cold neutrons in coherent scattering experiments becomes more apparent after multiplication of the wavelength spectra with  $\lambda^2$ :



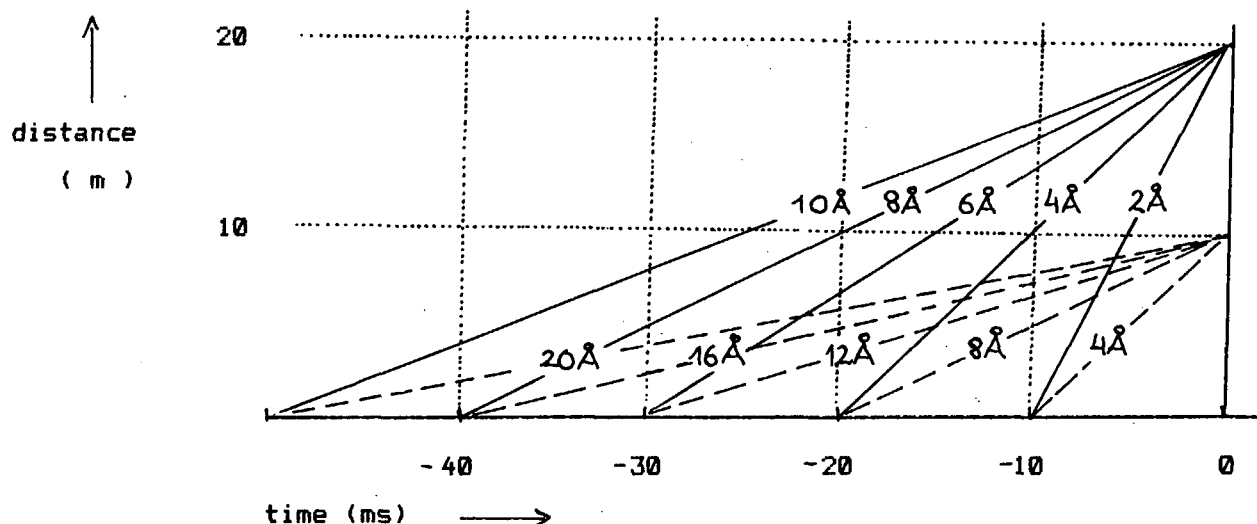
## THE USE OF A PULSED NEUTRON SOURCE

Each 10 ms a pulse of about 0.5 ms duration will leave the heavy water moderator of the SNQ. If the neutrons would propagate with equal velocity, say 1000 m/s for 4 Å neutrons, then the shape of the pulse would be conserved. A snap shot of the spatial distribution of neutrons would show pulses at 10 m spacing, as is shown on the next page.

The picture changes considerably when no velocity selection is foreseen. At 10m distance from the source, we will find neutrons with wavelengths of 4Å, 8Å, 12Å, 16Å, .. while a new pulse of neutrons is created in the moderator. At greater distances the situation gets even more confusing: at 40 m distance we would find at the same time neutrons with wavelengths of 1Å, 2Å, 3Å, and so on. In general we have

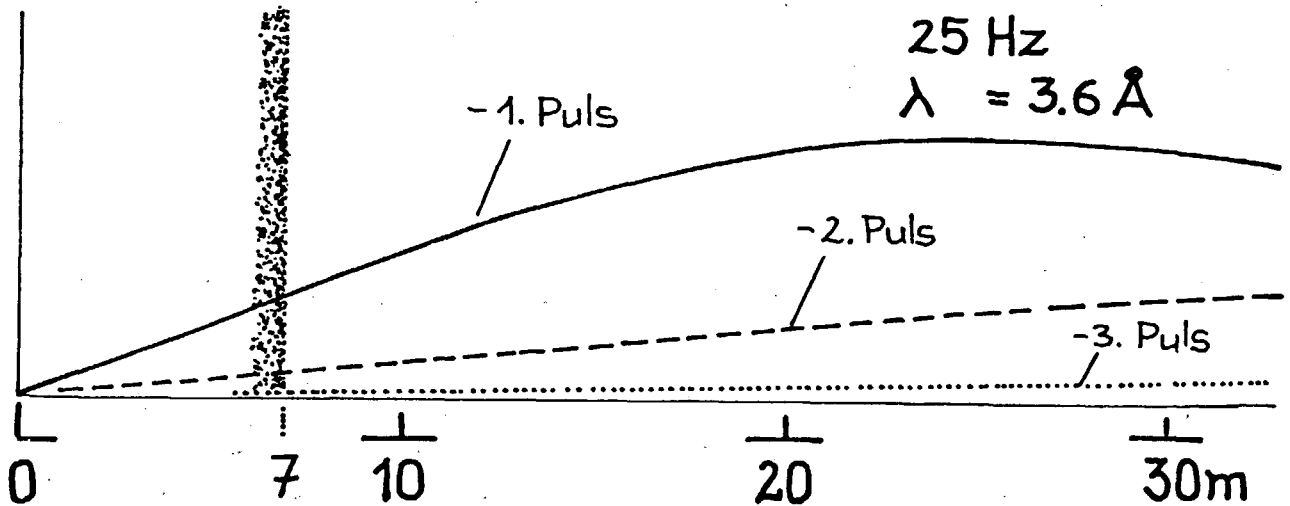
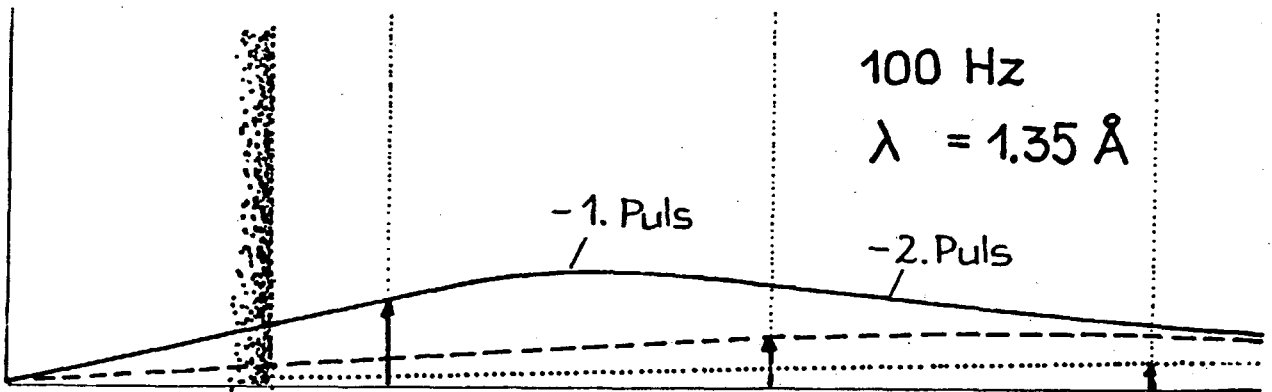
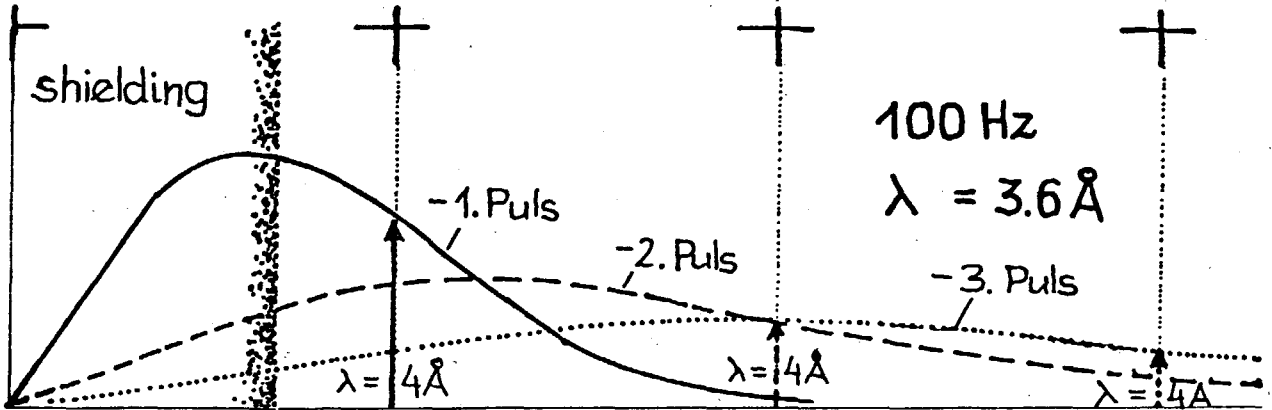
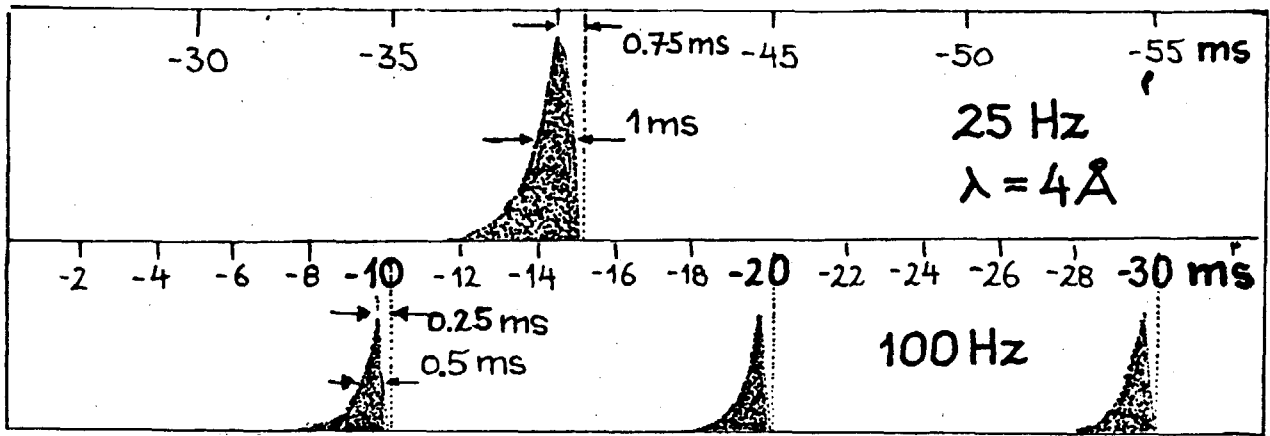
$$n \lambda_0 [\text{\AA}] = 4000 / (\text{frequency} [1/\text{s}] * \text{distance} [\text{m}]) \quad n = 0, 1, 2, \dots \quad (13)$$

This is illustrated by the time-table of the free neutrons given below.



At a lower puls frequency, say 25 Hz, the same degree of pulse overlap is shifted to distances four times larger.

This is illustrated by the following figure, which shows the spatial distribution of three neighbouring pulses. Pulses of cold neutrons emitted at a frequency of 100 Hz tend to overlap already within the 7m shielding of the SNQ. Thermal neutrons are faster on the average. Therefore the same degree of overlap will occur at a larger distance. In this respect they compare to cold neutrons emitted at the much lower frequency of 25 Hz.

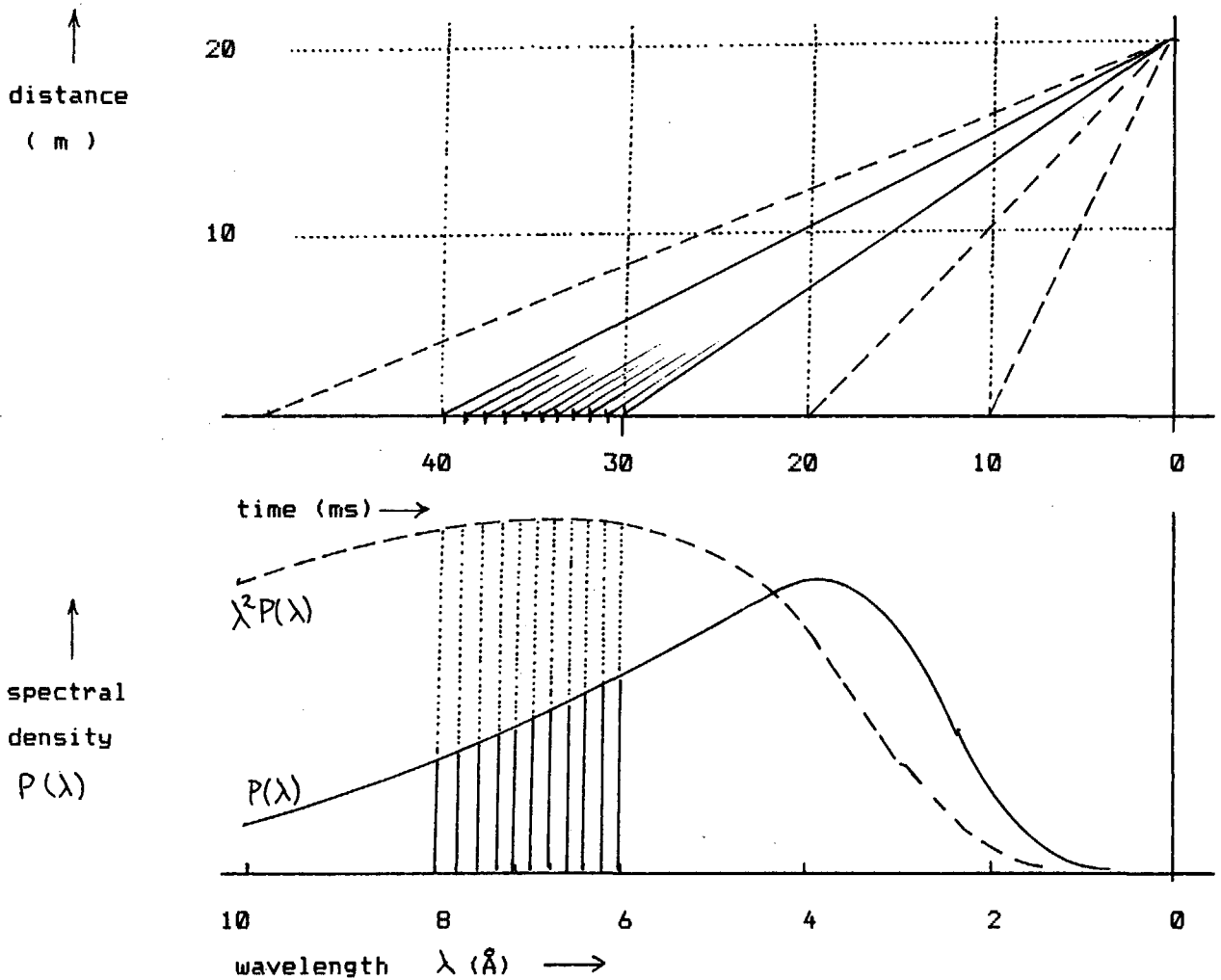


Spatial distribution of monochromatic and polychromatic neutron pulses .

For a well-defined time-of-flight experiment, pulse (or frame) overlap has to be avoided, that is, one has to know the age of the free neutron for each scattering event. The wavelength range has to be chosen in such a way that no pulse overlap will occur. If there are  $M$  measurements of neutron scattering per period, then the following wavelengths  $\lambda_m$  may be chosen:

$$\lambda_m = \lambda_0 (n + m/M) \quad n = \text{const.}, m = 1, 2, 3, \dots, M \quad (14)$$

In practice,  $m = 1$  will have to be suppressed, because during this time epithermal neutrons may lead to an increased background count rate on the detectors. The choice of  $n = 0, 1, 2, \dots$  allows an optimization of the scattering experiment.



With the wavelength distribution given in Eq.(12) we obtain the scattering intensity  $I(2\theta)$  on the detector at a given time, characterized by  $m$  :

$$I(2\theta) = P[\lambda_0(n + m/M)] I[4\pi \sin \theta / (\lambda_0(n + m/M))] \quad (15)$$

As the shielding of the spallation neutron source will amount to about 7m the minimum flight path of the neutron from the source to the detector will be 10m. The collimation length should be as long as the distance d from the sample to the detector. Allowing 1m for velocity selection facilities in addition to the 7m shielding the total flight path of the neutrons will be  $a = 8 + 2 d$ . For small angles we have  $\sin \theta = x / 2d = x / (a - 8)$ . With the time  $t$  in ms between two neighbouring pulses the intensity given by Eq.(13) is changed to

$$I_m(x) = P [ 4 t (n + n/M)/a ] I [ a x / ( 4 t (a-7) (n+m/M) ) ] \quad (16)$$

$x$  and  $a$  are given in m. This equation is used for the calculation of the scattering intensity produced by the SNQ.

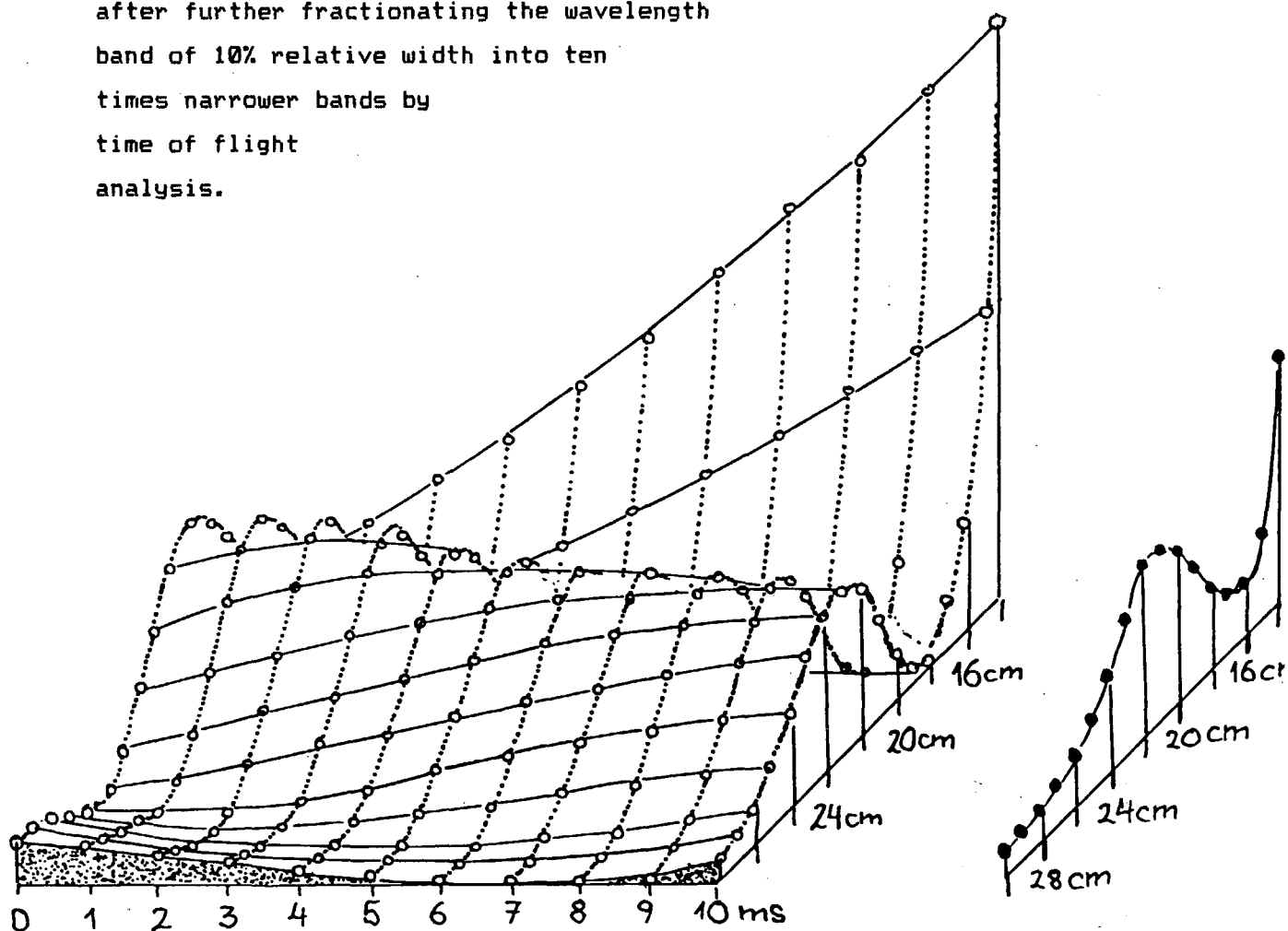
#### THE D11 INSTRUMENT OF THE ILL AT A PULSED NEUTRON SOURCE.

Although D11 is optimized for the use at the high flux reactor of the ILL, its possible performance at a pulsed source deserves some attention. The velocity selector at about 40m distance from the source transmits wavelength band of 9% full half width. Using wavelengths from 5 Å to 5.4 Å serious pulse overlap will occur at  $a(m) = 4 t(ms) \cdot (\lambda_2 - \lambda_1) = 4 \cdot 10 / 0.4 = 100$  m. This is roughly the length of the neutron flight path at D11. If the available wavelength band is just right, then no further gain can be achieved. D11 then would neither win nor lose at a pulse source, the mean neutron flux being equal to that of the HFR of the ILL.

A wavelength resolution better than 9% may be required with ordered structures, e.g. viruses. These structures may be described by rather few multipoles. In solution the scattering of each of the multipoles adds up independently to the total scattering function of the dissolved virus. As the particle is limited, each of these multipole scattering functions has its zeroes, which usually will not coincide. The depth of the minima of the scattering function of the total molecule are of particular interest, as the scattering of one of the multipoles will vanish there. This is most easily observed with the scattering function of the monopole, the zeroes of which easily can be identified in the small-angle region. The depth of these minima is a measure of the remaining multipoles, which describe the angular structure of the scatterer. This very desirable information is often obscured by the two broad wavelength band very often used with neutron scattering.

Time of flight analysis increases the wavelength resolution beyond the 10% available at D11:

The smeared out zeroes of the spherical particle ( $R = 600 \text{ \AA}$ ) on the right side of the figure will come out clearly after further fractionating the wavelength band of 10% relative width into ten times narrower bands by time of flight analysis.

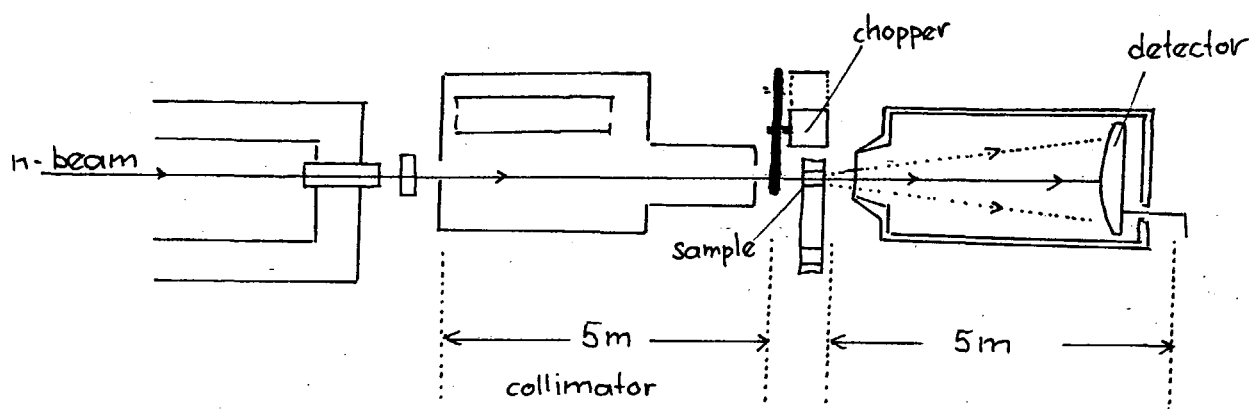


The flight distance of the neutron pulse is 72m in this case. The distance between sample and detector is 32m. The picture element of the detector is  $1\text{cm} \times 1\text{cm}$ . - Here it has been assumed that the finite cross section of the beam would not distort the scattering pattern. In the case of D11 the wavelength resolution of 10% roughly matches the spatial resolution of the detector. If a higher resolution of momentum transfer is required then the divergence of the beam has to be decreased as well. This will decrease the incident intensity at the sample quite independently of the nature of the neutron source. More stringent requirements on wavelength selection are most easily met by pulsed neutron sources using TOF-methods, without further reduction of the incident beam.

## A TIME-OF-FLIGHT INSTRUMENT AT A REACTOR

This is just the inverse of what has been described in the previous section. The use of the time-of-flight method relies on a pulsed neutron source. The steady neutron stream of the reactor has to be interrupted by a chopper creating pulses of the desired duration. It is interesting to see how the parameters are chosen, if there is free choice of the pulse structure.

We refer to the low-Q instrument PACE which just started to work at the reactor ORPHEE du C.E.N. Saclay. Here, only its time-of-flight option is presented schematically:



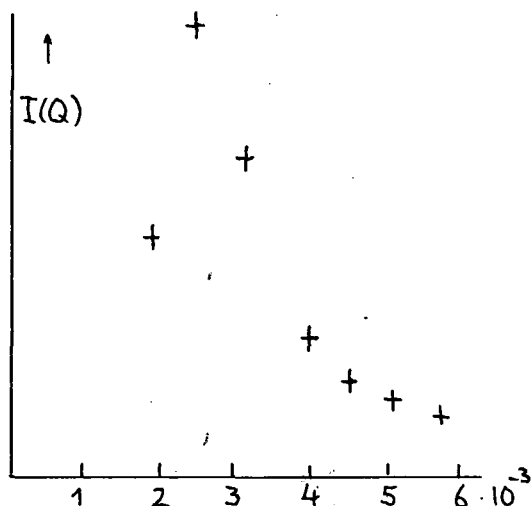
This set up uses a beam emitted by a neutron guide cutting off the wavelength spectrum below  $6 \text{ \AA}$ . The beam is collimated by two holes of 1cm diameter at 5m distance. The chopper is just behind the collimator and in front of the sample holder. It runs at less than 3000 rpm cutting down the intensity by a factor 25. Time-of-flight analysis occurs at a position sensitive area counter. The distance between sample and detector can be chosen freely to meet the requirements of the experiment. The detector consists of 30 concentric rings with radii between 3 and 32cm. At a distance of 2.5m the range of momentum transfer extends from  $3 \cdot 10^{-3}$  to  $0.35 \text{ 1/\AA}$ .

The large range of  $Q$  accessible to one measurement at the instrument PACE may be a considerable advantage. Low angle diffraction from block copolymers are a good example. These may have spacings of several  $1000 \text{ \AA}$ . As has been shown by one of us (J.P. Cotton) a classical set up ( $\lambda = 18 \text{ \AA}$ , distance sample-detector = 5m) will hardly reveal the  $2300 \text{ \AA}$  diffraction peak from the mesomorphous phase of polystyrene-polyisoprene block copolymers. Time-of-flight analysis at the innermost rings of the detector ( $\theta = 19^\circ, 25.9^\circ, 32.4^\circ$  and  $38.9^\circ$ ) gives a much clearer result.



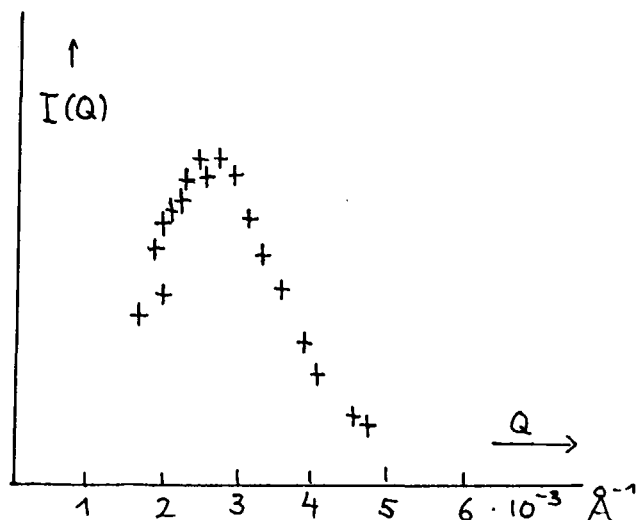
Low Q- scattering using a  
classical set up.

(  $\lambda = 18\text{\AA}$ ,  $D = 5\text{ m}$  )



Low-Q scattering using  
time-of-flight analysis.

(  $7.39 < \lambda < 20.7\text{\AA}$ ,  $D = 5.15\text{m}$ ,  $36\text{ Hz}$  )



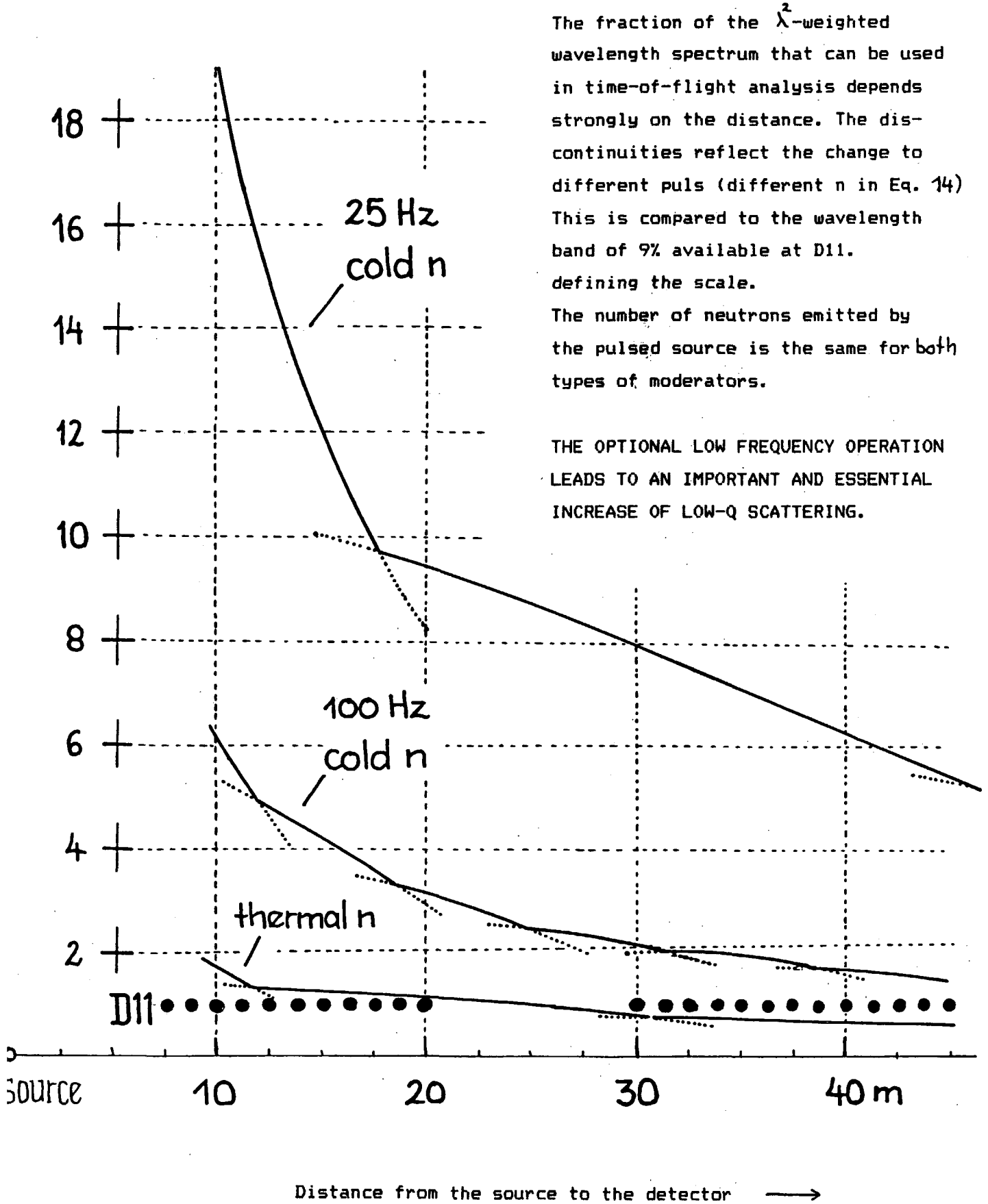
This result could also be obtained at the 20m position of D11, although with poorer resolution. - Time of flight measurements using neutrons from a reactor will not be faster than conventional small-angle scattering. Its advantage lies in the greater flexibility.

#### LOW - Q SCATTERING AT THE SNQ.

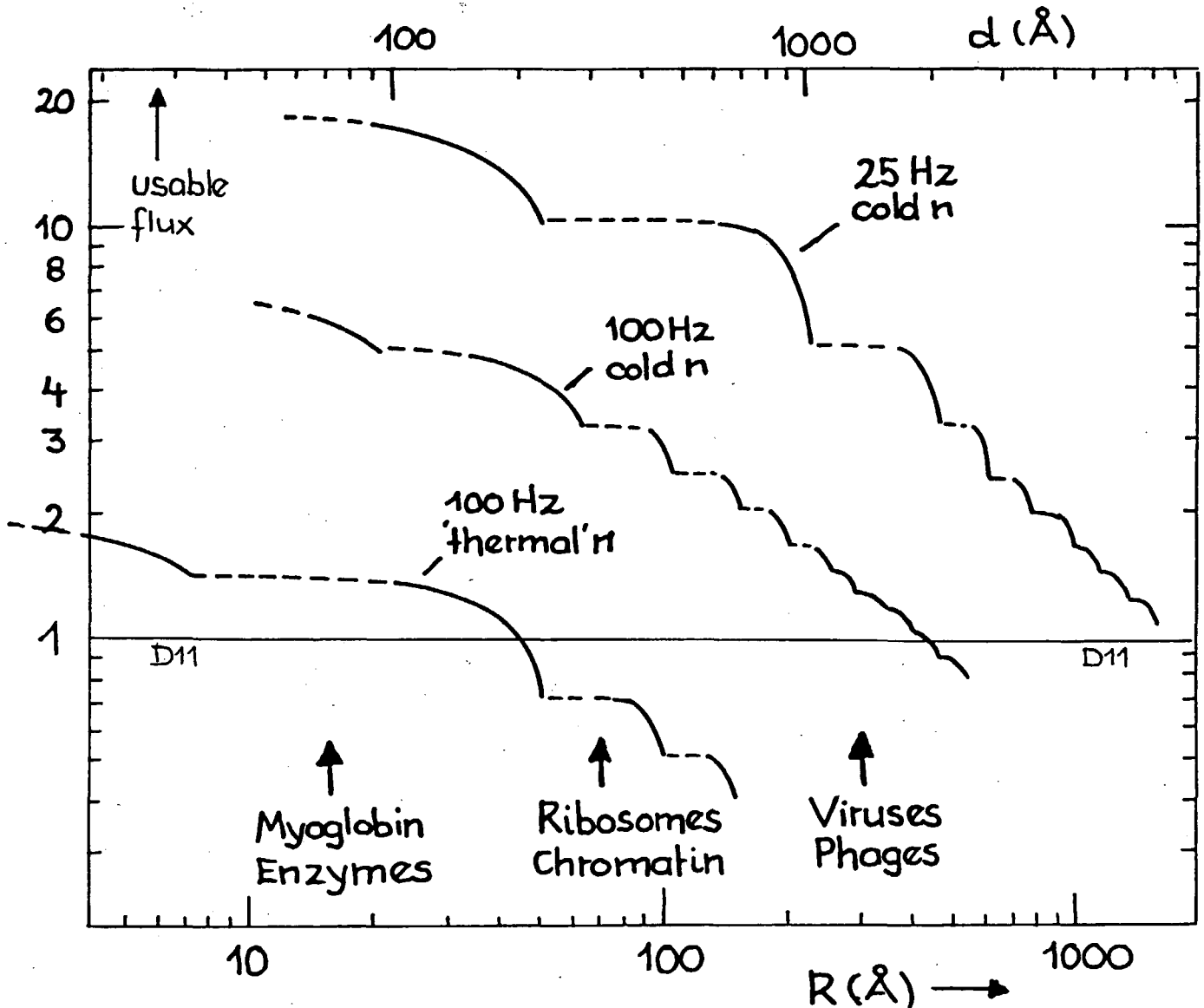
The gain factor at a pulsed neutron source, like the spallation neutron source SNQ will depend on the fraction of the wavelength spectrum of each pulse, which can be used in time-of-flight analysis without frame overlap. It depends on

- the temperature of the moderator
- the length of the flight path
- and in a most crucial way on the frequency of neutron puls emission.

As small-angle scattering increases with the square of the wavelength, the long wavelength spectrum from a cold source would be preferred. Cold neutron pulses tend to penetrate each other after relatively short flight paths. Thermal neutrons are better in this respect. The initial pulse length of not more than about 1 ms is acceptable in most cases. A program has been written in "basic" in order to make the sums of the useful  $\lambda^2$ -weighted neutrons as a function of the parameters mentioned above.



From the user's point of view, a gain factor should be related to a problem of molecular structure research. This we do by using Eq. 2 which relates the half-width of small angle scattering to the radius of gyration of the scattering particle. It is assumed that this  $Q$ -value should be measured on the detector at 10 cm distance from the primary beam.



The GAIN FACTORS with respect to D11 of the ILL given in this figure stress even more the importance of cold neutrons. The puls structure of the SNQ can best be exploited by operating it at a fairly low frequency, say 25 Hz.

## IF SAMPLES ARE VERY SMALL.....

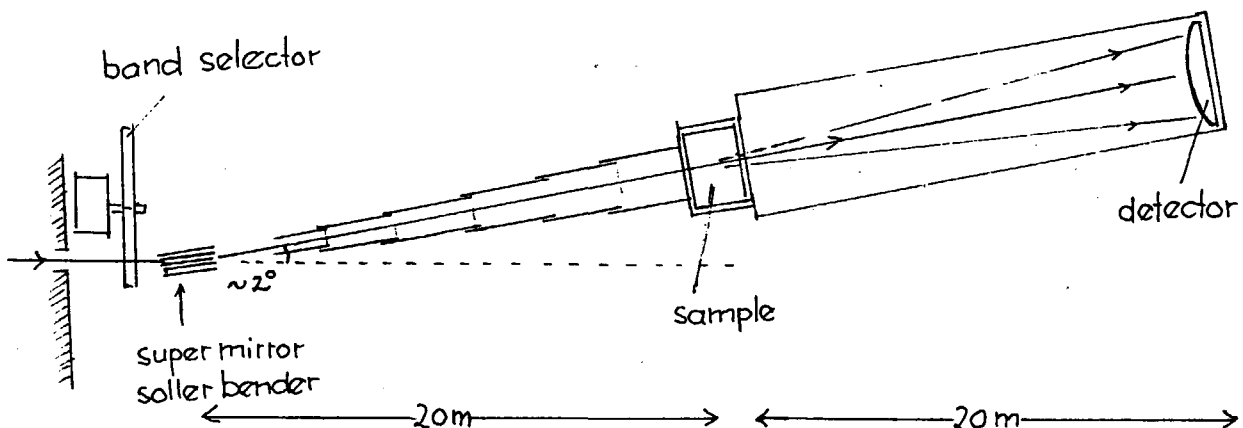
For this purpose rather compact diffractometers have been proposed. One is presently built by the European Molecular Biology Laboratory in collaboration with the ILL. Although it is now being installed at the high-flux reactor of the ILL, its place should at a pulsed source in the future. Due to the short distance between the detector and the source, an improvement by an order of magnitude is easily achieved there even if the source is run at a neutron pulse frequency of 100 Hz. A very lucid discussion of this "1m camera" has been given by B. Alefeld and J. Schelten in "SNQ Informationen" in March 1984.

## FUTURE LOW-Q SCATTERING INSTRUMENTS

We propose two instruments, both of which are similar to those presently planned by the Neutron Division of the Rutherford Appleton Laboratory.

These are a modified D11 instrument and a special low-Q diffractometer for small samples. As for the latter, a detailed description of this camera has been given by B. Alefeld and J. Schelten (1984).

A sketch of the modified D11 instrument is given below:



Neutrons from the cold source are deflected by a Super Mirror Sollér Bender. This device bends (through 40 mrad) neutrons of  $\lambda \gtrsim 2\text{\AA}$  away from the natural axis defined by the fast neutrons emerging from the primary collimator inside the shielding. This device would also act as a polarizer if required. Heavy shielding on the outside of the curve in the path of the straight through

neutrons. Following the design of the small-angle instrument of the ILL (K.Ibel, 1982), the collimation path will have a variable length from 3 to 20m.

Wavelength selection is done by a chopper at 7.5 m distance from the source. For producing very narrow bands,  $\Delta\lambda$  less than  $1 \text{ \AA}$ , of neutrons, another chopper will be put at the end of the collimation path. Both chopper are phase-locked to the SNQ.

The detector is housed in a 20m long vacuum tube where it can move freely. This design is now being adopted at various places. The detector may be a  $^3\text{He}$ -filled multi-wire chamber. A very attractive alternative is offered by  $^6\text{Li}$  loaded scintillators. 0.5mm thick lithium glass gives detection efficiencies of about 35% for  $1 \text{ \AA}$  neutrons going up to 90% at  $6 \text{ \AA}$ .

A very elegant arrangement of the scintillators in a telescopic form has been proposed by D. Cebula (RAL). With the outer rings positioned at a sample detector distance of 0.7, 2.0, 5.0 and 10m with the main detector placed at 20m a complete and continuous Q-range from 0.003 to  $1 \text{ 1/\AA}$  can easily be covered using a band from 4 to  $10 \text{ \AA}$  at the SNS pulse frequency of 50 Hz.

## CONCLUSIONS

Time of flight analysis of low-Q scattering is a very flexible technique, which can most easily match the experimental requirements, even at steady state reactors. This is so, as this technique lends itself easily to efficient control of the measurement by microprocessors. There is no reduction of measuring time at this stage. With neutrons from a pulsed spallation source the power of time-of-flight analysis will fully develop.

The analysis of the gain factor with respect to the small-angle instrument D11 at the high-flux reactor of the Institute Laue-Langevin shows that a cold neutron source emitting pulses of not more than 1ms duration each 40 ms (25 Hz) would provide the best conditions for low-Q experiments. The gain factor would be about one order of magnitude for a very wide class of structures, ranging from rather small ones, like enzymes to subcellular dimensions. At the presently for other reasons favoured 100 Hz pulse frequency of the SNQ, similar gain factors would be restricted to investigations of smaller structures with dimensions below  $50 \text{ \AA}$ .

Low-Q scattering from fibres ( muscles, connective tissue ), isometric structures ( viruses, phages ), copolymers etc. may show a much more structured scattering pattern, which would need a resolution in Q of a few percent. Time-of-flight analysis meets this requirement easily. An eventual loss of intensity may be due to a decrease of the divergence of the incident beam only.

The initial lenght of the neutron puls of 0.5ms at 100 Hz and 1 ms at 50 Hz is acceptable. Further pulse compression would improve the measurements at higher momentum transfer Q.

The deconvolution of low-Q scattering affected by frame overlap has not yet been succesful. A further gain with respect to D11 could be obtained in this way.

#### References

B. Alefeld and J. Schelten (1984) "Two Neutron Scattering Instruments for a Source with Time Structure", SNQ - Informationen, 2, 11-14, KFA Jülich

D.J. Cebula (1983) "The Small-Angle Neutron Scattering Instrument At The Rutherford Appleton Laboratory SNS" , NDR/PB/83

J.P. Cotton (1984) "Monochromation par temps de vol en diffusion aux petits angles", Laboratoire Léon Brillouin /84/758/JPC/CD, C.E.N. Saclay

K. Ibel (1982) "Neutron Small Angle Diffractometer. A Universal Camera at the Second Cold Source", 81IB07S, Internal Report of the ILL

K.H. Nierhaus (1984) "Anwendungen der Neutronenstreuung in der molekular-biologischen Forschung", SNQ-Informationen, 3, 7-11, KFA Jülich

## SMALL-ANGLE SCATTERING INSTRUMENT FOR BIOLOGICAL SAMPLES

B. Alefeld, H. Conrad, SNQ-KFA, Jülich

### (1) The concept

The instrument is meant for small angle scattering studies with small samples for a wavelength resolution of about 10%; for instance for studies on the morphology of biological molecules or cell components. The instrument is conceived such that it accepts a very wide band of incident wavelengths (1 to 8 Å) and utilizes the pulse structure of the neutron source to achieve the said resolution. This is illustrated in fig. 5. In this way, the instrument uses the peak flux almost continuously.

A Q-range  $0.01 \leq Q \leq 1 \text{ Å}^{-1}$  and a resolution  $\Delta Q = 0.012 \text{ Å}^{-1}$  demand the following dimensions:

- Distance moderator-sample L1: 7 m
- Maximum distance sample - detector L2: 2 m
- (with L2 = 2 instead of 1 m, the instrument is better matched).
- Anger camera with about 2 mm spatial resolution and an area of about  $60 \times 60 \text{ cm}^2$ , movable in the range of L2 = 0.3 m to L2 = 2 m.
- Two diaphragms
- Chopper 2 m from the moderator
- Small samples, some  $\text{mm}^2$  area

The best moderator for the instrument would be the one at 90 K.

The following options are foreseen, but require still further considerations and test measurements:

- For suppression of frame overlap, replacement of the chopper by adapted totally reflecting mirrors.
- Use of a pepper-pot collimator, if larger samples are available.
- Use of diaphragms dynamically adjustable to the resolution required for different wavelengths.

- Use of a bent neutron guide between sample and moderator to enhance the intensity and to avoid direct view of the moderator.

## (2) Count rate and resolution

We compare an instrument with diaphragms to one with a neutron guide.

### (a) Instrument with diaphragms

The full line in Fig. 1 shows the count rate in the entire detector for a 10 % isotropic scatterer as a function of flight time, that is: wavelength.

The diaphragms are:

|                         |                                           |
|-------------------------|-------------------------------------------|
| In front of the sample: | 1.5 mm radius<br>(4 mm <sup>2</sup> area) |
|-------------------------|-------------------------------------------|

|                                       |                            |
|---------------------------------------|----------------------------|
| In 1.5 m distance from the moderator: | 4.2 mm radius<br>(matched) |
|---------------------------------------|----------------------------|

|                  |      |
|------------------|------|
| The length L2 is | 2 m. |
|------------------|------|

The count rate is sharply peaked around 2 Å. For the calculated incident neutron spectrum, the averaged value is ~10 neutrons/sec.

Dividing this count rate by  $/K/$  one arrives at the dashed line (arbitrary units). This takes into account that for a given  $/Q/$  the solid angle is larger for smaller  $K$ 's, (if integrating over the azimuthal angle).

Fig. 2 shows the  $Q$ -resolution  $\Delta Q$  as a function of flight time:

$$\Delta Q = K \cdot \Delta \vartheta$$

The  $\Delta \vartheta$ -resolution (FWHM) for the scattering angle is calculated from /P.A. Seeger, Nucl. Instr. and Methods 178 (1980) 157/. The contribution of the pulse width  $\Delta t$  to  $\Delta Q = Q \cdot \Delta t / t$  can be neglected for the smaller  $Q$ 's of interest here.

The dashed line for the minimum  $Q$  is calculated from geometry with an additional margin of 25%.

### (b) Instrument with neutron guide

Figs. 3 and 4 show the corresponding curves for a fully illuminated glass neutron guide in front of the sample, with the instrument unchanged behind



the sample (not matched resolution).

The count rate is dramatically enhanced, but the resolution is dramatically worse.

The resolution is independent of  $K$  and is due to the large vertically reflected component for glass of  $K_{\perp} = 0.0063 \text{ \AA}^{-1}$ . (For Ni ( $K_{\perp} = 0.0107$ ) this would be even worse.) Considerable improvement of the resolution is possible, if  $k_{\perp}$  can be reduced, for instance either by a trumpet-like shape of the neutron guide or by depositing two layers of materials onto the glass surface, one layer for absorption of neutrons, for example Gd, and one for total reflection, for example Al.

$Q_{\text{MIN}}$  is calculated for the largest reflecting angle of the neutron guide ( $0.46^\circ$  for 8 Å neutrons). The time structure may permit to measure at smaller angles and thus to reduce  $Q_{\text{MIN}}$  for the smaller wavelengths.

An adapted totally reflecting mirror for suppression of frame overlap would be most effective, if its  $K_{\perp}$  component is larger than the one for the neutron guide.

Fig.1

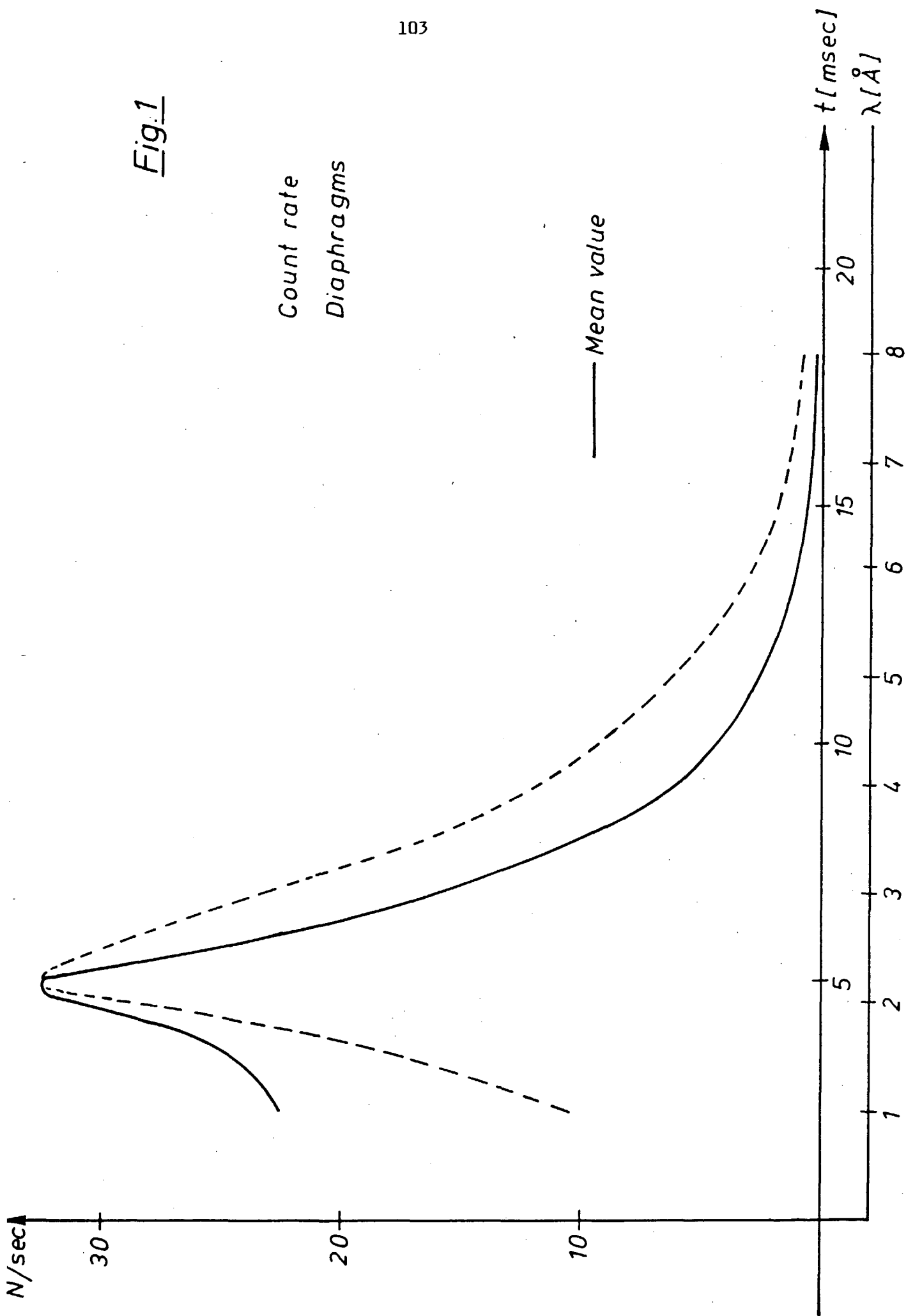


Fig. 2

Resolution  
Diaphragms

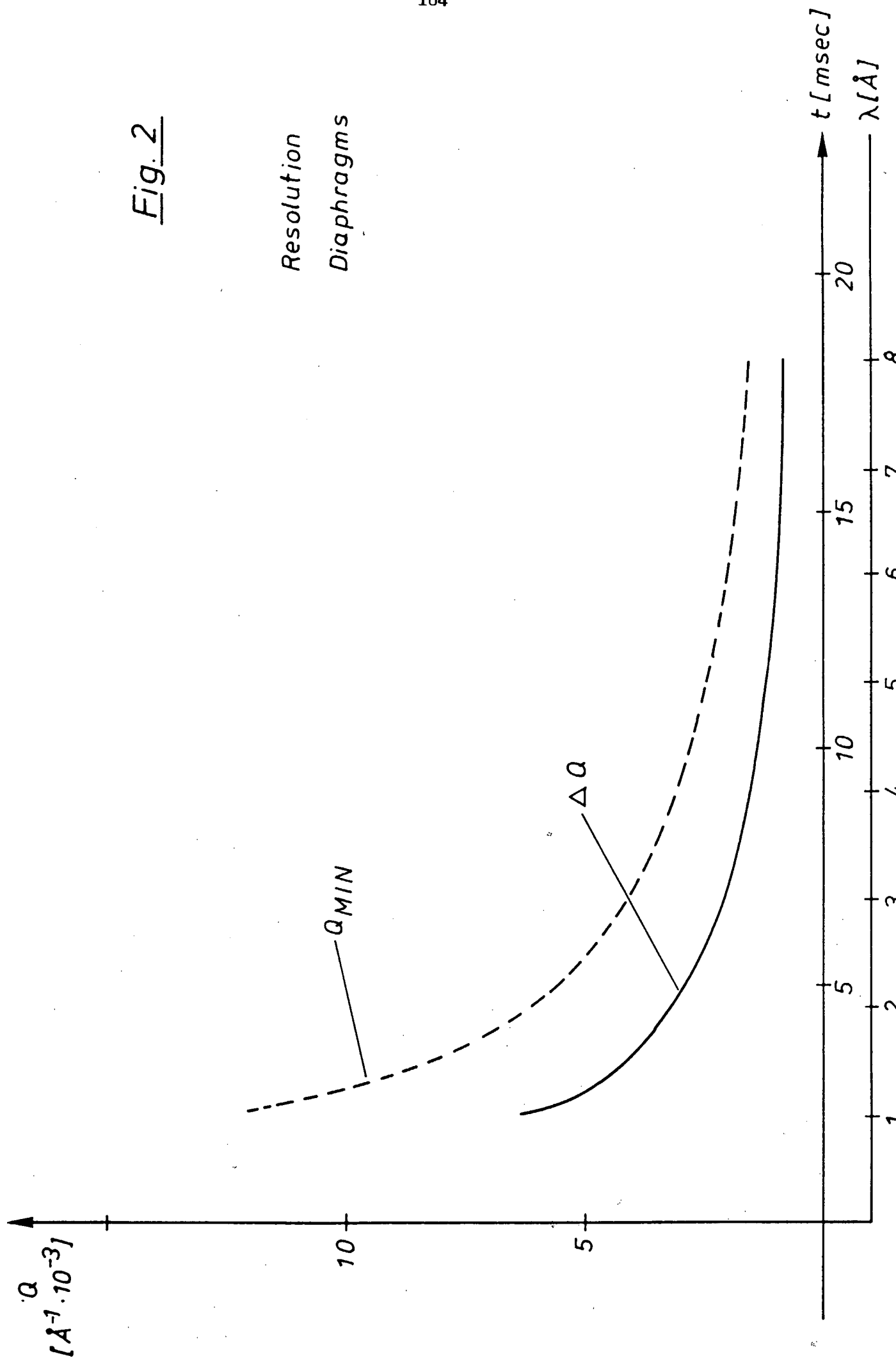
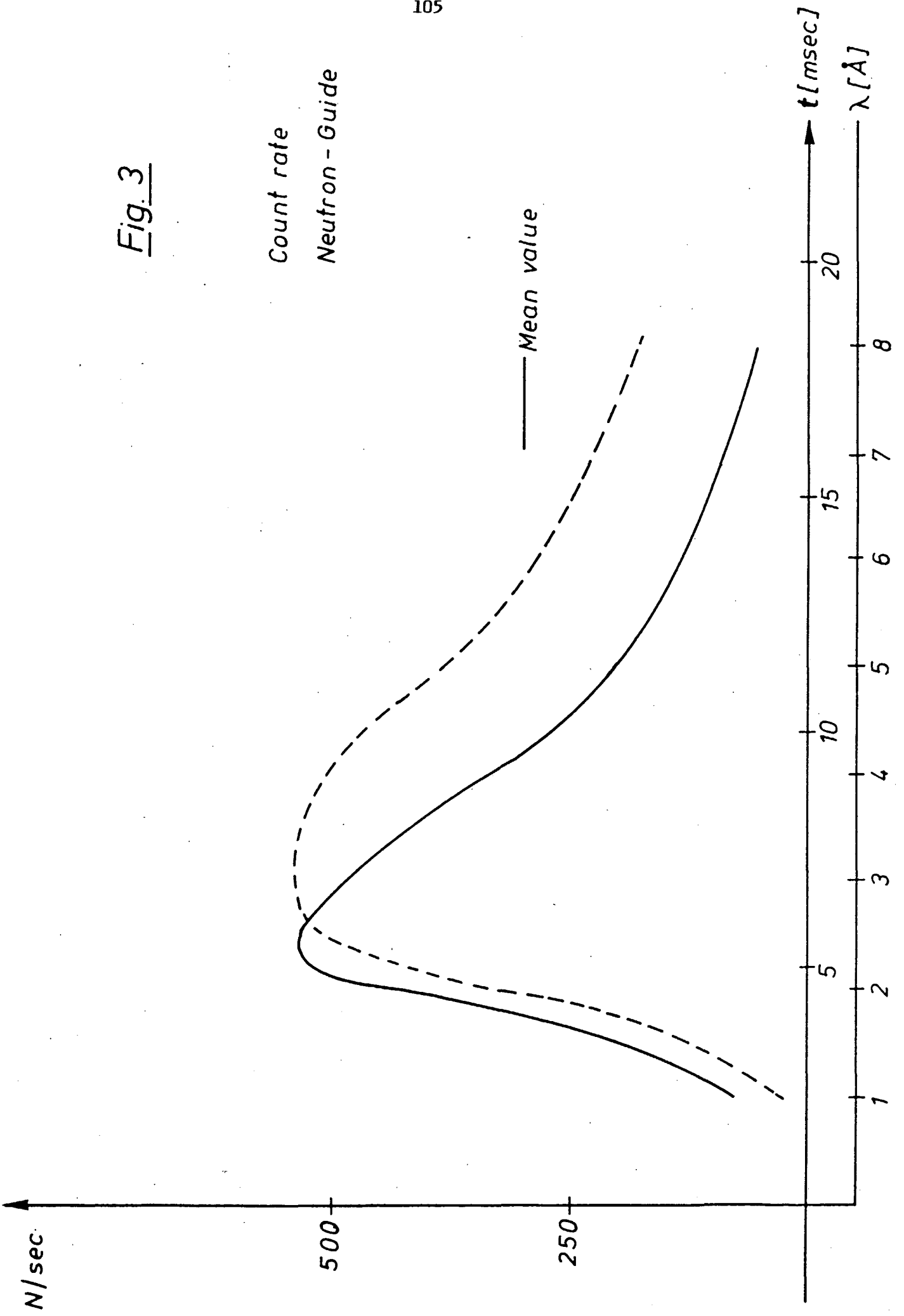


Fig. 3

Count rate  
Neutron - Guide

— Mean value



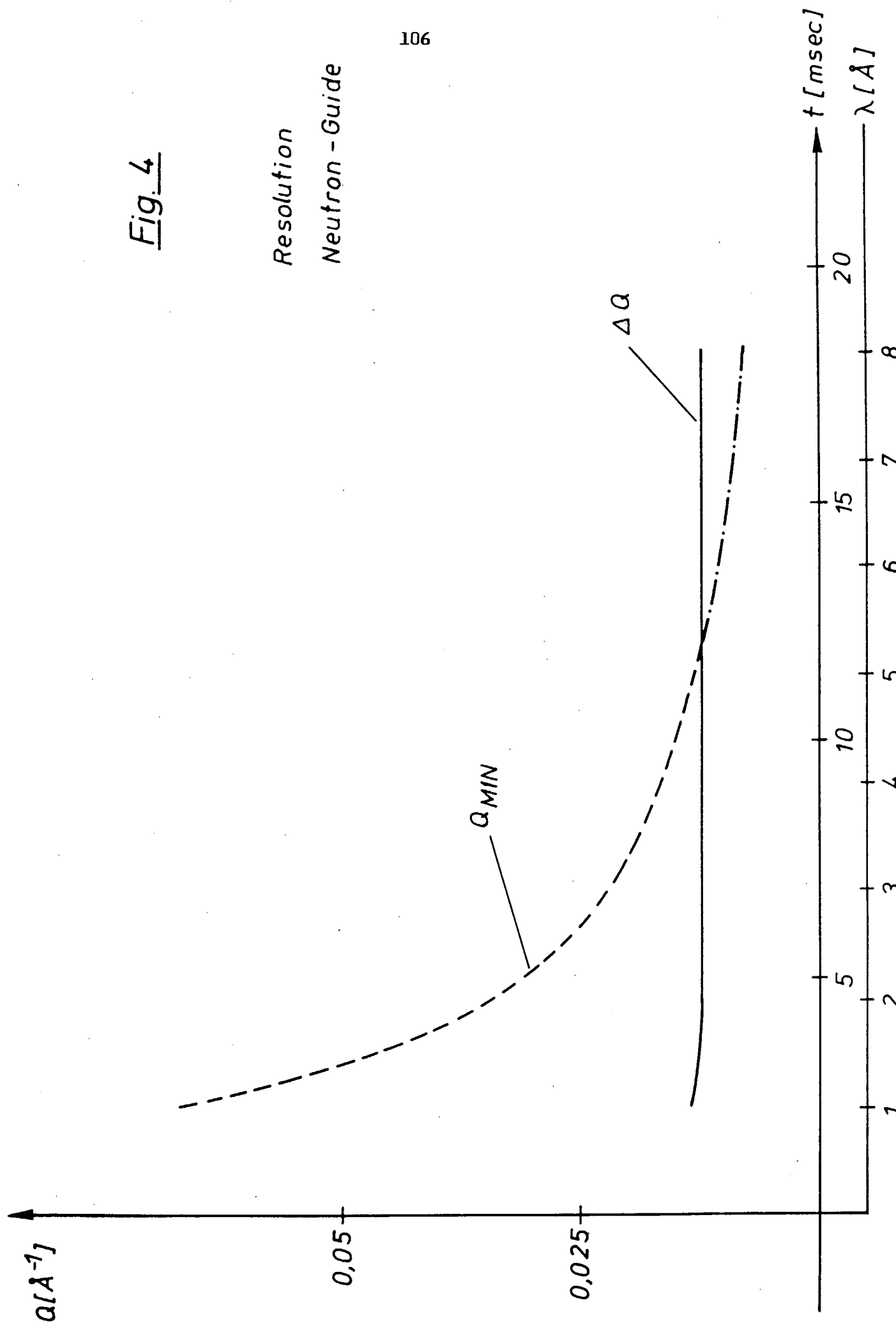


Fig. 4

Resolution  
Neutron - Guide

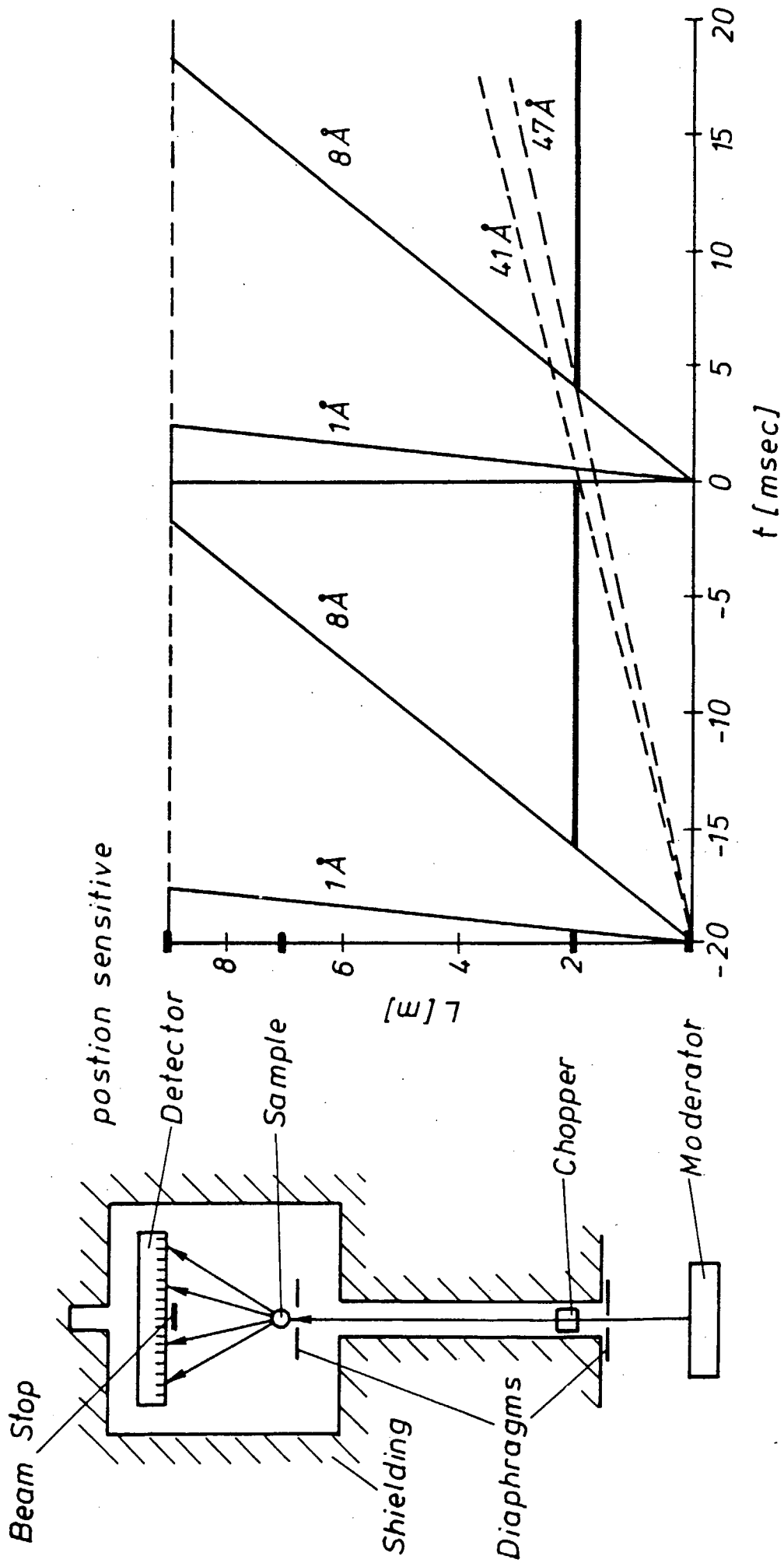


Fig. 5

SURFACE STUDIES WITH NEUTRONS

B. Farnoux

Laboratoire Léon Brillouin, CEN-Saclay, France

A. Steyerl

Physik-Department E21 der Techn. Universität, München, Germany

J. Deutz, B. Alefeld

Kernforschungsanlage, 5170 Jülich, Germany

ABSTRACT

Neutron Scattering as a possible tool for surface structure studies in different fields is described and some existing activities are mentioned. In the second part experimental methods are discussed in some detail, especially in view of a pulsed neutron source. For low resolution studies with conventional geometry, the peak flux of the source can be exploited. Gain factors for the gravity refractometer were calculated. The gain becomes essential only for high resolution experiments.

## 1. General aspects

There are numerous subjects for surface studies by slow neutrons, and we may quote the following partial list:

- Physical or chemical adsorption:
  - adsorption of gases on metallic surfaces
  - variation in the density of a liquid at a liquid-gas or liquid-liquid interface
  - conformation of adsorbed polymers
  - Langmuir-Blodgett layers
  - wetting liquid-vapour interface of binary liquid mixtures
  - oxidation phenomena of metallic surfaces and semiconductors
  - electrochemistry of electrodes
  - critical phenomena in two dimensions
  - in biology, adsorption of proteins of blood on surfaces of polymers or metals
  - etc. ...

All these subjects are studied by different techniques (light, x-rays particularly), but neutrons can offer some advantage (especially as far as surface magnetism is concerned) due to their particular interaction (isotope substitution, nuclear and magnetic interaction).

These studies concern the determination of concentration profiles near a surface by measuring the reflectivity curve around the and beyond the angle of total reflection. There have already been recent experiments, e.g.:

- critical reflection of neutrons by Hayter et al. /1/, who give a good review of the possibilities of the technique.

More precise experiments have been carried out by H. Scheckenhofer and A. Steyerl /2/ and by G.P. Felcher et al. /3/, who describe an experiment at the pulsed source in Argonne to measure the penetration depth of the magnetic field in superconductors. This is a good example for the interest in neutrons and pulsed sources.

At Saclay an experimental program has been set up to determine the concentration profile of polymers adsorbed on metallic surfaces. During the test



measurements a time of flight method has been studied which simulates an analogous experiment at a pulsed source. This type of technique is perfectly suited to measurements on liquid interfaces which cannot be inclined towards the beam to study the variation of scattered intensity as a function of angle.

The principle of the measurement is the following:

The direct beam coming out of the neutron guides is precisely collimated by two horizontal slits (of  $1 \times 10 \text{ mm}^2$ ) situated at a mutual distance of about 2 m. A chopper, placed behind the second slit, produces the neutron pulses which are reflected by the mirror to be studied. The detector is placed at a distance of 5 m to get a good angular resolution. The rotation frequency of the chopper is 30 Hz for the actual distribution of incident neutron wavelengths. First results have already been obtained.

As far as "real time low-Q scattering" is concerned, one of the interesting problems would be to study the variation (as a function of time) of the dimensions of polymer chains under stress.

In a classical steady state low angle scattering experiment the measuring times are some times too long with respect to characteristic times of the sample. The characteristic disentanglement time of a polymer chain in the solid state is about  $6 \cdot 10^5$  seconds. Other characteristic times are the conformation reorganization time  $T_R$  and the reptation relaxation time  $T_{Rep}$ . At 128°C e.g. the variation of these times is

$$\begin{aligned} 150 \text{ sec} < T_R &< 1500 \text{ sec} \\ 1.5 \cdot 10^4 \text{ sec} < T_{Rep} &< 1.5 \cdot 10^5 \text{ sec} \end{aligned}$$

With a pulsed source one can envisage to do these measurements "in situ": using the incident spectrum - limited by choppers synchronized with the frequency of the source- one can register the variation of intensity as a function of time. During these measurements the sample can be kept under constant tension, after elongation by a given value; or else one can let the sample relax and register the scattered intensity as a function of time.

Another type of measurement is the study of polymers under shear stress in a flow. One can synchronize the speed of the flow with the frequency

of the source by integrating the scattered intensity over a certain number of cycles.

Other measurements of conformation can also be made, for example the study of conformation in a critical mixture.

## 2. Experimental techniques

### 2.1 Intensity and resolution

An idealized surface reflection experiment is shown in Fig. 1

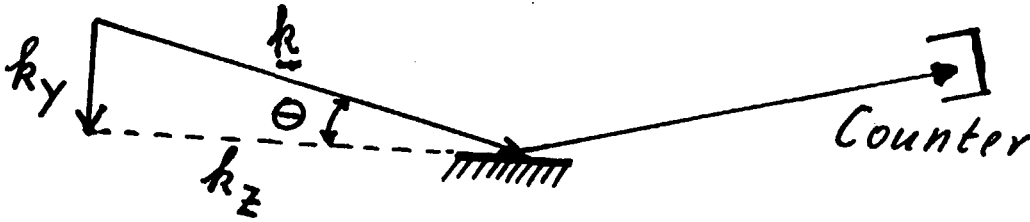


Fig. 1 Schematic surface reflection. A monochromatic and ideally collimated neutron beam is reflected by the surface and detected in the counter.

A monochromatic, ideally collimated neutron beam with a wave vector  $k$  hits the surface of the sample,  $\theta$  is the angle between the beam and the surface. Physical information on the surface is obtained from the intensity and direction of the reflected beam. The most important information on the surface frequently is obtained near and beyond the total reflecting edge of the reflecting curve. This edge is given by

$$k_y^m = 2 \cdot \sqrt{\pi N b_{\text{coh}}} \quad (1)$$

$N$ : Number density of atoms,  $b_{\text{coh}}$ : coherent scattering length.  $k_y^m$  depends only on the scattering density and is typically of the order of  $0.005 \text{ \AA}^{-1}$ . In this sense a surface reflection experiment is very similar to a small angle scattering experiment with  $Q = 2 k_y$

$$\begin{aligned} k_y &= k_z \cdot \theta \\ (\sin \theta &\approx \theta) \end{aligned} \quad (2)$$

As always in a realistic experimental situation, a compromise has to be made between resolution and intensity. The resolution of  $k_y$  is given by the uncertainty of the two experimental parameters  $k_z$  and  $\theta$

$$\frac{\Delta k_y}{k_y} = \frac{\Delta k_z}{k_z} + \frac{\Delta \theta}{\theta} \quad (3)$$

In the case of surface reflection a slit geometry is appropriate, and it can easily be shown that  $\Delta k_x$  does not influence the resolution. (Slit in x-direction). The intensity of the beam hitting the surface of the sample is given by

$$\Delta J = \frac{\phi}{2\pi} \frac{k_z^4}{k_T^4} e^{-\frac{k_z^2}{k_T^2}} \frac{\Delta k_x}{k_z} \frac{\Delta k_y}{k_z} \frac{\Delta k_z}{k_z} \cdot F \quad (4)$$

$\frac{\Delta k_x}{k_z}$  : horizontal collimation

$\frac{\Delta k_y}{k_z}$  : vertical collimation

$\frac{\Delta k_z}{k_z}$  : wavelength resolution

F: Area of the slit in front of the sample. If a resolution  $R = \frac{\Delta k_y}{k_y}$  is desired for the experiment the two contributions  $\Delta k_x/k_z$  and  $\Delta \theta/\theta$  have to be matched for intensity reasons. It follows

$$1/2 R = \frac{\Delta k_z}{k_z} = \frac{\Delta \theta}{\theta} \quad (5)$$

$$J = \frac{\phi}{2\pi} \frac{k_z^4}{k_T^4} e^{-\frac{k_z^2}{k_T^2}} \cdot \frac{1}{2} R^2 \frac{\Delta k_x}{k_z} \frac{k_y}{k_z} \cdot F \quad (6)$$

### 2.1.1 Scanning modes.

The reflection curve of the sample can be scanned in two different ways.

In equ. (2) ( $k_y = k_z \cdot \theta$ ) either  $k_z$  is changed keeping the angle constant or  $k_z$  is kept constant and the reflection angle  $\theta$  is changed. The intensity is equivalent for both methods. This also holds for the time of flight method, which consists in cutting the primary beam into short pulses with a wavelength spread given by the frame overlap condition. This can be shown in the following way. There are two important quantities in a time of flight experiment, the resolution and the available wavelength band between two pulses.

The time of flight resolution is given by

$$\frac{\Delta k_z}{k_z} = \frac{t_n}{t_{fl}} = \frac{t_n}{L} \cdot v \quad (7)$$

$t_n$ : neutron pulse length

$L$ : flight path

$v$ : neutron velocity.

The wavelength band between two pulses is given by

$$\left( \Delta \frac{k_z}{k_z} \right)_b = \frac{T}{L} v \quad (8)$$

$T$ : time between two pulses.

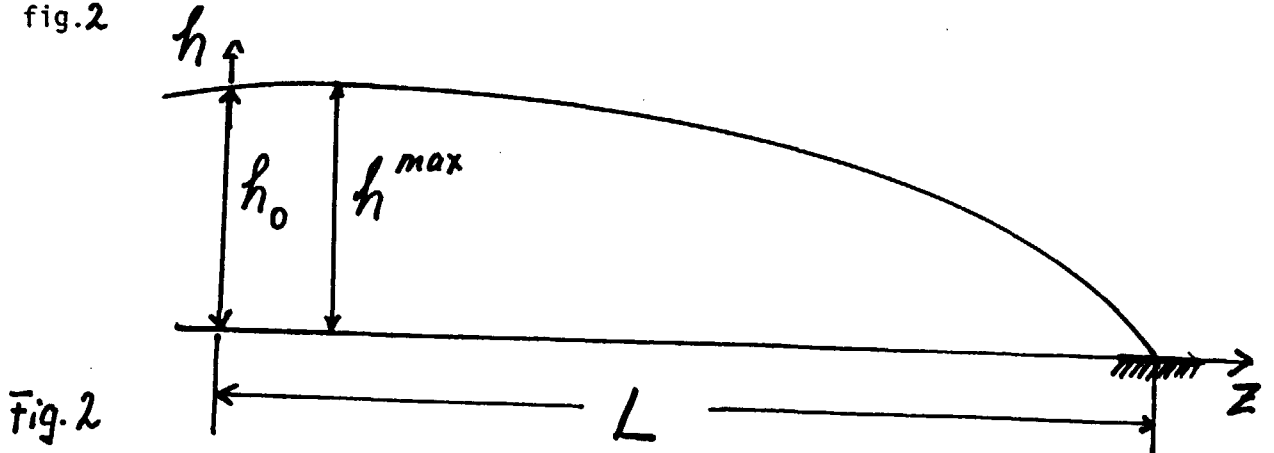
The number of resolution elements within the wavelength band is given by the ratio between equ. (8) and equ. (7) and is  $T/t_n$ . This is exactly the inverse ratio, by which the intensity of one resolution element is reduced by chopping the beam.

Assuming that the spectral density is constant within the wavelength band, the reduction of intensity by the factor  $t_n/T$  due to chopping the beam is exactly compensated by the available number  $T/t_n$  of neighbouring resolution elements. At the projected pulsed neutron source  $T = 0.01$  sec and  $t_n$  at the cold source will be about 500  $\mu$ sec, so the ratio  $T/t_n$  is 20. The scanning over twenty resolution elements seems to be reasonable for surface reflection studies. It follows that this factor is gained compared to a steady state source with the same average flux.

### 3. The Gravity Refractometer

At the Munich research reactor a very sophisticated instrument for surface studies was built [4,5]. It has mainly been used for high precision measurements of coherent scattering lengths by measuring  $k_y^m$ . A momentum space transformation due to the influence of the gravity force is used to obtain a focusing effect of  $k_y$ . Due to this transformation the uncertainty of  $k_y$  is independent of wavelength in first order. An intensity gain of two orders of magnitude is obtained compared to a simple experiment without gravity focusing.

The intensity gain is mainly due to the fact that a rather wide wavelength band of the order of 10 % can be used without deteriorating the resolution which is about 0.3 % at the Munich instrument. In the following the resolution and intensity of the gravity refractometer is studied in connection with a pulsed neutron source. The principle of the instrument is shown in fig.2



Neutrons pass a horizontal slit and move on a parabola due to the force of gravity. The coordinate  $h$  is given by

$$h = -\frac{1}{2}gt^2 + v_y t + h_0 \quad (9)$$

$v_y$  and  $h_0$  are the vertical velocity component and space coordinate at the slit.

A neutron arrives at  $h = 0$  at time

$$T = \frac{v_y}{g} + \left( \frac{v_y^2}{g^2} + \frac{2h_0}{g} \right)^{\frac{1}{2}} \quad (10)$$

To hit a surface at  $h = 0$  at a distance  $L$  from the slit the horizontal velocity component has to be

$$v_z = \frac{L}{\frac{v_y}{g} + \left( \frac{v_y^2}{g^2} + \frac{2h_0}{g} \right)^{\frac{1}{2}}} \quad (11)$$

The perpendicular  $k$ -component  $k_y$  at the mirror is given by the fall height  $h^{\max}$  from the top of the parabola to the mirror

$$mgh^{\max} = \frac{\hbar^2 k_y^2}{2m} \quad (12)$$

$h^{\max}$  can be derived from equ. (9)

$$\frac{dh}{dt} = 0 = -gt^{\max} + v_y$$

$$t^{\max} = \frac{v_y}{g} \quad (13)$$

Inserting this value into equ. (9) gives

$$\frac{h^{\max} - h_0}{h_0} = \frac{v_y^2}{2gh_0} \quad (14)$$

It follows that the instrument has two contributions to the resolution: one is due to the finite width of the slit, the other one is due to the divergence of the beam which is proportional to  $v_y$ .

For intensity reasons both contributions should be equal and each half the resolution,

$$\frac{v_y^2}{2gh_0} < \frac{1}{2} R \quad \text{or}$$

$$\left| \frac{v_y}{g} \right| < \sqrt{\frac{Rh_0}{g}} \quad (15)$$

Inserting these values into equ. (11) the velocity range which is tolerable for the resolution  $R$ , can be calculated.

Table 1 shows calculations for an instrument of 100 m length and for different resolution  $R$ . The first column shows the  $k_y$ -value to be measured. The second column shows the corresponding fall height of a neutron, equ. (12). The next column shows the mean velocity  $\bar{v}_z = L/\sqrt{2h_0/g}$  equ. (11) with  $v_y = 0$ . In the next three columns the tolerable wavelength resolution for different resolutions of the instrument is given. The last column shows the available wavelengthband of a pulsed source with the SNQ-parameters ( $T = 0.01$  sec, time between two pulses, calculated by  $(\Delta v/v)_b = \frac{T}{L} \bar{v}_z$ ). Table 2 gives the gain factors of a pulsed source compared to a steady state source with the same average flux. This gain factor is the ratio between the available wavelength band between two pulses and the tolerable wavelength band for the desired resolution.

Table 1

| $k_y (10^{-3} \text{ Å}^{-1})$ | $h_0/\text{cm/}$ | $\bar{v}_z (\text{m/sec})$ | $\Delta v_z / \bar{v}_z (\%)$ |               |               | $(\Delta v/v)_D$ |
|--------------------------------|------------------|----------------------------|-------------------------------|---------------|---------------|------------------|
|                                |                  |                            | $R = 10^{-3}$                 | $R = 10^{-4}$ | $R = 10^{-5}$ |                  |
| 1                              | 2                | 1559                       | 4,5 %                         | 1,4 %         | 0.45 %        | 15,6 %           |
| 2                              | 8,1              | 797                        | "                             | "             | "             | 7,8              |
| 3                              | 18,2             | 519                        | "                             | "             | "             | 5,19             |
| 4                              | 32,3             | 389                        | "                             | "             | "             | 3,9              |
| 5                              | 50,5             | 312                        | "                             | "             | "             | 3,11             |
| 6                              | 72,6             | 260                        | "                             | "             | "             | 2,6              |
| 7                              | 98,9             | 223                        | "                             | "             | "             | 2,23             |
| 8                              | 129,2            | 195                        | "                             | "             | "             | 1,95             |
| 9                              | 163,5            | 173                        | "                             | "             | "             | 1,73             |
| 10                             | 201,8            | 156                        | "                             | "             | "             | 1,56             |
| 11                             | 244,2            | 142                        | "                             | "             | "             | 1,42             |
| 12                             | 290,6            | 129                        | "                             | "             | "             | 1,29             |

Table 2

| $k_y (10^{-3} \text{ Å}^{-1})$ | G             |               |               |
|--------------------------------|---------------|---------------|---------------|
|                                | $R = 10^{-3}$ | $R = 10^{-4}$ | $R = 10^{-5}$ |
| 1                              | 3,5           | 11            | (35) 20       |
| 2                              | 1,74          | 5,5           | 17            |
| 3                              | 1,16          | 3,67          | 11,8          |
| 4                              |               | 2,75          | 8,7           |
| 5                              |               | 2,2           | 7             |
| 6                              |               | 1,8           | 5,8           |
| 7                              |               | 1,57          | 4,3           |
| 8                              |               | 1,37          | 3,9           |
| 9                              |               |               | 3,75          |
| 10                             |               |               | 3,2           |
| 11                             |               |               | 2,9           |
| 12                             |               |               | 2,7           |

#### 4. Conclusion

If only moderate resolution is desired, surface studies with neutrons are best performed with a monochromatic and collimated neutron beam with slit geometry. From the point of view of intensity the steady state method and the time of flight method are equivalent, however for practical reasons the time of flight method seems to have advantages. (Scanning of the reflection curve during a short time and averaging long time instabilities). In general at least 20 resolution elements can be used to measure the reflection curve. For this reason a properly designed instrument at a pulsed source can exploit the peak flux during the whole time. In contrast, the high resolution gravity refractometer which inherently uses a rather wide wavelength band of the order of  $\sqrt{R}$ , (a simple instrument uses a wavelength band of  $\Delta k_z/k_z = R/2$ ) will gain from pulsing the source for a resolution only of the order of  $10^{-4}$  or better and for small fall heights.

This indicates that investigations of thin surface structures requiring large fall heights will hardly profit from a pulsed source.

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Spektrometer für diffuse Streuung  
ohne Energieanalyse an der gepulsten Quelle

J. Schelten und G. Bauer

KFA Jülich

1. Einleitung

Mit dem Spektrometer wird die elastisch diffuse Streuung von Legierungen, teildeutierten Polymeren von amorphen Substanzen, von paramagnetischen Substanzen, etc. gemessen. Aus den Messungen wird Information über die Unordnung, den Nahordnungsparameter, die Fernordnung, Überstrukturen, Konformationen von Polymermolekülen, magnetische Formfaktoren, etc. gewonnen. Es wird unterstellt, daß die Streuung zwischen den Bragg-Reflexen hauptsächlich elastischer Natur ist, was bei hoher Defektkonzentration der Fall ist.

Mit dem fokussierenden Einkristalldiffraktometer T4 am BER II wird dieses Arbeitsgebiet gepflegt. Ziel dieses Berichtes ist es, ein Spektrometer an der gepulsten SNQ-Neutronenquelle ( $\Delta T = 10$  ms,  $\Delta t = 250$   $\mu$ s) zu beschreiben, mit dem man die Pulsstruktur nutzen kann und die Spezifikationen des Diffraktometers T4 erreicht.

2. Prinzipieller Aufbau

Die gepulste Quelle legt es nahe, im Flugzeitbetrieb die Neutronen zu detektieren, wobei die Ankunftszeit am Detektor die Neutronenwellenlänge und mit dem Streuwinkel  $\theta$  den Q-Vektor festlegt. Einen Intensitätsgewinn kann man sich davon versprechen, daß nacheinander mehrere Wellenlängenbänder genutzt werden.

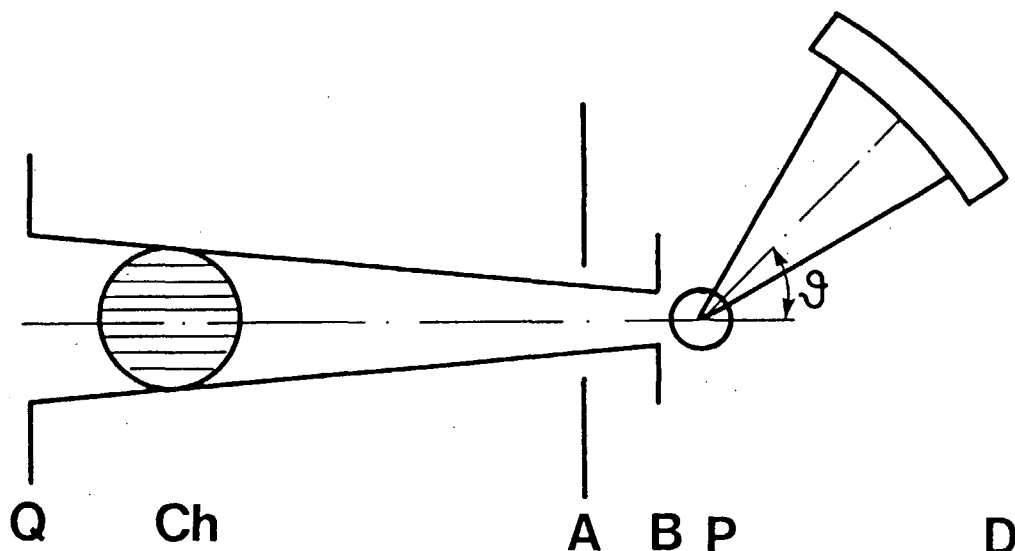


Fig. 1 Schematischer Aufbau des Spektrometers.

Q - Quelle, Ch - Chopper, A - biol. Abschirmung,  
B - Blende, P - Pröbe, D - Bananen-Detektor.

Abstände: Q - Ch :  $l \approx 1$  m; Q - D :  $L \approx 10$  m,  
P - D : 1.50 m.

In Fig. 1 ist die Apparatur skizziert. Sie hat keinen Monochromatorkristall und keine Fokussierungsvorrichtung. Ein Chopper verkürzt den Neutronenpuls von  $\Delta t = 250 \mu\text{s}$  auf  $\delta t$ . Die Wellenlängenauflösung ist damit

$$(1) \quad \frac{\delta\lambda}{\lambda_M} = \frac{\delta t}{T_M}$$

mit

$$(1') \quad T_M = \frac{L}{v_M}$$

Dabei ist  $T_M$  die Neutronenlaufzeit zum Detektor,  $v_M$  und  $\lambda_M$  sind die mittlere Geschwindigkeit und Wellenlänge des schmalen Bandes  $\delta\lambda/\lambda_M$ .

Die angestrebte und den anderen Auflösungsbeiträgen angepaßte Wellenlängenauflösung ist  $\frac{\delta\lambda}{\lambda} = 1\%$ . Dies bestimmt die Chopper-Öffnungszeit, die in Tabelle 1 angegeben ist. Die Öffnungszeiten sind kein technisches Problem.

Tabelle 1

| $\lambda_M$<br>(Å) | $T_M$<br>(ms) | $\delta_t$<br>(μs) | $\frac{\delta\lambda}{\lambda}$<br>(%) | $\frac{\Delta\lambda}{\lambda}$<br>l = 1 m |
|--------------------|---------------|--------------------|----------------------------------------|--------------------------------------------|
| 1                  | 2.5           | 25                 | 1                                      | 1                                          |
| 2                  | 5             | 50                 | 1                                      | 0.5                                        |
| 4                  | 10            | 100                | 1                                      | 0.25                                       |

Der Spektrometeraufbau nach der Probe ist identisch dem des Referenz-Diffraktometers.

### 3. Chopper

Die Position des Choppers bestimmt entscheidend, wie breit das Wellenlängenband  $\Delta\lambda/\lambda$  ist, das genutzt werden kann. Dies verdeutlicht das Wegzeitdiagramm für  $\lambda = 2 \text{ Å}$  und für  $l = 1 \text{ m}$  Chopperentfernung von der Quelle. Das durchgelassene Wellenlängenband hat die Halbwertsbreite  $\Delta\lambda = \lambda_1 - \lambda_2$ . Mit

$$\lambda_1 = \lambda_M \frac{t_M + \Delta t/2}{t_M} \quad \text{und} \quad t_M = \frac{l}{v_M} \quad \text{ergibt sich}$$

$$(2) \quad \Delta\lambda/\lambda = \frac{\Delta t}{t_M}$$

Werte für Durchlaßbänder finden sich in Tabelle 1. Sie sind groß bei kurzen Wellenlängen.

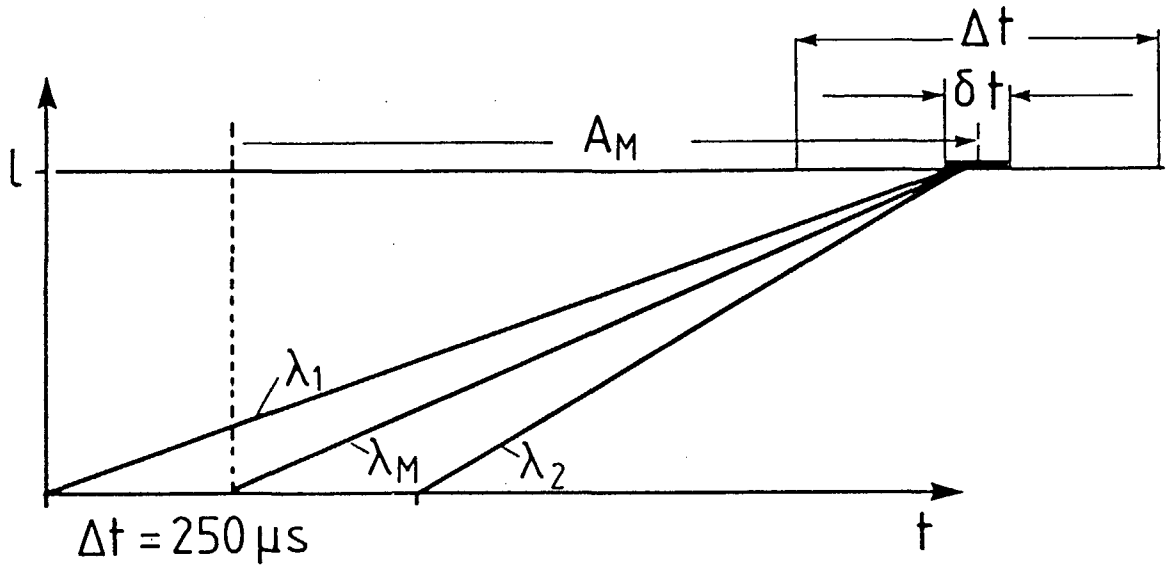


Fig. 2 Weg-Zeit-Diagramm und Chopper-Durchlaß-Wellenlängenbereich  $\lambda_1 - \lambda_2$  für  $\lambda_M = 2 \text{ \AA}$ ,  $l = 1 \text{ m}$

#### 4. Q-Auflösung

Die Q-Auflösung wird entsprechend (3)

$$(3) \quad \frac{\delta Q}{Q} = \frac{\delta \lambda}{\lambda} + \epsilon \tan(\vartheta/2) \cdot \frac{\delta \vartheta}{2}$$

von zwei Beiträgen bestimmt.

Dabei ist

$$(4) \quad \delta \vartheta = \delta \alpha_h + \delta \beta_h$$

gegeben durch die horizontalen Winkeldivergenzen vor und nach der Streuung. Sie sind aufgrund von Quellgröße und Probengröße

$$(5) \quad \Delta \alpha_h = \frac{10 \text{ cm}}{8.5 \text{ m}} = 1.2 \cdot 10^{-2} \text{ rad}$$

$$\Delta \beta_h = \frac{1 \text{ cm}}{1.5 \text{ m}} = 0.7 \cdot 10^{-2} \text{ rad}$$

Mit diesen Werten zeigt sich, daß für den mittleren Streuwinkel von  $\vartheta = 90^\circ$  ein  $\frac{\delta\lambda}{\lambda} = 1\%$ , wie ausgelegt, angepaßt ist. Bei kleineren Streuwinkeln dürfte  $\delta\lambda/\lambda$  größer und bei größeren Streuwinkeln kleiner sein.

## 5. Intensitätsvergleich

Für das Referenzdiffraktometer ist der Neutronenstrom an der Probe gegeben durch

$$(6) \quad I' = \bar{\vartheta}' \cdot R_K' \cdot \delta\alpha'_h \cdot \delta\alpha'_v \cdot \frac{\delta\lambda'}{\lambda}$$

Es nutzt ständig den kontinuierlichen Fluß  $\bar{\vartheta}'$ . Der Monochromatorkristall habe die Reflektivität  $R_K'$ .

Für das Spektrometer an der gepulsten Quelle gilt

$$(7) \quad I = \bar{\vartheta} \cdot T_{Ch} \cdot \delta\alpha_h \cdot \delta\alpha_v \cdot \frac{\delta\lambda}{\lambda} \cdot \left(\frac{\delta t}{\Delta t}\right) \cdot \left(\frac{\frac{\Delta\lambda}{\lambda_M}}{\frac{\delta\lambda}{\lambda_M}}\right)$$

Dabei ist  $T_{Ch}$  die Choppertransmission,  $\delta t/\Delta t$  der Zeitnutzungsfaktor und der letzte Faktor ist die Zahl der genutzten  $\delta\lambda/\lambda$ -Wellenlängenbänder. Unter Verwendung von (1) und (2) ergibt sich für das Intensitätsverhältnis bei gleichem mittlerem Fluß  $\bar{\vartheta}' = \bar{\vartheta}$  und gleicher Wellenlängenauflösung  $\frac{\delta\lambda'}{\lambda} = \frac{\delta\lambda}{\lambda}$

$$(8) \quad \frac{I}{I'} = \frac{T_{Ch}}{R_K'} \cdot \frac{\delta\alpha_h}{\delta\alpha'_h} \cdot \frac{\delta\alpha_v}{\delta\alpha'_v} \cdot \frac{L}{l}$$

Das Intensitätsverhältnis ist unabhängig von der Wellenlänge, der Chopper-Öffnungszeit und Pulsbreite, allein im wesentlichen durch das Längenverhältnis  $L/l \ll 10$  bestimmt.

Für eine Abschätzung des Intensitätsgewinns zugunsten des Spektrometers müssen die weiteren Parameter festgelegt werden. Bei der Choppertransmission kann man einen Wert von 90 % annehmen. Für die Kristallreflektivität bei den Wellenlängen

um  $2^\circ$  ist  $R_K' = 0.5$  ein guter Wert. Die Divergenzen des Spektrometers an der gepulsten Quelle sind einfach anzugeben.

$$\delta\alpha_h = \frac{10 \text{ cm}}{8.5 \text{ m}} = 1.2 \cdot 10^{-2} \text{ rad}$$

$$\delta\alpha_v = \frac{10 \text{ cm}}{8.5 \text{ m}} = 1.2 \cdot 10^{-2} \text{ rad}$$

Die Divergenzen beim Referenzdiffraktometer sind nicht so einfach festzulegen. Vertikal ist die Divergenz nur in zweiter Ordnung auflösungsbestimmend. Sie ist Höhe des gekrümmten Monochromators durch Abstand zum Detektor

$$\delta\alpha_v' \leq \frac{30 \text{ cm}}{5 \text{ m}} = 6 \cdot 10^{-2} \text{ rad},$$

also sehr groß.

Horizontal muß man mit Kollimatoren arbeiten, um die hohe Auflösung bei der Kristallreflexion zu erzielen. Für den groben Kollimator ist

$$\delta\alpha_h' = 20' = 0.6 \cdot 10^{-2} \text{ rad}$$

Die horizontale Krümmung bewirkt eine Verschlechterung der Wellenlängenauflösung.

Mit diesen Zahlen ist der Intensitätsgewinn

$$\frac{I}{I'} = \frac{0.9}{0.5} \cdot \frac{1.2}{6} \cdot \frac{1.2}{0.6} \cdot \frac{10}{1.5} = 5$$

Bei dem 10'-Kollimator ist der Intensitätsgewinn sogar **10**-fach.

Für Experimente mit geringerer Q-Auflösung ist der Gewinn geringer, weil dann  $\delta\alpha_h'$  beim Referenzdiffraktometer größer gemacht werden kann, während beim Spektrometer an der gepulsten Quelle die Divergenzen durch Abstand und Quellenquerschnitt festgelegt sind.

Der Intensitätsgewinn läßt sich wohl nur durch zwei Maßnahmen steigern:

- (a) Der Chopper rückt noch näher an den Moderator.
- (b) Die vertikale Divergenz wird vergrößert durch ein Fokussierungselement, z.B. Minineutronenleiter.

## 6. Meßbetrieb

Die registrierten Neutronen werden nach Streuwinkel  $\vartheta$  und Ankunftszeit  $T_M$  sortiert. Beide Parameter legen den  $Q$ -Vektor nach Betrag und Richtung fest. Die in jedem  $(\vartheta, T_M)$ -Kanal registrierte Intensität wird bei Kenntnis des Primärspektrums auf Wirkungsquerschnitte umgerechnet. Entlang der  $\vartheta$ -Kanäle ändert sich  $Q$  nach Richtung und Betrag bei festem  $T_M$ . Bei festem  $\vartheta$  ändert sich  $Q$  dem Betrage, aber nicht der Richtung nach entlang der  $T_M$ -Kanäle.

## 7. Charakteristische Daten des Spektrometers an der gepulsten Quelle

|                                     |                                                                         |
|-------------------------------------|-------------------------------------------------------------------------|
| Wellenlängenbereich                 | : $1 \text{ \AA} - 4 \text{ \AA}$                                       |
| Chopperöffnungszeit                 | : $25 - 100 \text{ } \mu\text{s}$                                       |
| Chopperabstand                      | : <b>1.5 m</b> von der Quelle                                           |
| Probenort                           | : 8.5 m von der Quelle                                                  |
| Relative $Q$ -Auflösung             | : $\frac{\delta Q}{Q} \approx 1.5 \% \text{ bei } \vartheta = 90^\circ$ |
| Relative Wellenlängenauf-<br>lösung | : $\frac{\delta \lambda}{\lambda} = 1 \%$                               |
| Nachweis                            | : Bananen-Multidetektor                                                 |

Keine  $\lambda/2$  Kontamination.

Probenort fest.

Geringer Platzbedarf (nur Detektorbewegung).

Untergrund schneller Neutronen unterdrückt.

Intensitätsgewinn gegenüber dem Referenzdiffraktometer 3-10-fach je nach Auflösung.

## 8. Danksagung

Herrn Axmann, HMI, danken wir für die Erläuterungen des Referenzdiffraktometers und hoffen, daß die angegebenen Diffraktometerdaten kritisch geprüft und gegebenenfalls korrigiert werden. Für eine Stellungnahme sind wir dankbar.



# A Spectrometer for Diffuse Scattering at a Pulsed Source

J. Schelten, G. Bauer, SNQ, KFA Jülich

This instrument is meant for studies on small effects, for instance for the determination of small defect concentrations in crystals, of the lattice sites of such defects (by determining the distortion of the lattice around the individual defect) and for information on the intermolecular forces as they can be deduced from such experimental data by means of theoretical models. Such studies require the separation of elastic from inelastic scattering. Otherwise, the weak scattering from the few defects is unobservable underneath the large thermal diffuse scattering. But only a modest energy resolution (of about 10 %) is required. A range of momentum transfers  $Q$  between 0.1 and  $2 \text{ \AA}^{-1}$  must be covered, again with modest resolution. Thus wavelenghtes of 4 to  $8 \text{ \AA}$  are best suited, and the instrument is best operated at the end of a long neutron guide. In the following, we propose a novel device to fullfill these conditions.

The device is a concentric arrangement of 6 cylinders rotating around the sample which is placed in the center as shown in fig. 1. Each cylinder is equipped with gaps and bars of equal widths such that only neutrons of velocities  $v_0, \frac{1}{3} v_0$  and  $\frac{1}{6} v_0$ , that is: of wavelenghts  $\lambda_0, 3\lambda_0$  and  $6\lambda_0$ , can enter radially from the outside and leave radially from the inside. This is accomplished in the way shown in fig. 2. With the requirement that the selector should have a transmission of 50 % and a resolution  $\Delta\lambda/\lambda = 12,5 \%$  one may arrive at the following specifications:  $R_1 = 30 \text{ cm}$ ,  $R_6 = 80 \text{ cm}$ ,  $d_1 = 0.38 \text{ cm}$ ,  $d_6 = 1 \text{ cm}$ . For  $v_0 = 1000 \text{ ms}^{-1}$  ( $\lambda_0 = 4 \text{ \AA}$ ), and  $\alpha = (R_6 - R_1) \frac{\omega_s}{v_0} = 0.16 \text{ rad}$  (see fig. 2.2) the rotation frequency then must be  $\omega_s/2\pi = 50.9 \text{ Hz}$ .

The device acts as a monochromator and a selector of elastically scattered neutrons at the same time. The scattered neutrons are detected as a function of the scattering angle and of their flight time in the neutron guide between moderator and spectrometer. The flight time is proportional to the neutron wave-

length. Hence the signal in the individual detector is

$$I(\lambda_i) = g(\lambda_i) \cdot \int S(\lambda_i, \lambda_f) \cdot g(\lambda_f) \cdot d\lambda_f$$

where  $S(\lambda_i, \lambda_f)$  is the scattering law and the  $g(\lambda)$  are the transmission of the spectrometer. Ideally, the  $g(\lambda)$  have a triangular shape, so that for elastic scattering  $[S(\lambda_i, \lambda_f) = \delta(\lambda_i - \lambda_f)]$   $I(\lambda_i) = I_{el}(\lambda_i) = (g(\lambda))^2$  will look as shown in fig. 3. The determination of the function  $I(\lambda_i)$  is extremely important as deviations of  $I(\lambda_i)$  from  $(g(\lambda))^2$  indicate the presence of inelastic scattering, for instance due to multiple scattering.

In principle, the same instrument can be used also at a steady-state neutron source. However, if then again not only  $I(Q)$  but also  $I(\lambda_i)$  are to be measured, the signals would be lower by about a factor 10, even if the neutron flux were the same as the SNQ time-averaged flux. For with the required resolution  $\Delta\lambda/\lambda = 12,5\%$  and with a flight path of 20 m the full SNQ pulse width of 250  $\mu s$  and about 1/5 of the 10 ms between pulses can be used. In this fraction of the time the peak flux is employed.

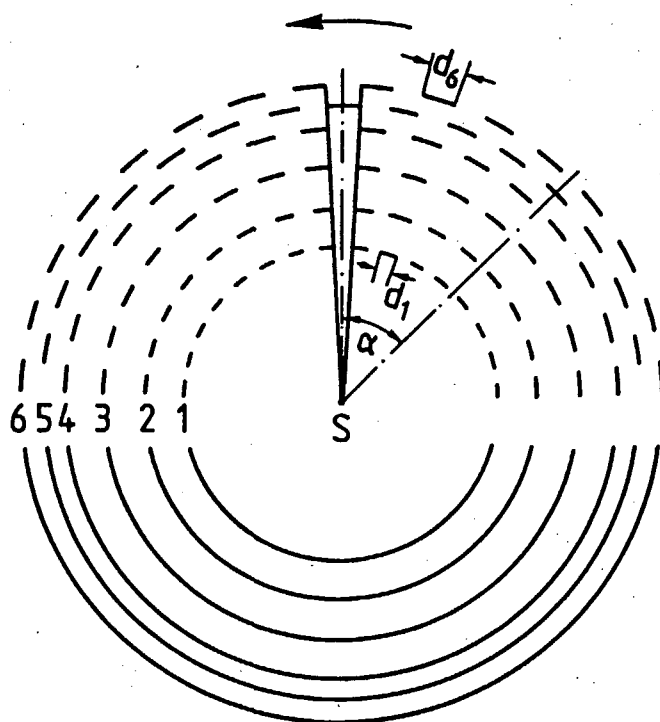


Fig. 1

Principle of the diffuse scattering instrument. The sample is placed in the center of six concentric rotating cylinders as described in the text. The neutron beam enters radially from one side, say from the left. The neutrons scattered in the sample are detected with counters placed around the outer cylinder.

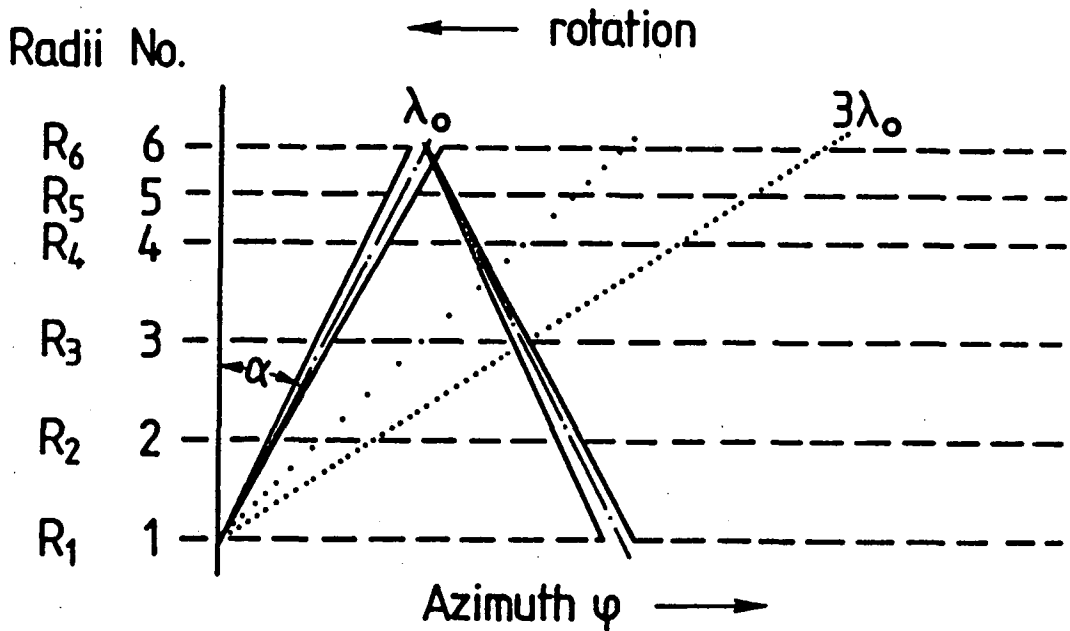


Fig. 2

With steady rotation, the azimuthal angle  $\psi$  is proportional to the time  $t$ . The solid lines hence represent the path-time diagram of neutrons moving radially to the outside (against the cylinder-rotation) or to the inside (with the cylinder-rotation). The slopes of the lines are the velocities. The neutron flight path between the innermost and the outermost cylinder is presented in units of the distance  $D = R_1 - R_6$ . The cylinders 1, 2, 3, 4 and 6 have neutron gaps arranged radially one behind the other (see fig. 2.1). Obviously, if these cylinders rotate with angular velocity  $\omega$ , only neutrons of velocity  $v_0 = D \cdot \omega$  can pass through ( $n = 1, 2, 3, \dots$ ). Cylinder no. 5 is inserted in order to suppress  $n = 2, 4$  and 5, so that only neutrons with  $v_0/3$  and  $v_0/6$  also can pass. These neutrons can be eliminated by a conventional velocity selector in the incident beam (or also ~~for~~ <sup>with</sup> appropriate gating of the detectors).

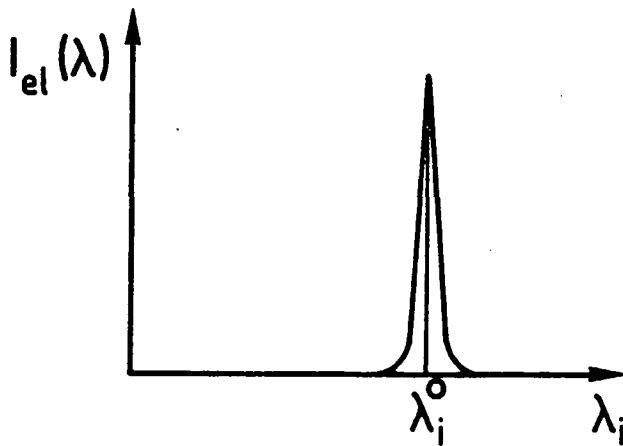


Fig. 3

The signal in one detector measured as a function of the flight time of the incident neutrons, i.e. of their wavelength  $\lambda$ , for elastic scattering.

Working Group "Inelastic Scattering at the SNQ"

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Participants: H. Arnold, G. Badurek, B. Dorner, H. Grimm,  
A. Kollmar, M. Loewenhaupt, H. Rauh,  
W. Reichardt, V. Wagner

Contributors to the papers and guests: R. Geick, L. de Graaff,  
L. Pintschovius, J.-B. Suck, A.D. Taylor, W.G. Williams,  
C.G. Windsor

Chairman's Report , W. Reichardt, KfK Karlsruhe

This report is intended to summarize and critically review the work of one of the working groups. It cannot and - I think - it should not be avoided, that such a report will reflect a lot of the author's own point of view. Therefore some of the members may object to the way, how things are dealt with, but I am sure that they agree with the essential statements, which are mostly based on discussions within the working group.

When we met in Bad Kissingen this April, the task, to think about instruments at a pulsed neutron source, was completely new to most of us. We started setting up the following list of topics, which should be studied until the next meeting:

1. Polarization Analysis
2. Fourier Methods and Correlation Techniques
3. Realtime Experiments
4. Comparison Between Crystal Spectrometer Methods  
and Time-of-Flight Methods
5. Measurements of Collective Excitations with Triple  
Axis Spectrometers

6. Measurements of Collective Excitations with Time-of-Flight Methods
7. Measurements of Isotropic Scattering Laws
8. Spectroscopy with High Energy Neutrons
9. Spectroscopy with Cold Neutrons.

It was obvious from the very beginning, that it would not be possible to cover this large field in an exhaustive manner. Therefore it was left to the individual authors' own decision, whether they would like to treat a larger field in a more general way, or to pick out specific aspects. As a consequence the contributions range from a detailed description of a particular spectrometer to general discussions about methods and physical applications.

This is the list of the papers, which are the output of the working group's studies:

- (1) Polarization Analysis  
V. Wagner, M. Loewenhaupt
- (2) Correlation and Fourier Time-of-Flight Spectroscopy  
at a Pulsed Neutron Source  
G. Badurek
- (3) Real Time Experiments  
B. Dorner
- (4) Comparison Between Time-of-Flight Methods and the  
Crystal Spectrometer Method at the SNQ for Measurements  
of Collective Excitations  
W. Reichardt
- (5) Triple Axis Spectroscopy at the SNQ  
W. Reichardt, L. Pintschovius, B. Dorner
- (6) Multi-Arm Crystal Analyzer TOF Spectrometer (TOF-XXX)  
H. Rauh, R. Geick

- (7) Computer Simulations of Phonon Measurements on a Multi Detector Time-of-Flight Spectrometer  
W. Reichardt
- (8) Some Considerations Concerning the Measurements of Dynamic Structure Factors and Frequency Distributions of Isotropic Samples at the SNQ  
J.-B. Suck and M. Loewenhaupt
- (9) High Energy Neutron Scattering  
M. Loewenhaupt, H. Rauh, V. Wagner
- (10) A Small Angle Spectrometer for Brillouin Scattering  
W. Reichardt

I add two more titles, which may be cited also in connection with the studies of the working group "Special Techniques", because their topics are closely connected with that of paper (6).

- (11) Constant-Q Scan an der gepulsten Quelle  
J. Schelten, B. Alefeld
- (12) A Constant-Q Scan at Constant Scattering Angle  
C.G. Windsor

At the last session of the workshop in Maria Laach we were asked to set up a list of spectrometers, which we recommend for the SNQ.

List of Instruments Recommended for the SNQ:

- 1. TAS for thermal neutrons
- 2. TOF-X spectrometer with multi-analyzer (MAX-spectrometer)
- 3. TOF-X spectrometer dedicated to PA
- 4. TOF-TOF spectrometer for thermal neutrons
- 5. X-TOF for thermal neutrons with option of Fourier chopper
- 6. Inverted geometry fitter spectrometer for chemical spectroscopy
- 7. TOF-TOF spectrometer for epithermal neutrons
- 8. Resonance detector spectrometer for epithermal neutrons
- 9. TAS for cold neutrons at neutron guide with option of PA



10. TAS for cold neutrons at a beam tube with option of PA
11. TOF-TOF for cold neutrons at neutron guide
12. X-TOF for cold neutrons at a beam tube with option of Fourier chopper and PA

(TAS = triple axis spectrometer, TOF-TOF = chopper time-of-flight spectrometer, X-TOF = direct geometry hybrid spectrometer, TOF-X = inverted geometry hybrid spectrometer, PA = polarization analysis).

These instruments should be considered as first day's instruments for the SNQ without compressor ring, unless it is specified differently in the subsequent comments.

#### A. Pulse Frequency of the Source

A pulsed source, where experiments will be done with neutron energies covering several orders of magnitude, cannot have the optimum repetition rate for each individual measurement. The working group agreed that the proposed 100 Hz is a satisfactory choice. Some restrictions will be imposed on the spectroscopy with cold neutrons because the flight times of the neutrons between the moderator and the detector can cover several periods of the source. Therefore experiments have to be planned in such a way, that the background arising from fast neutrons during and after the proton pulse is shifted to a region of the time-of-flight spectrum, which is of little importance.

#### B. Compressor Ring

All TOF experiments will profit from the compressor ring. For the spectroscopy with epithermal neutrons the installation of the compressor ring is crucial. I would like to quote here Ref. (9). "The loss factor of 125 for the version without compressor ring will settle the SNQ well below the SNS, roughly at the level of the IPNS". The factor 125 is specific for 1 eV neutrons, but the statement is generally valid for energies above about 200 meV.

### C. Moderator Concept and Tail Cutters

The present moderator concept is probably the best solution to provide the optimum intensity for all beam tubes and serve equally well TOF experiments and stationary experiments. All TOF experiments will need a tailoring of the moderator pulse. Therefore, in most cases mechanical choppers have to be installed close to the moderator. This is certainly a very hard technical problem. I believe, that in the thermal and sub-thermal range tailoring the pulse by a chopper should be preferred to all other methods (thin moderator, poisoning of the moderator), because the burst width can be chosen individually for each experiment and the pulse shape of a chopper is more valuable than the asymmetric distribution of the moderator pulse. Above 200 meV the use of mechanical tail-cutters becomes more and more impracticable. Therefore it should be considered, whether one side of the moderator (relative to the proton beam) can be optimized with respect to good pulse shapes in the high energy region, whereas the opposite side is optimized with respect to high overall intensity.

### D. Polarization Analysis

There was general agreement, that at the SNQ a wide use of polarization analysis should be possible. As the SNQ is a very powerful source, many experiments will become feasible, which fail at the existing sources due to insufficient intensity. There are several spectrometers in our list, which should have the option for polarization analysis, and one instrument which is dedicated exclusively to the application of polarized neutron.

### E. Fourier and Correlation Methods

Fourier and correlation methods are very similar concerning the gain they may provide compared to conventional TOF methods. At a pulsed source their performances can be improved considerably, because their major drawback at stationary sources - large elastic scattering spoils the statistical accuracy in the inelastic region - can be eliminated. The Fourier method

offers several advantages compared to the correlation technique: choppers do not impose restrictions on the cross-section of beam; with one properly designed rotor one can achieve almost any desired resolution; in specific applications it is sufficient to measure only a few Fourier components, which leads to a considerable reduction in measuring time.

There appears to be only one type of spectrometer, which allows a fruitful application of Fourier spectroscopy, the direct geometry hybrid TOF spectrometer. There are two instruments of this type in our list, where a Fourier chopper may be used as an option.

I must say, that at present I have no clear idea, to which extent the Fourier technique will be applied successfully at the SNQ.

#### F. Realtime Experiments

Realtime experiments will gain at the SNQ to the same extent as TOF spectroscopy will profit in general from the pulsed operation of the source. In addition to that I do not see a particular advantage of the SNQ. At present there is no indication, that realtime experiments will require specialized spectrometers.

#### G. Spectroscopy with Cold Neutrons

We have largely neglected this field in our discussions. This is partly my own fault, because I have not done my homework. On the other hand, this field has been dealt with extensively in the two other working groups "High Resolution Spectroscopy" and "Special Techniques". We have recommended that, in addition to the neutron guides, the cold source should have also a few beam tubes into the experiment hall. At these beam tube experiments, which can exploit a coarse angular resolution, will be performed.

#### H. TOF Spectrometers for High Energies

It was argued that the proposed small angle spectrometer (10) might be too restricted and therefore should be equipped with additional detectors at larger scattering angles in order to extend its range of application. This instrument will require the narrow pulses delivered by the compressor ring. Nevertheless, it was recommended to have such a spectrometer already at the SNQ without compressor ring. Although it will not be as powerful as it should be, already a lot of good research can be done and experience can be gained about spectroscopy with high energy neutrons.

The spectrometer with a resonance detector is useful only after the installation of the compressor ring.

#### I. Inverted Geometry TOF Spectrometers

The multi-analyzer TOF-spectrometer is a particularly exciting instrument due to its high flexibility. It is my impression, that it is mostly regarded as a "high-symmetry direction" spectrometer. This is only one mode of operation out of many. It must be pointed out, that sticking to high symmetry directions will prevent, that the various analyzers can be optimized individually with respect to resolution and intensity. In the design studies of this spectrometer one should take into account, that many experiments will profit from a dense spacing of the multi-analyzers. This is certainly not an easy technical problem.

The working group has recommended, that a spectrometer of this type should be built, if the German neutron community participates at the SNS.

There are two proposals, by J. Schelten and B. Alefeld and by C.G. Windsor, which demonstrate, that const. Q scans can be performed at this spectrometer, if certain components

of the spectrometer vibrate in a specific manner. There is no doubt, that these methods are technically feasible, whether they are also practicable, has to be demonstrated by further studies.

In spite of its very good intensity the inverted geometry filter spectrometer is not liked very much among physicists because of its limitations in the momentum transfer. It is particularly well suited for chemical spectroscopy, which was the reason, why it was included in our list. While writing this report, I got some doubt, whether at the SNQ this is really the proper instrument for the intended applications. At the SNQ this spectrometer cannot operate without a tail-cutter close to the moderator. Besides changing the resolution characteristic ( $\Delta t_p = \text{const}$  compared to  $\Delta t_p \sim \frac{1}{\sqrt{E_i}}$  at other pulsed sources) at the costs of intensity, the chopper cuts out an energy band from the source spectrum, which may be too narrow for many experiments.

#### J. Direct Geometry TOF Spectrometers

There is still no agreement about the best choice for a versatile TOF spectrometer with multi-detector in the thermal range for measurements of isotropic scattering laws. In my opinion it is very important, that this instrument allows to vary the primary energy continuously. With a double chopper spectrometer (TOF-TOF) this can be achieved rather easily by changing the phase between the rotors. However, there are several aspects, which favor a hybrid instrument (X-TOF):

- a. Decoupling of primary and secondary energy resolution.
- b. There is no need for a chopper close to the moderator.
- c. The sample is outside the direct beam.
- d. The chopper is placed in the monochromatic beam, which reduces considerably the background produced by the chopper. It is easily accessible, which allows fast replacements of rotors for a change in resolution.

- e. Possibility to use Fourier choppers for high resolution work.
- f. Curved monochromators can be used for experiments with small samples.

Some of these arguments are specific for the SNQ, because the present moderator concept will require tailoring of the source pulse by a chopper for all experiments.

A serious drawback of this instrument is the necessity to move the sample table and the secondary flight-path together with the heavily shielded multi-detector when the primary energy is changed. Up to now no satisfactory design exists for a spectrometer with vertical scattering plane, which is required by the limited space available.

A possible solution might be the installation of an inclined beam tube into the basement of the experimental hall with an angle of inclination  $\phi$  of about 20 to 30 degrees. The monochromatic beam can be rotated around a vertical axis for changing the primary energy. In this way it is possible to achieve monochromator take-off angles between  $\phi$  and about  $90^\circ$ , and the scattering plane can be arranged horizontally. This proposal removes the most serious technical problems, because all components of the secondary spectrometer can move on air cushions, but it requires a modification of the moderator design. Whether this is possible without influencing the performance of other instruments, remains an open question.

These comments are restricted to only a few topics. More information should be obtained from the reports listed above.

Finally I would like to thank all participants of the workshop and also those, who have contributed to the various studies without being present at our meetings, for their readiness to spend such a large amount of effort and time in this work.

### Triple Axis Spectroscopy at the SNQ

W. Reichardt, L. Pintschovius, Kernforschungszentrum  
Karlsruhe, INFP, Postfach 3640, D-7500 Karlsruhe, FRG.

B. Dorner, Institut Laue Langevin, 156 X, F-38042 Grenoble  
Cedex, France.

#### Abstract

It is investigated, how the time structure of the pulsed neutron source SNQ can be exploited to improve the performance of a triple axis spectrometer. Major advantages are the elimination of spurions and the use of higher order reflections of crystal monochromators by gating the detector. Several methods, which can provide a gain in measuring time are discussed. It is concluded, that these methods are either very limited in their applicability or are mainly suited for such types of experiments, which can be performed equally well on time-of-flight spectrometers. In addition some problems concerning the design of a TAS at the SNQ are pointed out.

## 1. INTRODUCTION

It has been shown in a separate report /1/ that also at the pulsed neutron source SNQ the conventional triple axis spectrometer (TAS) is a very useful instrument for the measurements of collective excitations in single crystals. This applies in particular to the range of thermal and subthermal neutrons. In this report we discuss special features which arise from the pulsed nature of the neutron source. Furthermore we deal with problems, which may influence the design of a TAS at the SNQ.

At the SNQ the time averaged leakage current from the thermal moderator corresponds to a reactor flux of  $\bar{\phi} = 1.2 \times 10^{15} \text{ ncm}^{-2} \text{ sec}^{-1}$ . This value is comparable to the flux at the Grenoble HFR. Due to the pulse structure of the source the neutron intensity is confined to narrow time intervals with a width of  $\Delta t_p = 140 \text{ } \mu\text{s}$  for the SNQ with compressor ring. The time between the pulses is 10 ms. Therefore time-of-flight measurements with a coarse time resolution are possible. This raises the question, whether this additional degree of freedom can be exploited in order to improve the performance of the instrument.

## 2. ELIMINATION OF SPURIONS

A serious drawback of triple axis spectrometers at stationary sources is the occurrence of spurions, which are mostly caused by elastic scattering in the sample (Bragg-reflections in first order or elastic scattering, both coherent and incoherent, for configurations  $n^2 E_i \approx m^2 E_f$ ). Although most of the origins of spurions are known, very often serious restrictions are imposed on the optimization of the experiment by the requirement that



"dangerous" configurations have to be avoided. As elastically and inelastically scattered neutrons arrive at the detector at different times, the contributions from elastic scattering processes can be eliminated by gating the detector only for a short time interval at the appropriate time. The same applies also to inelastic scattering processes like  $\hbar\omega(2Q) = 4E_i - 4E_f$ . Gating of the detector does not help much in the case of small excitation energies close to a Bragg-point (elastic region) because of insufficient time resolution and the short distance between sample and detector.

### 3. CRYSTAL MONOCHROMATORS: HIGHER ORDER REFLECTIONS AND FOCUSING

Gating of the detector will also allow to use higher order reflections of the monochromators and/or analyzers. This will extend the applicability of the few good monochromator crystals, which are available today, and improve the flexibility of the spectrometer.

Monochromators curved perpendicular to the scattering plane for focussing the beam onto the sample are already widely used at stationary sources. At the SNQ they are even more important due to the larger distance between source and sample (about 10 m compared to 6-8 m at reactors).

Only limited use has so far been made of the Rowland focussing. In all cases this was done with the analyzer. On the monochromator side one meets with the difficulty, that the distances source-monochromator  $L_1$  and monochromator sample  $L_2$  have to be very different because of the thick biological shields of neutron sources, whereas only for  $L_1 \approx L_2$  the reflecting planes of the monochromator are approximately parallel to the Rowland circle. At the SNQ

this asymmetry of the distances will be even more pronounced. Nevertheless this method of focussing should be further studied because it may influence the design of the beam tubes. The proposal given in Fig. 1 requires a variable diaphragm inside the biological shield and an increase of the beam tube's width towards the moderator.

#### 4. PROBLEMS, CONCERNING THE DESIGN OF A TAS AT THE SNQ

Triple axis spectrometers need a lot of space close to the biological shield. This should be taken into account, when the arrangement of beam tubes at the SNQ is planned. Experimentalists will object to the proposal, to use vertical scattering planes in order to save space, because this causes serious problems for the sample environment (cryostats, cryomagnets). Also serious problems will arise for the mechanics of the spectrometer. Therefore it is necessary to perform design studies of a TAS with a vertical scattering plane, that they can be discussed critically by the future users.

At a reactor the different compounds of a TAS, monochromator, analyzer and detector, are individually shielded with heavy, neutron moderating and absorbing materials. At the SNQ even larger shielding masses are required because of the high energy neutrons. This may lead to unreasonable dimensions and weights. Therefore it might be more favorable, to place the whole instrument into a thick-walled shielding house, whereas the individual components of the spectrometer are shielded against each others only by light neutron absorbing, but not moderating materials. Such a concept is only possible by the pulsed operation of the source. It is hoped, that the background of fast neutrons has already decayed sufficiently, when the thermal neutrons are recorded in the detector. The background level must be

smaller than 1 pulse per minute in order to meet the values obtained at today's instruments. This may be the only practicable solution, if the scattering plane of the spectrometer has to be vertical.

The concept of a lightly shielded spectrometer in a surrounding shielding house will not allow, that the user stays close to the experiment, while the spectrometer is in operation. All adjustments of the sample have to be automatized and visual inspection is only possible via video cameras.

The frequently used W-configuration of a TAS causes, that the detector gets often very close to the primary beam. At present we have no experience, whether this will cause serious background problems.

#### 5. USE OF THE TIME STRUCTURE TO IMPROVE THE PERFORMANCE OF A TAS

A TAS at a pulsed source may be considered also a TOF-spectrometer. Is it possible to improve by an additional time analysis the performance of the spectrometer in a single scan, or to achieve a gain in measuring time by a multiple use of the instrument?

Gains can be achieved by the following methods:

1. Use of several analyzer devices
2. Selection of several neutron energies in the primary beam
3. Combination of 1. and 2.
4. Use of neutrons, which are not scattered or absorbed in the sample, and are therefore wasted in the beam dump.
5. By phase space transformation with moving crystals.

Some examples are given in section 6.

The use of several analyzer devices is also possible at stationary sources. Spectrometers of this kind were not very successful, because the flexibility of the instrument is reduced and there are not many experiments, which allow a successful application of multi-analyzers.

When investigating the multiple use of a TAS one has to consider the following aspects, which have been discussed in detail in Ref. 1:

- a) At the SNQ a TOF spectrometer is superior to a TAS with respect to intensity, if the scanning range is large compared to the resolution of the instrument.
- b) The unique advantage of a TAS is given by the possibility, to choose the optimum type of scan, intensity, resolution, focussing and measuring time for each scan.
- c) In the range of thermal neutrons ( $E_i \lesssim 100$  meV) the resolution in energy of the TAS is smaller than the energy window resulting from the time structure.

This will be explained more explicitly by a special example:

If we want to determine an excitation energy of 25 meV with a resolution  $\Delta\hbar\omega/\hbar\omega = 10\%$ , a resolution  $\Delta E_i/E_i = 3.5\%$  is required for a primary energy of 50 meV.

Assuming a width of the moderator pulse of  $140\ \mu\text{s}$  and distances moderator-sample  $L_1 = 10$  m and sample-detector  $L_2 = 2$  m a pulse width at the detector of  $\Delta t_0 = 158\ \mu\text{s}$  is obtained. Two distributions, whose centers of mass are separated by only 1 FWHM can no longer be separated uniquely. The separation has to be at least twice as large in order to avoid strong overlap. If one wants to select in addition to the first energy  $E_i = 50$  meV a second primary energy by another monochromator, a unique

attribution of the detector counts is only possible, if the two energies differ by more than

$$E = 2 \cdot 2 \frac{\Delta t_o}{t_i} \Delta E_i = 9.8 \text{ meV} \quad .$$

This value has to be compared with  $\Delta h\omega = 2.5 \text{ meV}$ ; the energy window has to be 4 times as large as the resolution of the TAS.

The above example demonstrates, that for a single scan the use of several primary energies will provide a gain in intensity only in those cases, where the length of the scan is large compared to the resolution of the spectrometer.

The simultaneous measurement of several excitation energies by using several monochromators and/or analyzers largely reduces the advantages of a TAS which are mentioned in b. As the sample orientation has to be the same for all simultaneous measurements the different scans in the  $\hbar\omega$ - $\vec{Q}$ -space are no longer independent of each others. Only two of the three variables  $\hbar\omega$ ,  $Q_x$ ,  $Q_y$  (measured in the coordinate system of the sample) can be freely chosen. It is for instance possible to choose  $|\vec{Q}|$  for all simultaneous measurements and to perform const.  $|\vec{Q}|$  scans.

## 6. SOME EXAMPLES FOR THE MULTIPLE USE OF A TAS

A very simple method is the simultaneous use of first and second order reflections from the monochromator and the analyzer. If a scan is performed in first order at  $(\hbar\omega, \vec{Q}=\vec{q}+\vec{\tau})$ , the second order yields a similar scan at  $(4\hbar\omega, 2\vec{q}+2\vec{\tau})$ . The relative resolutions  $\Delta\hbar\omega/\hbar\omega$  are the same for both measurements. Only two counters with variable time windows are necessary to separate the counts of the two scans.

The ideas of B. Alefeld /2/ for a modified backscattering spectrometer "Musical", can also be applied to a TAS (see Fig. 2): A monochromator system consisting of  $n$  pairs of crystal monochromators select  $n$  equidistant neutron energies with increments  $\Delta E_i$ . With a single setting of the analyzer a whole scan is performed consisting of  $n$  hw-points separated by  $\Delta E_i$ . The energy increments have to be chosen in such a way, that at the detector there is no overlap in time between counting events resulting from different primary energies. As at the SNQ the energy windows resulting from the pulse width of the source and a time-of-flight smearing is usually much larger than the energy resolution of the TAS, it is not possible to obtain a continuous scan (see insert of Fig. 2). The "dead" areas may be filled by subsequent measurements with different primary energies.

The operation of this spectrometer is basically identical to that of a TOF spectrometer of type TOF-X, where the continuous source spectrum is replaced by discrete energies. However, the primary energy resolution is determined by the monochromators, whereas at a TOF-X spectrometer it is determined by the width of the moderator pulse. The instrument can be equipped also with several analyzers.

The selection of equidistant, closely spaced primary energies is a special case. It is also possible to aim simultaneously at several excitation energies  $\hbar\omega_i(\vec{Q}_i)$ . In this mode of operation an instrument with  $n$  monochromators and  $n$  analyzers corresponds to  $n$  triple axis spectrometers. However, there is the restriction, that the orientation of the sample is the same for all sub-instruments. Furthermore the spectrometer configurations have to be chosen in such a way, that there is no overlap of counting events arising from different measurements. The planning of such an experiment is certainly very difficult.

An alternative version of the spectrometer with a very similar performance is shown in Fig. 3

The proposal shown in Fig. 4 is intended to make a better use of the primary beam in experiments with small samples. The monochromatic beam is reflected several times in backscattering between two crystals. Crystal I is attached to a rotating disc, which has an entrance window, in order to allow the beam to pass through the disc before the first reflection at crystal II. There are two identical analyzers, which are arranged in an inverted geometry relative to the monochromatic beam. Thus identical measurements are done with analyzer I for the even number of reflections and with analyzer II for the odd number of reflections. The distance between the two crystals has to be chosen in such a way, that there is no overlap in time between detector signals arising from different reflections. If we describe by  $\alpha$  the losses of the beam by scattering and absorption in the sample, and by  $R$  the reflectivity of the crystals, we obtain a gain in intensity after  $m$  reflections according to

$$\frac{1 - (\alpha R)^{m+1}}{1 - \alpha R}$$

For  $\alpha R = 60\%$  and 3 reflections the gain is 2.2.

There are two requirements, which are hard to fulfill:

- 1) The reflectivities of the crystals have to be considerably larger than 50%.
- 2) The energy band selected by backscattering must correspond to the required resolution. This can only be obtained by crystals, which have a gradient in the lattice constants.

It is obvious, that experiments are restricted to certain wave-lengths with  $\lambda = 2d/n$ , where  $d$  is the lattice constant and  $n$  the order of the reflection.

B. Alefeld /3/ has proposed a TOF spectrometer, where a gain in intensity is achieved by Doppler shifting neutrons from the cold source to higher energies. The same principle can also be applied to a TAS. As in this case the neutron pulse has to be reflected during its full length, several crystals have to be placed close together on the Doppler wheel.

## 7. CONCLUSION

Triple axis spectroscopy will profit considerably from the pulsed operation of the SNQ. The possibility of a simple time analysis, which in fact is only a gating of the detector, will improve the quality of the measurements and will allow a greater flexibility of the spectrometers. We studied several methods, which can achieve an increased output of the triple axis spectrometers. We found, that these methods are either very restricted in their applicability or are only suited for experiments, which at the SNQ can be equally well performed on TOF spectrometers.



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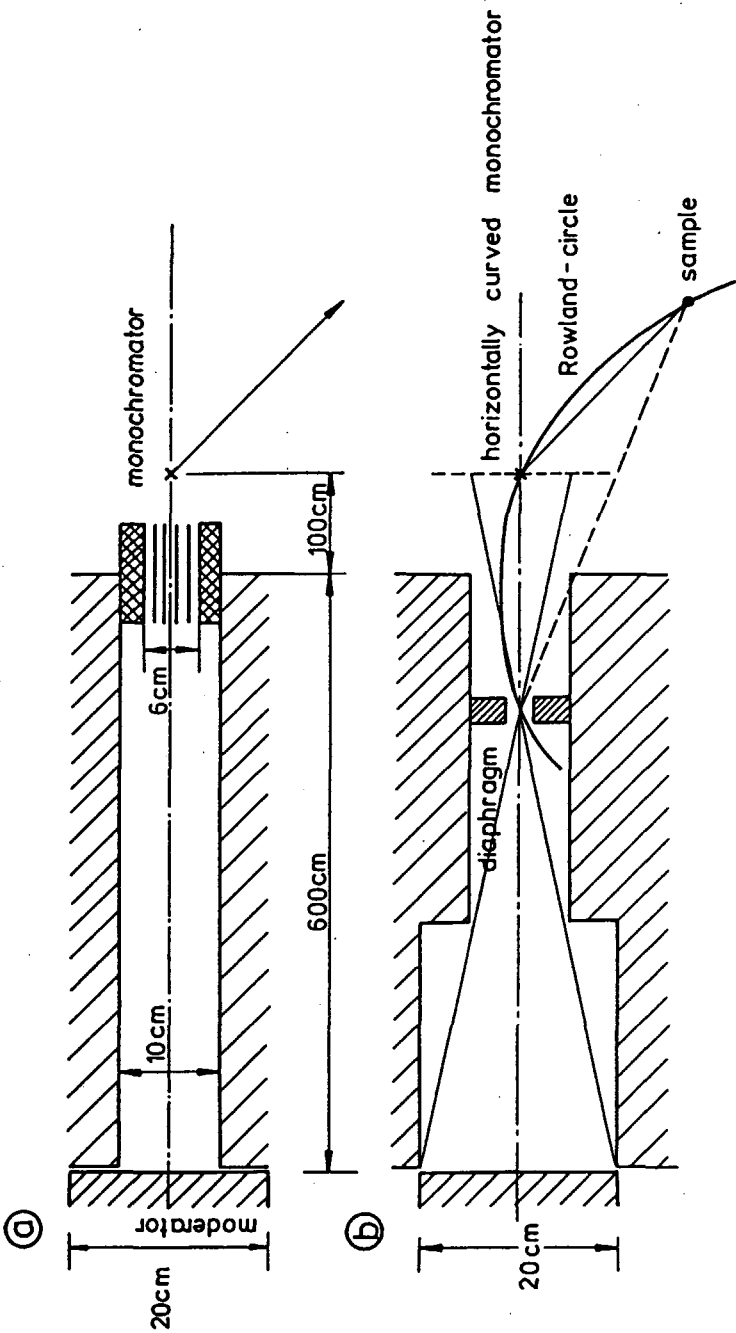


Fig.1: (a) Beam tube for a flat monochromator  
(b) Beam tube for a horizontally curved monochromator (Rowland focussing)

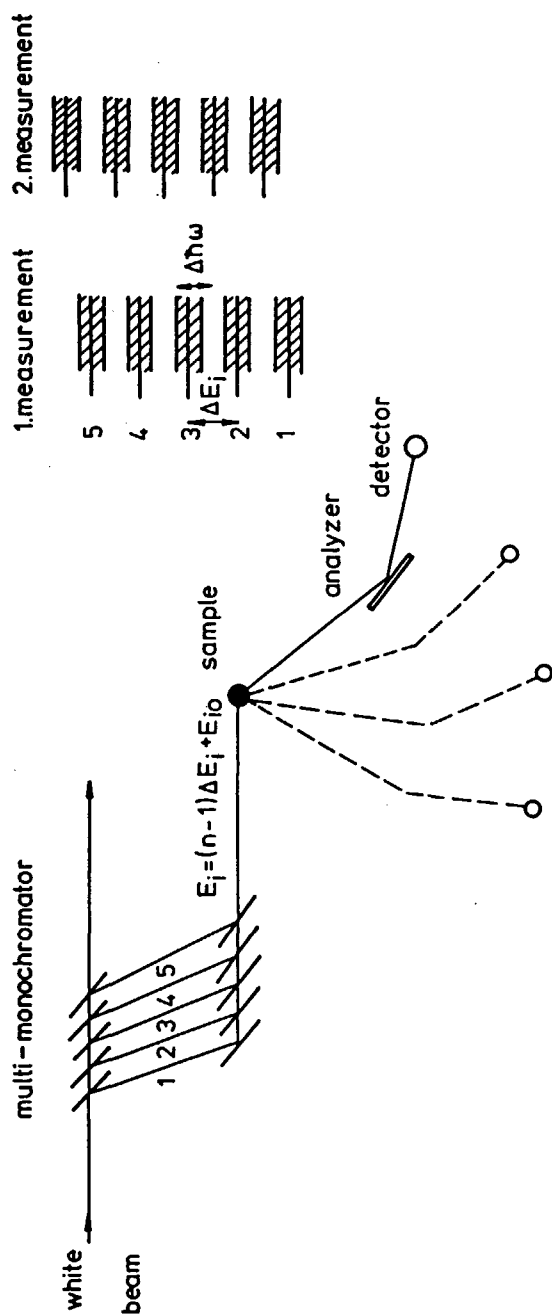


Fig.2 TAS with a multi - monochromator

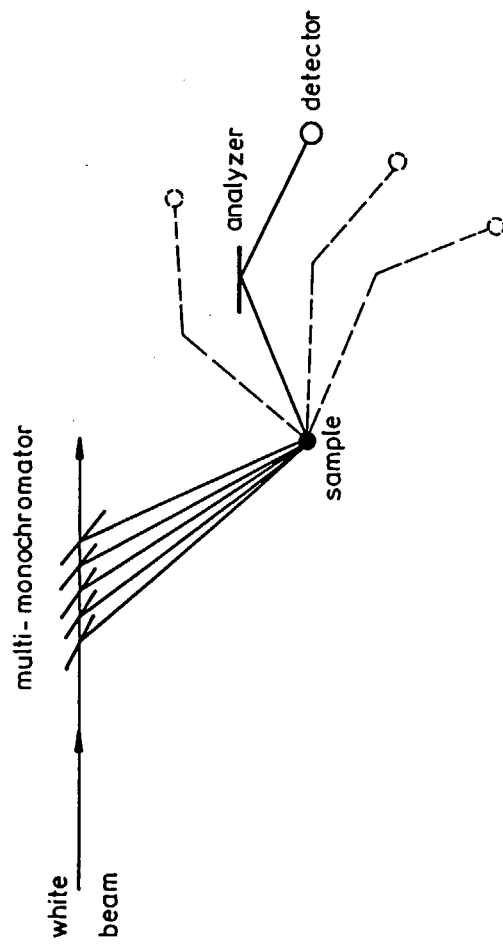


Fig.:3 TAS with a multi-monochromator

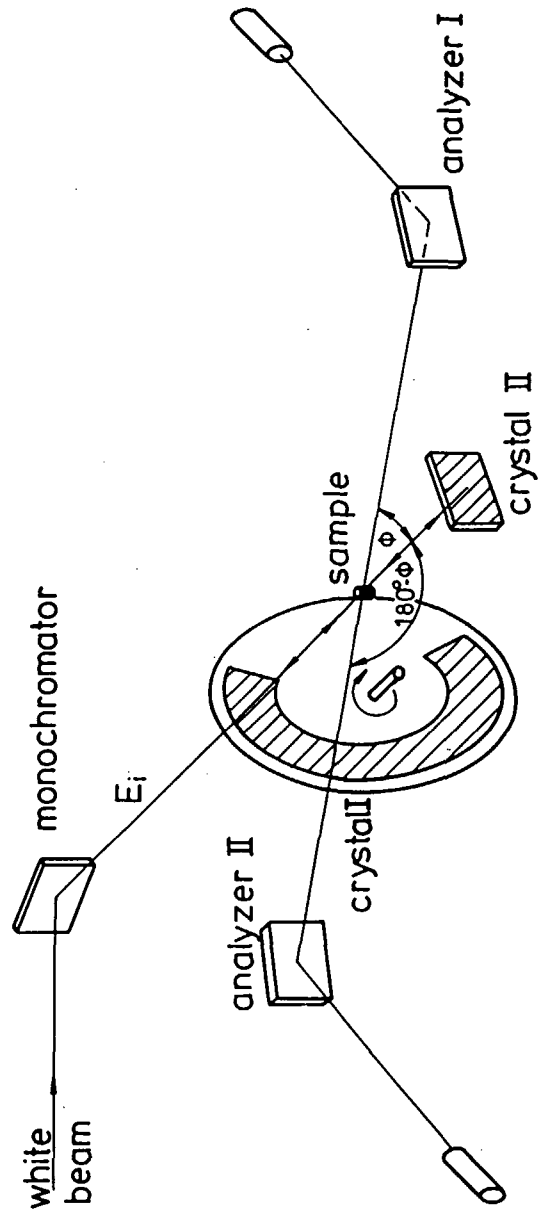


Fig.: 4 Multiple use of the primary beam on a triple axis spectrometer

## Constant-q Scan at the Pulsed Source

J. Schelten and B. Alefeld

KFA - Jülich

1. Introduction

Classical neutron spectroscopy is used for selective measurements in  $h\omega, \underline{q}$ -space. This is frequently done with the aid of the constant  $q$  scan, adjusting the spectrometer parameters in such a way that the phonon vector  $\underline{q}$  remains unchanged and only the energy transfer  $h\omega$  is varied. This favourable measurement technique can be carried out at a pulsed source with a gain in intensity. For this purpose, the classical triple axis spectrometer must be equipped with a vibrating Soller collimator and a vibrating analyzer crystal. The vibration amplitudes and phases should be adjusted in such a way that neutrons of different wave numbers are processed consecutively during one semivibration while the phonon vector  $\underline{q}$  is kept constant.

The spectrometer set-up is outlined in Fig. 1. Neutrons in the wavelength range of 2 - 4 Å are passed to the spectrometer in a neutron guide tube of 25 m - 50 m length. The mechanical velocity selector transmits a narrow ( $\pm 10\%$ ) wave number band of these neutrons and stops neutrons of shorter wave lengths coming from later neutron pulses. The transit time  $t_M$  of a 4 Å neutron over the distance of 25 m from the source to the spectrometer is 25 ms, i.e. exceeding the pulse interval of 10 ms. The wave number is determined within 1 % for a pulse width of 250  $\mu$ s by the time of arrival. Further numbers of this type are contained in Table 1.

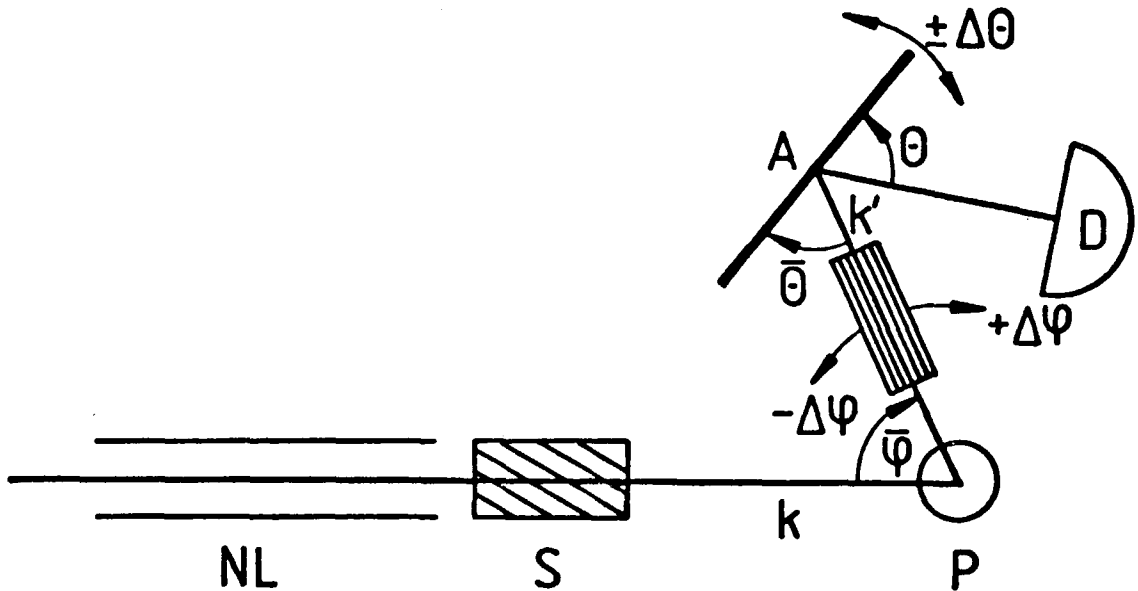
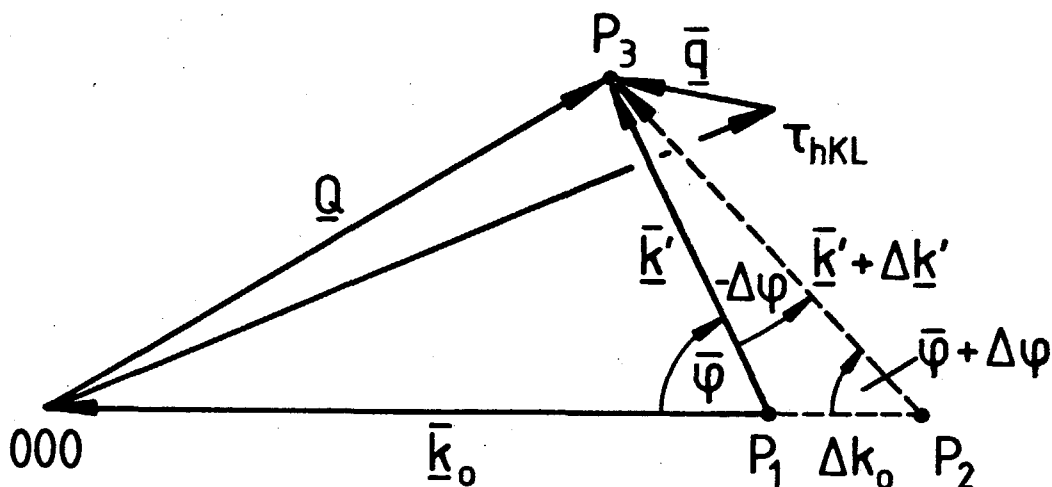


Fig. 1 Basic set-up of the modified triple axis spectrometer at a pulsed source

Table 1 Transit time and wave number resolution for various distances and neutron wave lengths at  $\Delta t = 250 \mu s$

| L (m)                  | 25 m |      |     | 50 m |     |       |
|------------------------|------|------|-----|------|-----|-------|
| $\lambda (\text{\AA})$ | 1    | 2    | 4   | 1    | 2   | 4     |
| $t_M$ (ms)             | 6.25 | 12.5 | 25  | 12.5 | 25  | 50    |
| $\delta k/k$           | 4 %  | 2 %  | 1 % | 2 %  | 1 % | 0.5 % |

The neutrons scattered in the sample are passed to the analyzer at the instantaneous angle of scattering  $\phi(t)$ , defined by the vibrating collimator. They impinge upon the analyzing crystal under the instantaneous Bragg angle  $\theta(t)$ .



$\Delta P_1 P_2 P_3$  : geg.:  $k'_M, \varphi_M, \Delta k$   
 ges.:  $\Delta \varphi, \Delta k'$

Fig. 2 Pulse diagram for constant- $q$  scan

Scattered neutrons satisfying the Bragg equation will reach the detector.

Fig. 2 illustrates the conditions in  $k$ -space leading to constant  $q$ . The spectrometer should be set up in such a way that neutrons of wave number  $k_M$ , scattered to wave number vector  $k'_M$ , lead to a desired  $q$ -vector  $q_M$  under the angle of scattering  $\varphi_M$  involving the desired  $h\omega$ -energy transfer  $h\omega_M$ , see also Fig. 3. When  $k$  is changed by  $\Delta k$ , the angle of scattering is changed from  $\varphi_M$  to  $\varphi_M + \Delta \varphi$  and the analyzer crystal is rotated so as to reflect the wave number  $k'_M + \Delta k'$ . In order to keep  $q$  constant, the changes  $\Delta k$ ,  $\Delta k'$  and  $\Delta \varphi$  must satisfy certain equations which can be read from the triangle  $P_1 P_2 P_3$ .

In a linear approximation the changes are as follows:

$$(1) \quad \Delta \varphi = - \frac{\Delta k}{k'_M} \sin \varphi_M$$

$$(2) \quad \Delta k' = + \Delta k \cos \varphi_M$$



The time-dependent changes of both variables result from those of  $\Delta k$  as

$$(3) \quad \Delta k = - \frac{k_M}{t_M} t$$

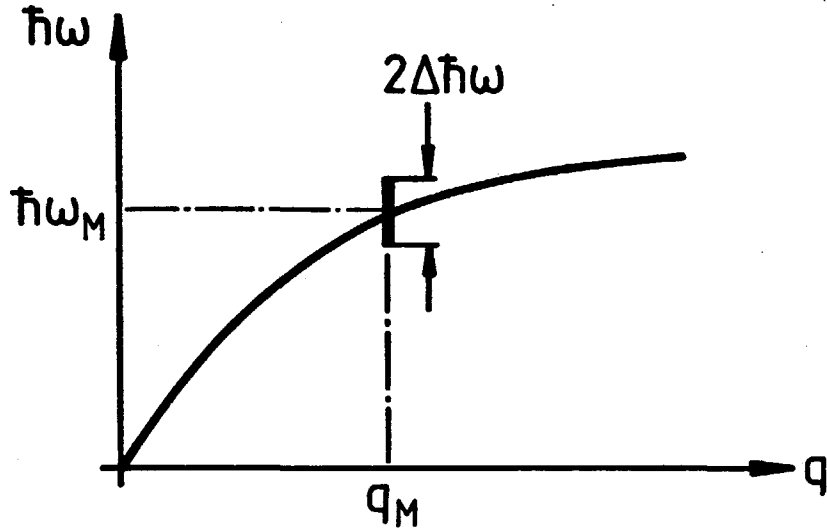


Fig. 3 Phonon dispersion curve and  $\hbar\omega$  deviation

The vibration amplitudes of the angular vibrations of the collimator and analyzing crystal will be determined in the following. A mismatch between desired and actual motion will be described causing  $q$  to change even less. Moreover, the entire change of the energy transfer of a scan will be assessed including information about the intensity gain.

## 2. Vibration Amplitudes

The scattering angle is

$$(3') \quad \varphi = \varphi_M + \Delta\varphi_0 \cdot \sin \omega t$$

and the Bragg angle on the analyzer crystal is

$$(4) \quad \theta = \vartheta_M - \Delta\varphi(t) + \Delta\vartheta(t),$$

in which

$$(5) \quad \Delta\vartheta(t) = \Delta\vartheta_0 \sin \omega t$$

and

$$(6) \quad \Delta\varphi(t) = \Delta\varphi_0 \sin \omega t.$$

It is advisable that the angular frequency  $\omega = 2\pi/T$  be equal to the pulse repetition frequency, i.e.  $2\pi/T$  with  $T = 10$  ms. The zero points in the time dependencies of  $\Delta k, \Delta\vartheta$  and  $\Delta\varphi$  are chosen so as to ensure that neutrons of wave number  $k_M$ , scattered at the angle  $\varphi_M$ , hit the analyzer at the Bragg angle  $\vartheta_M$  where they are reflected if their wave number is  $k_M'$ . For these reference values the phonon vector has the value  $q_M$  and the energy transfer is

$$(7) \quad \hbar\omega_M = E_i - E_f \text{ with}$$

$$E_i = \frac{\hbar^2}{2M} k_M^2 \text{ and } E_f = \frac{\hbar^2}{2M} k_M'^2.$$

The actual angular changes lead to an actual change of  $k'$  resulting from the Bragg equation on the analyzer crystal

$$(8) \quad \Delta k' = -k_M' \operatorname{ctg}(\vartheta_M) \cdot [\Delta\vartheta(t) - \Delta\varphi(t)].$$

By putting the required change in time at  $t = 0$  of  $\Delta\varphi$  from (1) equal to the actual change from (6),  $\Delta\varphi_0$  is determined as is  $\Delta\vartheta_0$  by equating (2) and (8).

The results are

$$(9) \quad \Delta\varphi_0 = \frac{1}{2\pi} \frac{T}{t_M} \frac{k_M}{k_M'} \sin \varphi_M$$

$$(10) \quad \Delta\vartheta_o = \Delta\varphi_o \left[ \frac{\operatorname{tg} \vartheta_M}{\lambda g \varphi_M} + 1 \right]$$

Both vibration amplitudes are of magnitude

$$(11) \quad \Delta\varphi_{o_{\max}} = \frac{1}{2\pi} \frac{T}{t_M} \frac{k_M}{k'_M}$$

For  $k'_M = k_M/2$ , one obtains  $\Delta\varphi_{o_{\max}} = 3.65^\circ$  if  $L = 50$  m and  $\lambda = 4$  Å. The vibration amplitudes are four times larger if  $L = 25$  m and  $\lambda = 2$  Å.

At  $\varphi_M = 90^\circ$ ,  $\Delta\varphi_o$  and  $\Delta\vartheta_o$  are of equal size. In the special case of  $\operatorname{tg} \varphi_M = -\operatorname{tg} \vartheta_M$ , entailing  $\varphi_M > 90^\circ$ , one gets  $\Delta\vartheta_o = 0$ . The analyzing crystal need not vibrate. It should be borne in mind that a rotation of the collimator by  $\Delta\varphi$  with the analyzing crystal at rest changes the Bragg angle by the same angular amount.

### 3. Mismatch

The vibration amplitudes were determined from the condition that the required and actual changes of  $\Delta\varphi$  and  $\Delta k'$  be equal at  $t = 0$ . Since the required change is linear in time, whereas the actual change is sinusoidal, there is a mismatch in the form of

$$(12) \quad \delta\varphi = \Delta\varphi_{\text{desired}} - \Delta\varphi_{\text{actual}} = \Delta\varphi_o (\omega t - \sin\omega t)$$

and

$$(13) \quad \delta\theta = (\Delta\vartheta_o - \Delta\varphi_o) [\omega t - \sin\omega t]$$

This is illustrated in Fig. 4.

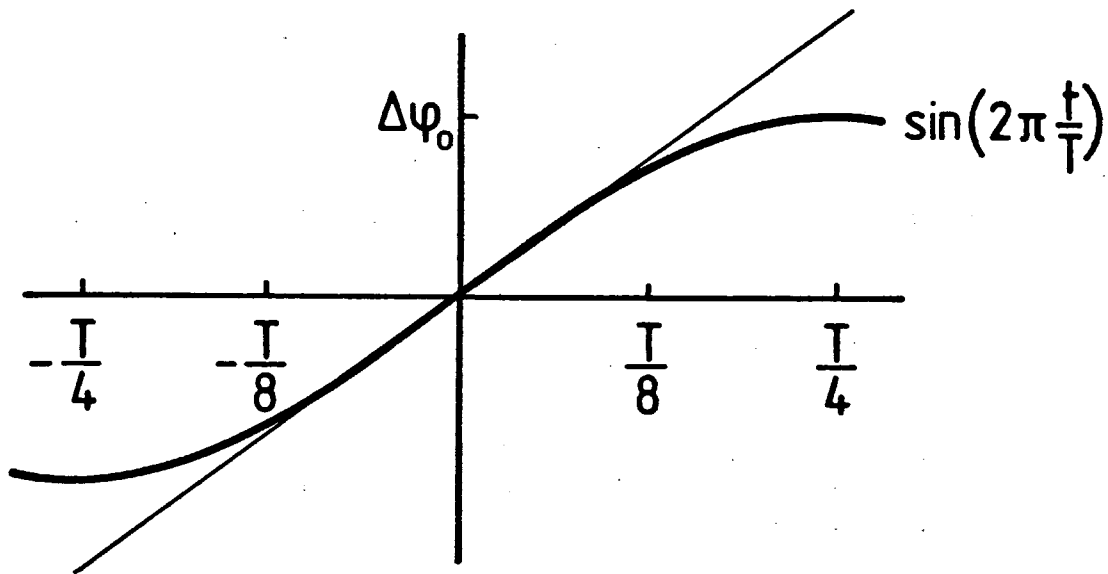


Fig. 4 Illustration of mismatch

For the time interval of the duration  $T/4 = 2.5$  ms, the deviation is sufficiently small, amounting to

$$\frac{\delta \varphi}{\Delta \varphi_0} = \frac{\delta \vartheta}{\Delta \vartheta_0} = 0.08$$

which means that  $\underline{q}$  may be regarded as constant over this time interval. The deviation from constancy is a deviation in modulus and in direction which depends on the length and direction of  $\underline{q}$  itself and is influenced, moreover, by  $k_M'$  and  $\vartheta_M$ .

#### 4. $h\omega$ -Change

The nominal energy transfer  $h\omega_M$  is given in (7). Its change during the scan can be calculated from

$$(14) \quad \frac{\Delta h\omega}{E_i} = 2 \frac{\Delta k}{k_M} \left[ 1 - \frac{k_M'}{k_M} \cos \varphi_M \right]$$

For the measuring time interval per pulse of  $[-T/8, T/8]$  derived from the mismatch, there results a change in the energy transfer of

$$(15) \quad 2\Delta h\omega = \frac{E_i}{2} \cdot \frac{T}{t_M} \left[ 1 - \frac{k_M'}{k_M} \cos \varphi_M \right].$$

This quantity must be related to the energy resolution of the spectrometer. It is essentially governed by monochromatization, i.e.

$$\delta E \approx 2 E_i \frac{\Delta t}{t_M}$$

In which  $\Delta t = 250 \mu s$  is the pulse width. The relative change of the energy transfer is therefore

$$(16) \quad \frac{2 \Delta h\omega}{\delta E} \approx \frac{T}{4 \cdot \Delta t} \left[ 1 - \frac{k_M'}{k_M} \cos \varphi_M \right]$$

$$\approx 10 \left( 1 - \frac{k_M'}{k_M} \cos \varphi_M \right)$$

The bracket is generally in the range of 1, it may be greater or smaller than 1 depending on  $\cos \varphi_M$ . Measurements therefore extend over approx. 10 half-widths. This is more than necessary if only the position and width of the phonon are of interest, even if a thorough background analysis is required. The situation is different for profile analyses.

## 5. Intensity Gain

The intensity profit of a pulsed source is evident. The spectrometer measures during the time interval  $[-T/8, T/8]$  per neutron pulse, i.e. over 2.5 ms. This means a nominal gain in intensity by a factor of 10 for the pulse width of 250  $\mu s$ !

## 6. Spectrometer Data

The most important spectrometer parameters are as follows:

|                                                             |   |                |
|-------------------------------------------------------------|---|----------------|
| distance between spectrometer and source                    | : | 25 - 50 m      |
| wavelength range                                            | : | 1 - 4 Å        |
| energy resolution $\delta E/E_i$                            | : | 1 - 8 %        |
| intensity gain at pulsed source                             | : | 10fold         |
| vibration amplitudes $\Delta\varphi_0$ and $\Delta\theta_0$ | : | 2 - 30°        |
| vibration frequency $\omega/2\pi$                           | : | 100 Hz         |
| relative change of energy transfer                          | : | 5 · $\delta E$ |

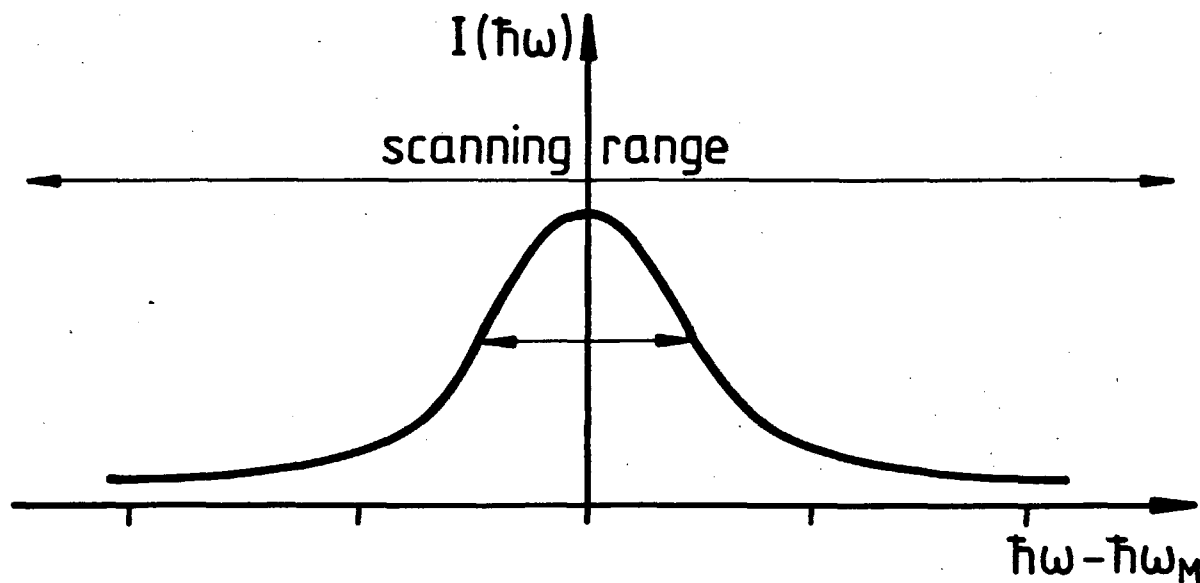


Fig. 5 Scan of a phonon

The spectrometer offers the particular advantage that any constant- $q$  scan without restriction can be performed at a continuous source similar to the conventional triple axis spectrometer. A disadvantage is the requirement of a large distance from the source, especially in those cases where high energy resolution is required in connection with short neutron wave lengths.

New development effort is required for the 100 Hz vibration arms for the analyzing crystal and the collimator. The Soller collimator must be short so that the crystal does not become too large. The technical difficulties do not appear excessive, however.

Fig. 6 shows a draft for the vibrating device of the collimator. A resonance frequency of 100 Hz is achieved with a rod of 1 cm radius and 1 m length for a moment of inertia formed by 10 kg moving mass on the 10 cm long rotating arm. The shear angle is  $\gamma = 1 \cdot 10^{-2} \cdot \Delta\phi_0$ . The stored energy is  $U = \frac{1}{2} M_K \cdot R_K^2 \cdot (\omega \cdot \Delta\phi_0)^2 = 20 (\Delta\phi_0)^2$  kWS. When the vibration decreases to  $e^{-1}$  after 1 s, a power of  $20(\Delta\phi_0)^2 \cdot [KW]$  must be applied. The assumption is based on a quality of  $Q \approx 300$ , which is a suitable value. Spring steels would probably require a higher  $Q$ . At  $\Delta\phi_0 = 20^\circ$ ,  $\gamma = 3 \cdot 10^{-3}$  in the elastic range, the power dissipation is 2 KW and considerably less for spring steel.

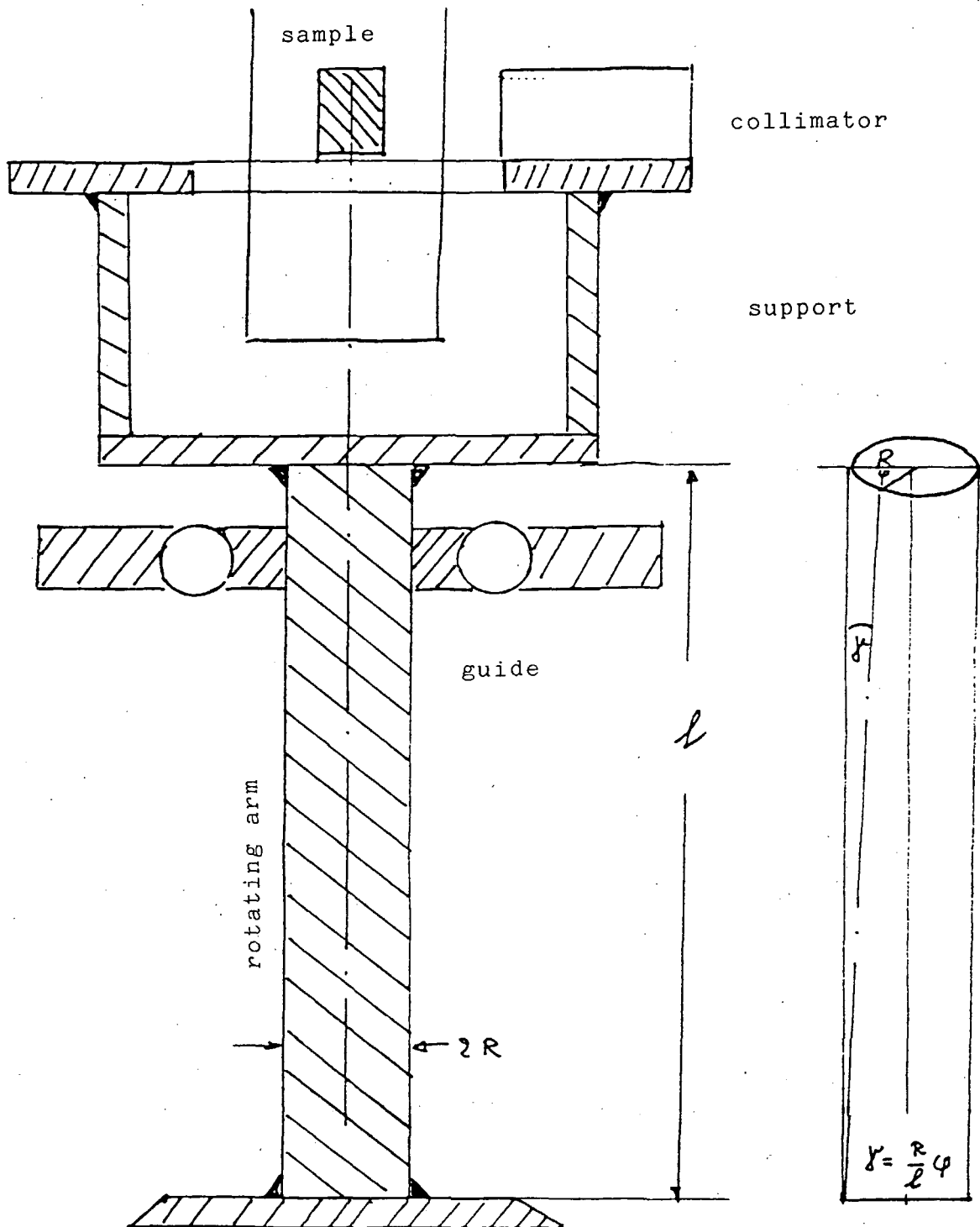


Fig. 6 Cross section of the rotating arm for the collimator rotational vibration



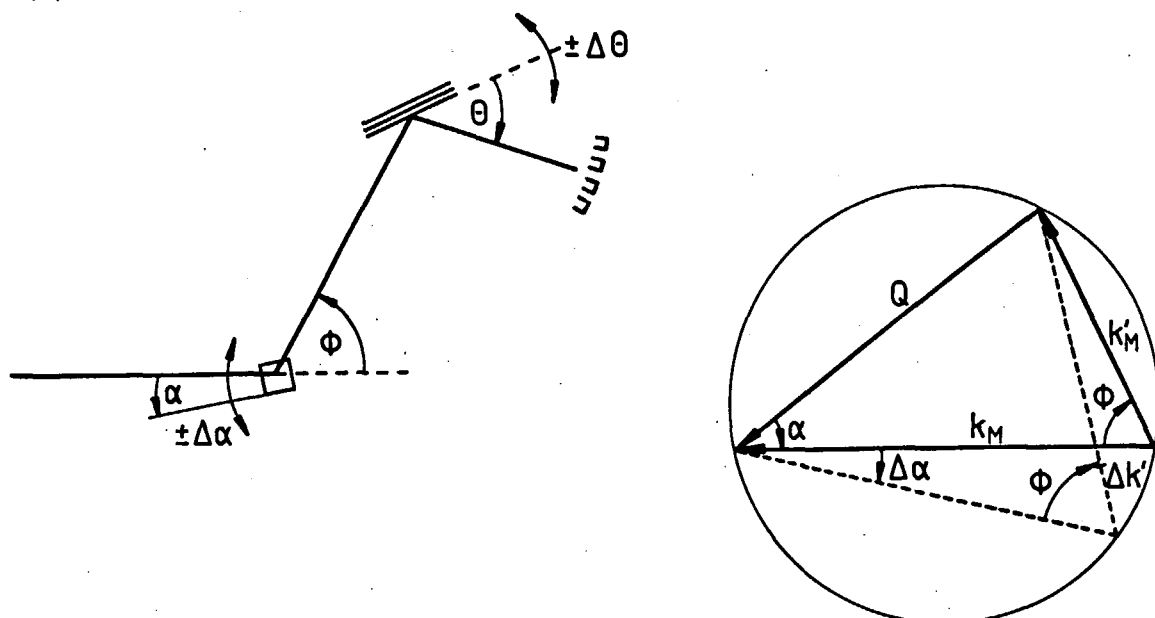
# Appendix to the paper "Constant-q Scan an der gepulsten Quelle"

by J. Schelten and B. Alefeld

## A Constant-Q Scan at Constant Scattering Angle

Colin Windsor, Bldg 418, MP & M Division, AERE, Harwell, OX11 0RA, UK

The above paper describes a pulsed source crystal analyser spectrometer capable of performing a short frequency scan at a chosen  $Q$  value. It operates by swinging the analyser angle and also the main arm scattering angle in a sinusoidal motion phased with the pulsed source. The swinging arm seems a major technical problem. This note describes an alternative based on a swinging sample angle  $\alpha(t)$  at a fixed scattering angle  $\phi$ . It still requires a swinging analyser angle  $\theta(t)$ .



To achieve a variable energy transfer  $\hbar\omega$  at fixed  $Q$  and fixed scattering angle  $\phi$  the scattering triangles in the sample axes must lie on the circle shown. The full line shows the median energy triangle  $k_M, k'_M$ , the dashed line one with the crystal angle displaced by  $\Delta\alpha$ . The corresponding changes  $\Delta k$  and  $\Delta k'$  are given from the cosine formula

$$\begin{aligned}
 Q^2 &= k_M^2 + k'_M{}^2 - 2k_M k'_M \cos \phi \\
 &= (k_M + \Delta k)^2 + (k'_M + \Delta k')^2 - 2(k_M + \Delta k)(k'_M + \Delta k') \cos \phi \\
 &= k_M^2 + k'_M{}^2 - 2k_M k'_M \cos \phi + 2k_M \Delta k + 2k'_M \Delta k' - 2k_M \Delta k' \cos \phi \\
 &\quad - 2k'_M \Delta k \cos \phi + O(\Delta k^2)
 \end{aligned}$$

$$\therefore \Delta k' = \frac{-\Delta k (k_M - k'_M \cos \phi)}{(k'_M - k_M \cos \phi)}$$

From the sine formula

$$\frac{\sin \phi}{Q} = \frac{\sin \alpha}{k'_M} = \frac{\sin(\alpha + \Delta\alpha)}{(k'_M + \Delta k')}$$

$$k'_M \sin \alpha + \Delta k' \sin \alpha = k'_M \sin \alpha \cos \Delta\alpha + k'_M \cos \alpha \sin \Delta\alpha$$

$$\Delta\alpha \approx \tan \alpha \frac{\Delta k'}{k'_M} \quad 2$$

As before the variation in the initial wavevector is trivially obtained from the change in time-of-flight

$$\Delta k = \frac{-k_M}{t_M} \Delta t \quad 3$$

The change in scattered wavevector must be made by swinging the analyser angle  $\phi$

$$\Delta k' = k'_M \cot \theta \Delta \theta \quad 4$$

Hence we have from equations 1 to 4

$$\Delta\alpha = \tan \alpha \cot \theta \Delta \theta \quad 5$$

$$\begin{aligned} \Delta \theta &= \frac{k_M}{k'_M} \frac{\Delta t}{t_M} \tan \theta \frac{(k_M - k'_M \cos \phi)}{(k'_M - k_M \cos \phi)} \quad 6 \\ &= \frac{\Delta t}{t_M} \tan \theta \frac{(k_M/k'_M - \cos \phi)}{(k'_M/k_M - \cos \phi)} \end{aligned}$$

Both angles are most conveniently swung in a sinusoidal motion with appropriate amplitude  $\Delta\alpha_0$  and  $\Delta\theta_0$  and phase with respect to the pulsed source. The median energy transfer and its shift are

$$\hbar\omega_M = \frac{\hbar^2}{2M} (k_M^2 - k'^2_M) = E_0 - E_1$$

$$\text{and } \Delta(\hbar\omega) = \frac{\hbar^2}{2M} (2k_M \Delta k_M - 2k'_M \Delta k')$$

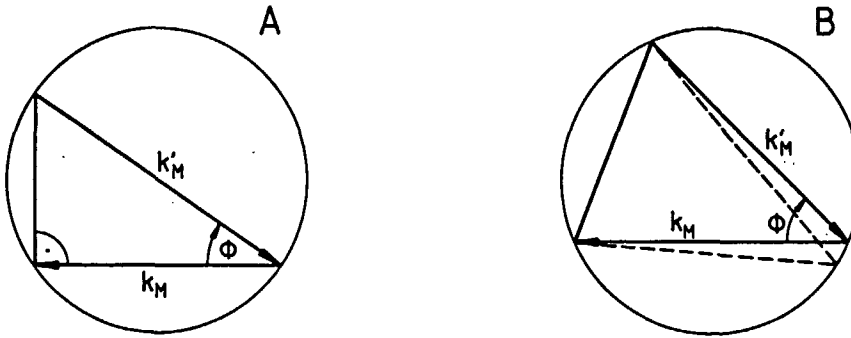
$$\begin{aligned} \frac{\Delta\hbar\omega}{E_0} &= \frac{2\Delta k_M}{k_M} - \frac{2k'_M}{k_M^2} \Delta k' \\ &= \frac{2\Delta k}{k_M} \left( 1 - \frac{k'_M}{k_M} \left( \frac{k_M - k'_M \cos \phi}{k'_M - k_M \cos \phi} \right) \right) \\ &= 2 \frac{\Delta t}{t_M} \frac{\cos \phi \left[ (k'_M/k_M)^2 - 1 \right]}{(k'_M/k_M - \cos \phi)} \quad 7 \end{aligned}$$

The time spread terms  $\Delta t/t_M$  may be eliminated to express the angular spreads  $\Delta\theta$  and  $\Delta\alpha$  directly in terms of the energy scan

$$\Delta\theta = \frac{\Delta h\nu}{E_o} \frac{\left[1 - k_M/(k'_M \cos \phi)\right]}{2(1 - (k'_M/k_M)^2)} \quad 8$$

$$\Delta\alpha = \frac{\Delta h\nu}{E_o} \tan\alpha \cot\theta \frac{\left[1 - k_M/(k'_M \cos \phi)\right]}{2(1 - (k'_M/k_M)^2)} \quad 9$$

here is clearly a problem here for scattering triangles where the numerator of equations 8 and 9 becomes small, that is  $k_M \simeq k'_M \cos \phi$ . Figure A



above illustrates this energy gain triangle which should not be too important in practice. There is similarly a problem when the denominator is small giving quasi-elastic scattering  $k_M \simeq k'_M$ . The angular amplitudes necessary for even a small energy scan are large as shown in B.

Consider a typical phonon scattering triangle with an energy transfer of 10 meV and a Q of  $3 \text{ \AA}^{-1}$  with half the incident energy lost. We assume Ge 220 planes ( $d = 2.00 \text{ \AA}$ ).

|                          |                                |                                |
|--------------------------|--------------------------------|--------------------------------|
| $h\nu = 10 \text{ meV}$  | $E_o = 20 \text{ meV}$         | $E_1 = 10 \text{ meV}$         |
| $Q = 3 \text{ \AA}^{-1}$ | $k_o = 3.107 \text{ \AA}^{-1}$ | $k_1 = 2.196 \text{ \AA}^{-1}$ |
| $\phi = 66.34^\circ$     | $\alpha = 42.10^\circ$         | $= 44.33^\circ$                |

The equations are all "well behaved" with these parameters.

$$\Delta\theta = -2.52 \Delta h\nu/E_o$$

$$\Delta\alpha = -2.33 \Delta h\nu/E_o$$

Thus a 2% energy scan requires a 5% angular scan of some  $2^\circ$  in  $\theta$  and  $\alpha$ .

A Simultaneous Energy Analysis of Scattered Neutrons by Means of the  
Renninger-Effect

A. Kollmar

SNQ-KFA Jülich

Principle

The energy analysis of scattered neutrons by Bragg-reflection from a single crystal suffers from the following facts:

1.

The reflectivity for the used reflection of a single crystal is reduced by the Renninger-effect. This means, that neutrons with the energy just analyzed are scattered by any other reflection lying on the Ewald-sphere into any other direction and do not hit the detector.

2.

Only neutrons of a certain energy, determined by Bragg-angle and used reflection, are detected. All other neutrons are lost.

3.

The reflectivity of single crystals is also reduced by phonon scattering.

These difficulties can be overcome to some extent if a cooled single crystal with any fixed orientation as analyser is almost entirely surrounded by a position sensitive detector.

Just by the Renninger-effect neutrons of any fixed energy will find one or more reflections corresponding to certain scattering-directions and are detected in the position sensitive detector. The distribution of the neutrons into these specific reflections is characteristic for their energy and can be used for the energy analysis. The figure shows the principle.

### Continuous Source

Neutrons scattered out of the monochromatic beam by the sample fall onto the analysing crystal of a certain mosaicity. Nearly all neutrons with different energies will be Bragg-reflected by reflections characteristic for their energy.

A calibration procedure will be necessary, that means that measurements of the distribution of the neutrons of known and sufficiently sharp energy into the various reflections have to be done, in steps of an appropriate small energy difference. The spectrum of the scattered neutrons from the sample will be composed out of these measured calibration functions by a linear fitting procedure of a dedicated computer. The energy analysis is simultaneous for all energies.

### Restrictions

Restrictions for this method are:

- For very broad and flat spectra the fitting procedure will not give precise results.
- The measurement of collective excitations will be difficult.
- Only neutrons above the Bragg-cut-off can be detected.
- Neutrons with a small wavevector will not find a sufficient number of possible reflections to cover more or less continuously the energy-scale. For the example of a Beryllium-crystal the average energy distance (in meV) between two reflections is  $dE = 7,32/\sqrt{E}$ . The total number of reflections within the Ewald-sphere is  $N = 0,091 E^{3/2}$ . This perhaps can be overcome by composing the crystal out of several pieces with different orientations or lattice parameters.
- For neutrons with large wavevectors there will be too much reflections which overlap in the position sensitive detector.

- Mosaic spread and necessary thickness of the crystal can be adapted only for a more or less broad energy range.
- The solid angle for the neutrons scattered from the sample will be restricted. Otherwise the dimension of the detector will be unreasonable.
- Because of geometrical hindrance only a small number of analysers could be installed around the sample.
- Because in general the reflections occur out off the measuring plane also the vertical collimation before the crystal influences the resolution.
- Small deviations of the direction of the beam onto the crystal from the nominal direction for the calibration and also sample size effects may cause inaccuracies.

#### Example

To overlook roughly the situation the simulatunously occuring reflections of a Beryllium single crystal ( $1,3 \times 0,7 \times 0,5 \text{ cm}^3$ , mosaic spread  $\eta = 0,4^\circ$ ) had been measured on the D9 diffractometer at the Institute Laue-Langevin. To see the change of the relative intensities of the different reflections this has been done for two wavelength of the incoming neutrons,  $\lambda_1 = 0,5175 \text{ \AA}$  ( $\hat{=} 305,4 \text{ meV}$ ) and  $\lambda_2 = 0,5306 \text{ \AA}$  ( $\hat{=} 290,5 \text{ meV}$ ), with a resolution  $\Delta E/E$  of about 5 %.

The crystal had been at the sample position with its 001 direction nearly parallel to the incoming beam. 2- $\theta$ -scans for the detector have been made from  $5^\circ$  to  $77^\circ$ . For detecting also the reflections which, at a fixed orientation of the crystal, do not lie in the measuring plane, these 2- $\theta$ -scans have been repeated for several  $\chi$ -angles of the Eulerian cradle between  $0^\circ$  and  $90^\circ$ , in small enough steps as not to miss a reflection. Because the  $\omega$ -angle was  $0^\circ$  by this the crystal was turned around the axis of the incoming beam so the scanned solid angle was about  $4\pi/10$ , naturally with some inaccuracies due to resolution effects.

The measured integrated intensities are listed in the table. The change with the incoming wavelength is significant. That equivalent reflections (with respect to the 001 direction, like for example Nr. 1,8 or 3,7,10) have so different intensities is due to a misorientation of the 001 direction out of the incoming beam direction of about  $1,5^\circ$ . For the strong reflection Nr. 1 the reciprocal lattice point lies, in a rather equal distance, just within or without the Ewald-sphere for  $\lambda_1$  or  $\lambda_2$  respectively.

The sum of counts for all reflections in the scanned solid angle (only about  $1/10$  of  $4\pi$ ) is about 50000/10000 monitor. To compare the method with the 3-axis performance a  $\theta$ - $2\theta$  scan with the  $02\bar{1}$  reflection had been done. The peak intensity was also about 50000 counts/10000 monitor. That means that with a position sensitive detector covering nearly the whole solid angle 4 much more neutrons would be counted at the same time. The factor may be in between 5 and 10. The effective medium resolution will be much better than for the  $\theta$ - $2\theta$ -scan with the  $02\bar{1}$  reflection because all the others contributing reflections have a larger Bragg-angle  $\theta$ . If one takes into account that for the determination of the incoming spectrum (in this case position and width of the Gaussian line) several points of a  $\theta$ - $2\theta$ -scan have to be measured, then with the position sensitive detector a large gain in measuring time for the same accuracy of the measured spectrum can be expected.

#### Pulsed Source

In this case the spectrometer would resemble a time of flight spectrometer with inverse geometrie, with the sample in the direct beam. Also the time-dependence of all reflections has to be measured. The total time resolution defined by pulse width, length of primary flight path etc, has to be adapted to the medium crystal resolution. The spectrometer can be regarded as a MAX-type spectrometer where at one scattering angle the time of flight spectra of many reflections and by this with different secondary resolution are measured simultaneously. There is not only one secondary energy-window (together with higher orders) but there is a broad energy band more or less tightly covered by the energy windows corresponding to all reciprocal lattice points lying between two Ewald spheres, the radius of which is given by a reasonable lower and upper limit.

With a calibration, for example with Vanadium, also the relative intensities of the different time of flight spectra then measured with the sample can be determined. This increases the accuracy for any fitting procedure.

By the time of flight analysis also reflections with different energies but which may overlap in the position sensitive detector and also higher order reflections may be resolved, for not too broad incoming spectra.



TABLE

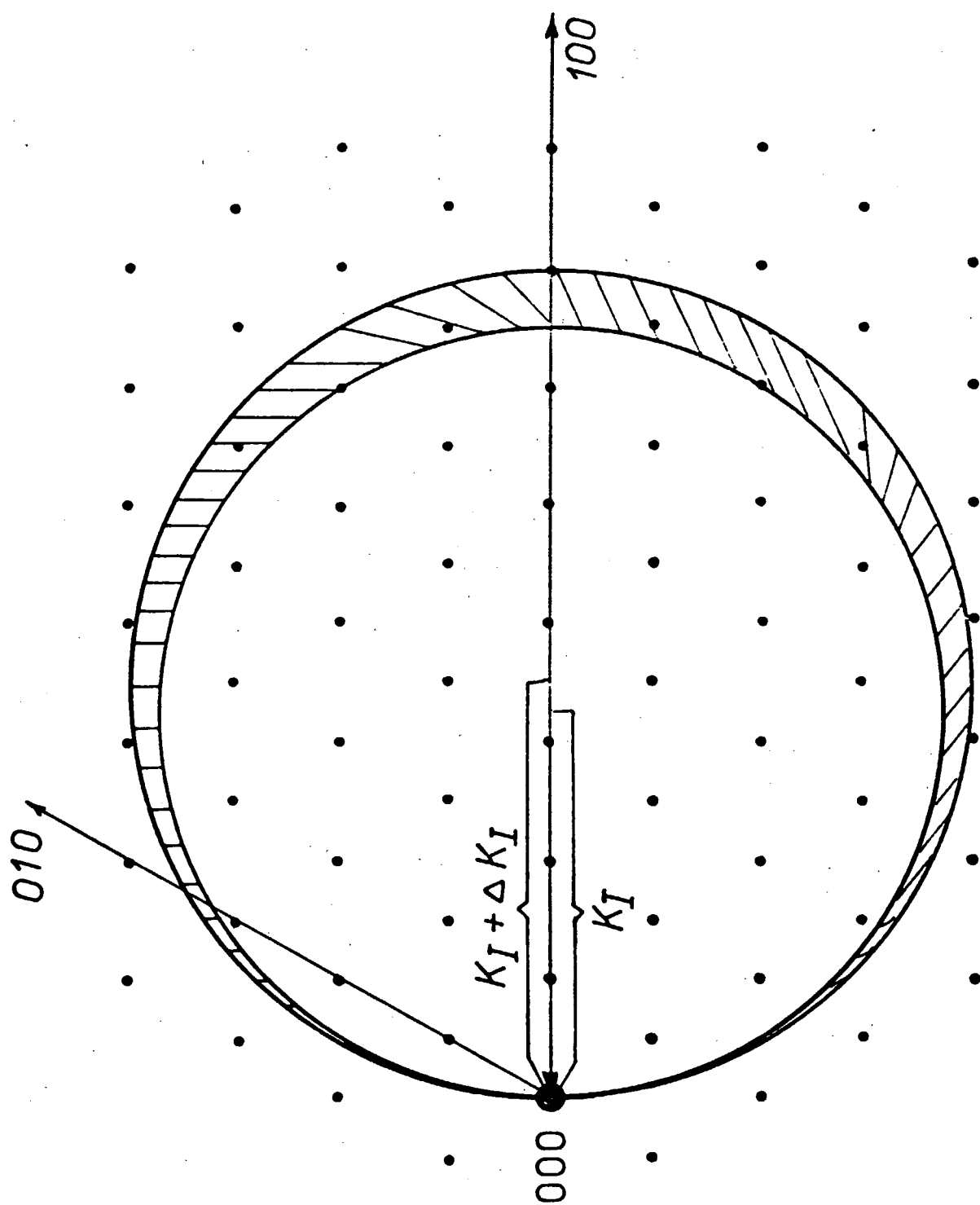
| Nr. | h k l                 | $\lambda_1=0,5175$ | $\lambda_2=0,5306$ | $2\theta$ | $ SF ^2$ |
|-----|-----------------------|--------------------|--------------------|-----------|----------|
|     |                       | I                  | I                  |           |          |
| 1   | 0 2 $\bar{1}$         | 25000              | 24200              | 32,1      | 3        |
| 2   | 1 6 $\bar{7}$         | 650                | 300                | 59,6      | 3        |
| 3   | 1 3 $\bar{5}$         | 800                | 4100               | 75,5      | 3        |
| 4   | 1 2 $\bar{2}$         | 700                | 7300               | 45,1      | 1        |
| 5   | 2 2 $\bar{4}$         | 7500               | 580                | 65,9      | 4        |
| 6   | 2 1 $\bar{2}$         | 7500               | 5000               | 44,8      | 1        |
| 7   | 3 1 $\bar{5}$         | 5500               | 2600               | 74,2      | 3        |
| 8   | 2 0 $\bar{1}$         | 450                | 260                | 32,1      | 3        |
| 9   | 6 0 $\bar{5}$         | 1000               | 260                | 50,8      | 0        |
| 10  | 4 $\bar{1}$ $\bar{5}$ | 7000               | 200                | 76,9      | 3        |
| 11  | 3 $\bar{1}$ $\bar{2}$ | 1300               | 100                | 44,5      | 1        |
| 12  | 4 $\bar{2}$ $\bar{4}$ | 170                | -                  | 64,4      | 1        |
| 13  | 4 $\bar{2}$ $\bar{5}$ | -                  | 100                | 75,0      | 3        |

h k l = indices of reflection

I = counts/10000 monitor ( $\approx$ ca. 1 sec)

$2\theta$  = measured  $2\theta$  of reflection (medium value for both wavelength)

$|SF|^2$  = square of geometrical structure factor for the reflection



Reciprocal lattice of Be  
(without mosaic spread)

fig.

Comparison of Time-of-Flight Methods and Crystal Spectrometer  
Methods at the SNQ for Studies of Collective Excitations

W. Reichardt  
Kernforschungszentrum Karlsruhe  
Institut für Nukleare Festkörperphysik  
P.O.B. 3640, D-7500 Karlsruhe, FRG

Abstract

We compare the performance of a conventional triple axis spectrometer with that of a direct geometry hybrid time-of-flight spectrometer when placed at identical beam tubes of the SNQ. Assuming identical resolutions for both instruments we find, that in the thermal region the intensities at the detectors are about the same for the SNQ with the compressor ring, whereas the triple axis spectrometer is superior by a factor of two for the version without compressor ring. In view of other well established advantages of the crystal spectrometer method we conclude, that also at the pulsed source SNQ the triple axis spectrometer will be the adequate instrument for studies of selected collective excitations.

Time-of-flight spectroscopy will be superior for high neutron energies, if the compressor ring is installed, and for those types of experiments which profit from the use of multi-detectors, at all energies.

## 1. INTRODUCTION

At stationary neutron sources the measurements of collective excitations using single crystal samples have been a domain of triple axis spectrometers (TAS). There are many reasons for the superiority of the crystal spectrometer method over the time-of-flight method, among which are:

1. Restriction of the measurement to a limited range in energy  $\hbar\omega$  or momentum transfer  $\vec{Q}$ , which can be estimated in advance in most cases. Possibility to repeat individual data points, which seem to be uncertain, or to extend scans, which had not been planned correctly.
2. High flexibility of the TAS for optimizing the measurement with respect to resolution and intensity (e.g. easy changes of collimators, monochromator- and analyzer crystals, application of focussing methods).
3. Possibility to select high symmetry directions for the measurement and to choose special scanning modes like const. Q-scans or const. E-scans.

While item 1 causes a reduction in measuring time that is calculable, the advantages mentioned in 2. and 3. are not so easily estimated on a quantitative basis. Because of the complexity of present experiments - the easy ones have already been dealt with - these advantages have a strong weight.

There are also some disadvantages, which are largely absent in TOF-spectroscopy; as for instance the occurrence of spurious and difficulties, if absolute intensities have to be determined. Very often spurious can be avoided by a careful

planning of the experiment. When the TAS uses a pulsed beam they can be eliminated almost completely, if the detector is suitably gated.

The future German neutron source SNQ has a pulsed time structure, where the peak flux  $\hat{\phi}$  is about 30 times ( $\sim 60$  times with compressor ring) higher than the time averaged flux  $\bar{\phi}$ . Therefore the question arises, whether at this source the TAS is still competitive.

A general comparison between the crystal spectrometer method and TOF spectrometry, that is valid for all cases, is certainly impossible. Each experiment has very special requirements. Therefore we want to present a special comparison, which is rather simple and may be easily extended to other cases.

## 2. COMPARISON OF INTENSITIES FOR TAS AND X-TOF AT A PULSED SOURCE

We compare the measurement of a single excitation  $\hbar\omega(\vec{Q})$ , performed on the following spectrometers

- a. a conventional TAS
- b. a hybrid TOF-spectrometer of type X-TOF with a single detector (selection of the primary energy by a monochromator crystal, determination of the energy distribution of the scattered neutrons by time-of-flight).

The two instruments use identical beams of the SNQ and have identical primary spectrometer parts. This implies that the intensities at the sample and the primary resolution in energy and angle are the same for both cases. For the TOF-spectrometer the area of the detector and the distance between sample and detector will be determined by the requirement, that also the secondary resolutions in energy and angle are identical to that of the TAS.

We measure the scanning length on the TAS in units of the energy resolution ( $n \cdot \Delta E_{\text{FWHM}}$ ), which is identical for both instruments. According to experience the average scanning length in these units is about 3. Of course, this refers to cases, where the width of the excitation is small compared to the resolution of the spectrometer, but most of the todays measuring problems are in this category.

We arrive at the following intensities I:

$$\begin{aligned} a = \text{TAS} & : I_{\text{TAS}} \sim R_{\text{An}} \epsilon_{\text{TAS}} / n \\ b = \text{TOF} & : I_{\text{X-TOF}} \sim \epsilon_{\text{X-TOF}} \end{aligned}$$

or

$$\frac{I_{\text{X-TOF}}}{I_{\text{TAS}}} = \frac{n \epsilon_{\text{X-TOF}}}{R_{\text{An}} \epsilon_{\text{TAS}}} \quad (1)$$

Here  $R_{\text{An}}$  is the reflectivity of the analyzer crystal and  $\epsilon$  are the sensitivities of the detectors.

A more accurate comparison should take into account, that the collimators of the TAS in the analyzer system have transmissions  $< 1$ . On the other hand the Gaussian shape of the resolution function of the TAS has a higher statistical value than the asymmetric resolution function (step function plus exponential decay) of the TOF spectrometer (see section 6). These two effects are not crucial and largely compensate each other. Therefore they will be neglected.

If we assume  $R_{\text{An}} = .5$ ,  $n = 3$  and  $\epsilon_{\text{TAS}} = \epsilon_{\text{X-TOF}}$  we obtain

$$I_{\text{X-TOF}} = 6 I_{\text{TAS}} \quad (2)$$

This relation, however, is only valid, if the width of the source pulse is sufficiently narrow, that one can obtain the resolution, which is required for the experiment.

If we have to tailor the pulse by a tail-cutter in order to reduce the pulse width by a factor  $\alpha_{\text{TC}}$  the new comparison reads

$$\frac{I_{X-TOF}}{I_{TAS}} = \frac{n \cdot \epsilon_{X-TOF} \cdot \alpha_{TC} \cdot T_{TC}}{R_{An} \epsilon_{TAS}} \quad (3)$$

( $T_{TC}$  is the transmission of the tail-cutter).

For the exponentially decaying pulse of the SNQ in the thermal range  $\alpha_{TC}$  is given to a good approximation by

$$\alpha_{TC} = \lambda \cdot \Delta t_{TC}, \quad (4)$$

where  $\lambda$  is the decay constant of the moderator and  $\Delta t_{TC}$  is the burst width of the chopper, that is used to reduce the pulse-width.

In these considerations we have neglected the background. We have learnt from existing instruments, that the major part of the background at the detector of a well shielded TAS arises from the sample itself. Because of the good filter characteristic of analyzer crystals with respect to neutrons of wrong energy, and because in phonon experiments the slowly varying background mostly is temperature dependent roughly in the same way as the phonon peak itself, we have to conclude, that this background arises mainly from inelastically scattered neutrons (incoherent scattering, multi-phonon processes, multiple scattering, Bragg-phonon scattering), whose energy correspond to the reflection energy of the analyzer.

Therefore we expect, that

- a. the background of a TAS is largely independent of the time behaviour of the neutron flux (stationary or pulsed). This does not apply to spurious caused by elastic scattering processes.
- b. The backgrounds of a TAS and a TOF spectrometer at the SNQ will not be very different.

### 3. THE RANGE OF THERMAL NEUTRON ENERGIES

$$(10 \leq E_0 \leq 100 \text{ meV})$$

For neutron energies between 10 and 100 meV the neutron pulse of the water moderator of the SNQ with compressor ring can be well described by step function with exponential decay

$$\phi(t) = \hat{\phi} e^{-\lambda t} \quad t \geq 0 \quad (5)$$

The lifetime of the neutrons  $\tau = 1/\lambda$  is about 140  $\mu\text{s}$ .

Routine measurements of dispersion curves are often performed in neutron energy loss scattering with resolutions of the order  $\Delta\hbar\omega/\hbar\omega = 10\%$ .

If we introduce  $m = E_i/E_f$  ( $E_i$  = primary energy,  $E_f$  = secondary energy) and postulate that  $\Delta E_i = \Delta E_f = \frac{1}{\sqrt{2}} \Delta\hbar\omega$  (because of  $\Delta\hbar\omega = (\Delta E_i^2 + \Delta E_f^2)^{1/2}$ ), we obtain the following relations for the TOF spectrometer of type X-TOF

$$\begin{aligned} \frac{\Delta E_i}{E_i} &= \frac{m-1}{\sqrt{2} \cdot m} \frac{\Delta\hbar\omega}{\hbar\omega} \\ \Delta E_i &= \frac{m-1}{\sqrt{2}} \frac{\Delta\hbar\omega}{\hbar\omega} = 2 \frac{\Delta t_f}{t_f} = 2 \Delta t_f \cdot \frac{v_f}{L_2} \end{aligned} \quad (6)$$

$\Delta t_f$  is the required time resolution at the sample,  $t_f$  is the flight time between sample and detector (path length  $L_2$ ), and  $v_f$  is the velocity of the scattered neutrons.

Due to the distance between source and sample  $L_1$  ( $\approx 10$  m) the source pulse of width  $\Delta t_p$  will be broadened. The resulting width  $\Delta t_s$  at the sample is

$$\Delta t_s = (\Delta t_p^2 + (\frac{1}{2} \frac{\Delta E_i}{E_i} \frac{L_1}{v_i})^2)^{1/2} \quad (7)$$

In order to obtain the required resolution  $\Delta t_f$  must be equal to  $\Delta t_s$ , which leads to a condition for the secondary flight path  $L_2$



$$L_2 = 2\sqrt{2} v_f \cdot \frac{(\Delta t_p^2 + (\frac{1}{2\sqrt{2}} \frac{L_1}{v_f} \frac{m-1}{m} \frac{\Delta \hbar\omega}{\hbar\omega})^2)^{1/2}}{(m-1) \frac{\Delta \hbar\omega}{\hbar\omega}} \quad (8)$$

Fig. 1 shows the pulse width  $\Delta t_s$  at the sample and the required flight-path  $L_2$  as a function of  $m = E_i/E_f$ . The parameters are  $\Delta t_p = 140 \mu s$ ,  $\hbar\omega = 25 \text{ meV}$ ,  $\frac{\Delta \hbar\omega}{\hbar\omega} = .1$ . For the SNQ without compressor ring ( $\Delta t_p \approx 300 \mu s$ ) the values of  $L_2$  have to be multiplied by a factor of about 2.

The most valuable region is  $m \leq 2$  ( $E_i \leq 2E_f$ ) because it gives the largest freedom in the choice of the momentum transfer  $Q$ . For measurement of magnetic excitations and low frequency acoustic phonons  $m$  usually has to be much smaller than 2. In measurements of optical phonons frequently  $m \approx 2$  is used.

The flight paths calculated in the above example for  $m \leq 2$ , namely  $L_2 \geq 10 \text{ m}$  and  $L_2 \geq 20 \text{ m}$  without compressor ring, are not practicable, as scattering angles between  $\sim 0^\circ$  and  $\sim 120^\circ$  have to be used in the experiments. A reasonable upper limit is  $L_2 = 4 \text{ m}$ .

One has further to consider that only a moderate energy resolution was assumed in our example. For improved resolution even longer flight-paths would be necessary.

This demonstrates that in most cases TOF experiments with thermal neutrons need a tailcutter to reduce the width of the source pulse. In addition the use of a tail-cutter will improve the flexibility of a TOF spectrometer and lead to better pulse shapes.

Thus the gain in intensity calculated for a pulsed source with very narrow pulses ( $\sim$ factor 6) cannot be reached with the SNQ in the range of thermal neutrons. This applies already to experiments with moderate resolution and even more if high resolution is required, unless new aspects arise by special methods

like the Fourier methods or the correlation technique.

Using  $\alpha_{TC} = .25$  ( $\Delta t_p \approx 35 \mu s$ ) and  $T_{TC} = 2/3$  we obtain from Eq. 3

$$I_{X-TOF} = I_{TAS}$$

For this case, which is based on the above example, the TOF spectrometer does not yield a gain in intensity compared to the TAS.

Near the upper limit of the thermal region ( $E_0 \approx 100$  meV) the time resolution must be much better than  $\Delta t_p = 35 \mu s$ . Therefore the comparison of the intensities gets even more disadvantageous for the TOF spectrometer. However, one has also to consider, that the reflectivities of the analyzer crystals of the TAS decrease with increasing neutron energy.

The above example shows also, that for special cases, if  $m \gg 3$ , the flight paths are in a reasonable range. A X-TOF spectrometer with a flight path of 4 m can be very attractive, if the experiment allows low final energies  $E_f$ . Although its applications are very limited, the intensity is very high. A possible field of application is the measurement of optical phonons in small sample. It is not suited for small energy transfers, as the broadening of the neutron pulse due to the large distance between source and sample leads to an unsatisfactory resolution. It is evident, that also the two other types of TOF spectrometers: TOF-X=inverted geometry hybrid spectrometer and TOF-TOF=chopper spectrometer can use the source pulse without tailoring if the primary energies are very low. This implies that only neutron energy gain scattering is possible and therefore experiments with samples at low temperatures are excluded. In case of a chopper spectrometer the burst width of the chopper must be considerably smaller than the width of the moderator pulse in order to allow a good time resolution.

This analysis has shown that in the thermal region ( $10 \text{ meV} < E_i < 100 \text{ meV}$ ) a TAS will be preferred to a TOF spectrometer also at the SNQ, if one wants to study selected excitations. This applies in particular to experiments with high resolution. As the TOF spectrometer provides no gain in intensity, the advantages of the TAS mentioned in the introduction will speak in favor of this instrument.

TOF spectroscopy is of interest only in those cases, where the moderator pulse can be used without tailoring. There are a few cases of this type:

- a.  $h\omega \gg E_f$  and  $E_f$  small, for energy loss scattering
- b.  $h\omega \gg E_i$  and  $E_i$  small, for energy gain scattering.

The number of experiments, which can tolerate these restrictions, is not very large.

#### 4. NEUTRON ENERGIES BETWEEN 300 meV AND 1 eV

Above 300 meV, the pulse width of the water moderator is given by

$$\Delta t_p = 2/\sqrt{E} \text{ } \mu\text{s} \quad (E \text{ in eV}), \quad (9)$$

if the proton pulse is compressed by the compressor ring. Time resolutions below 1% become possible with a flight path of 4 m. Therefore we can apply relation (1). If we take further into account, that the reflectivities of the crystal monochromators are considerably reduced at these high energies (we shall assume, that  $R_{An} < 1/3$ ), we arrive at

$$I_{X\text{-TOF}} > 9 I_{\text{TAS}}.$$

According to Carlile et al. (1) in this energy region a TOF spectrometer of type TOF-TOF is superior in intensity to the X-TOF spectrometer, which we have considered so far. Assuming an average gain factor of about 2 we obtain

$$I_{\text{TOF-TOF}} > 20 I_{\text{TAS}}$$

As experiments at high neutron energies always suffer from low intensities, the large intensity gain will always favor the TOF spectrometer.

Without compressor ring the pulse width is about 250  $\mu\text{s}$  for all neutron energies. Cutting down the source pulse to the necessary width of  $\Delta t_p < 5 \mu\text{s}$  will reduce the intensity to such an extent, that the application TOF spectroscopy becomes impossible.

## 5. SPECTROMETERS WITH MULTI-DETECTORS

One has to distinguish two cases:

- a. Several detectors or analyzer-devices are placed close together. Usually they can be positioned only as a whole. Such a system of small angular range probes one dispersion surface in a small range of momentum transfers  $Q$  around a selected midpoint. Devices of this kind with about 10 detectors are very easy to build for the spectrometer types X-TOF and TOF-TOF. In fact, they are necessary to allow interpolation onto a desired direction in reciprocal space. The maximum gain is obtained for flat dispersion surfaces. Triple axis spectrometers allow a similar extension of the  $Q$ -range with Rowland focussing at the analyzer or Multi-detectors on a MARX-spectrometer.

- b. Use of multi-detectors, which cover a large range of scattering angles (about  $100^\circ$ ). Examples are the IN4 at the ILL or the MAX-spectrometer at KENS. Large multi-detectors are used, if one wants to investigate simultaneously different dispersion sheets in different Brillouin zones, that is to say, in survey studies of the dispersion relations.

In experiments of this kind one has to consider the following restrictions: It is not possible to achieve favorable resolution and focussing for all detectors. The smallest counting rate in a multi-detector determines the duration of the experiment. Therefore simultaneous measurements of excitations with very different cross-sections are unreasonable.

Further details about the use of a multi detector for measurements of collective excitations in single crystals are given in a separate report (2).

## 6. RESOLUTION OF TIME OF FLIGHT EXPERIMENTS

In time-of-flight experiments at a pulsed source we have to consider essentially three types of resolution functions:

- a. The triangular transmission function of a chopper.
- b. The time dependence of the slowing down spectrum for the epithermal region, which for a homogeneous moderator is given by

$$\phi(v,t) = (\Sigma_S vt)^2 e^{-\Sigma_S vt} \quad (12)$$

( $\Sigma_S$  = scattering cross section of hydrogen).

- c. The time dependence of the thermal spectrum which can be approximated rather well by a step function at  $t = 0$  and an exponential decay characterized by the decay constant  $\lambda$  or the mean emission time  $\bar{t} = 1/\lambda$ .

In the table below we compare several frequently used quantities of those three resolution functions.

1. The mean emission time  $\bar{t}$  (all pulses start at  $t = 0$ ).
2. The full width half (quarter) maximum  $t_{1/2}$  ( $t_{1/4}$ ).
3. The variance, which is defined by

$$\Delta^2 = \overline{t^2} - \bar{t}^2 \quad (13)$$

This quantity is of primary importance for time-of-flight experiments, because it enters into the leading term of the resolution correction for the time-of-flight distribution  $Z(t)$  according to

$$Z(t))_{\text{true}} = Z(t))_{\text{measured}} + A \cdot \Delta^2 + \text{higher order terms} \quad (14)$$

4. The fraction of the intensity in the pulse outside  $t_{1/2}$ .

|                       | $\bar{t}$                      | $t_{1/2}$                                      | $t_{1/4}$              | $\Delta$                                         | relative intensity |                   |
|-----------------------|--------------------------------|------------------------------------------------|------------------------|--------------------------------------------------|--------------------|-------------------|
|                       |                                |                                                |                        |                                                  | outside $t_{1/2}$  | outside $t_{1/4}$ |
| chopper               | $t_{1/2}$                      | $\bar{t}$                                      |                        | $.408t_{1/2}$                                    | .25                |                   |
| slowing down spectrum | $\approx \frac{1.8}{\sqrt{E}}$ | $1.133\bar{t}$<br>$\approx \frac{2}{\sqrt{E}}$ |                        | $.510t_{1/2}$                                    | .26                |                   |
| thermal spectrum      | $\frac{1}{\lambda}$            | $\frac{.693}{\lambda}$                         | $\frac{1.39}{\lambda}$ | $\frac{1}{\lambda}, 1.44t_{1/2}$<br>$.72t_{1/4}$ | .50                | .25               |

From this table it is obvious that the resolution function of the chopper is the most valuable one, although there is no pronounced difference compared with the slowing down spectrum. It is further demonstrated, that it is unreasonable to use  $t_{1/2}$  of the thermal pulse for an estimation of the time resolution. We must conclude that the time uncertainties  $\Delta t = t_{1/2}$  for a chopper and  $\Delta t = t_{1/4} = 1.39/\lambda$  are corresponding quantities.

It has been pointed out before, that the pulse width of the fast moderator at the SNQ is too large for most of the time-of-flight experiments with thermal neutrons. Reduction of the pulse width may be achieved either by a chopper close to the moderator or by poisoning the moderator with neutron absorbers. Poisoning the moderator will essentially leave the peak intensity unchanged and decrease the mean emission time  $1/\lambda$ . In order to obtain a desired time uncertainty  $\Delta t$  the two methods will lead to the following reduction factors for the intensity in the pulse

$$\begin{array}{ll} \text{chopper} & : \Delta t \cdot \lambda \cdot \alpha_{TC} \quad (\alpha_{TC} = \text{transmission of the chopper}) \\ \text{poisoning} & : \Delta t \cdot \frac{\lambda}{1.39} \end{array}$$

With  $\alpha_{TC} \sim .7$  both methods will yield similar intensities. As poisoning does not give advantages concerning the intensity, tailoring the pulse with choppers should be preferred, because this allows to adjust the pulse width individually for each spectrometer or even for each experiment.

## 7. CONCLUDING REMARKS

In this report the application of triple axis spectrometers and time-of-flight spectrometers at the SNQ to measurements of collective excitations in single crystals was studied. It was shown that in the thermal region the TAS is competitive to TOF spectrometers concerning intensity, if selected excitations are investigated. Therefore, also at the SNQ the TAS will be the appropriate instrument for experiments of this kind. Experiments which make optimum use of TOF spectrometers with multi-detectors are for instance:

1. Survey experiments with the aim to probe the whole dispersion relation.
2. Measurements of collective excitations in samples with large, often strongly structured, incoherent background (Hydrogen-compounds, Vanadium-compounds).

3. Studies on systems with a large coupling between magnetic excitations and phonons. Here the measurements often need to cover large regions in  $Q$  and  $h\omega$ .
4. Investigation of collective excitations, where the linewidth is much larger than the resolution of the spectrometer (defect structures).
5. Measurements of optic phonons in compounds with large differences in the atomic masses of the constituents. In these systems the optic branches usually are flat and fluctuations of the dynamical structure factor as a function of  $Q$  are not very pronounced.
6. Experiments, where absolute intensities have to be determined.

In the range of neutron energies above 300 meV the intensities of TOF spectrometers are by far superior to those of triple axis spectrometers, that they largely overcompensate the other advantages of triple axis spectrometers. This, however, applies only to the SNQ with compressor ring.

The range of neutron energies between 100 meV and 300 meV has not been dealt with in this report, because no quantitative information exists about the pulse structure of the moderator. The width of the moderator pulse changes from 5  $\mu$ s at 300 meV to about 140  $\mu$ s at 100 meV at the SNQ with compressor ring. Therefore TOF spectroscopy is not possible without the use of tailcutters, which causes a considerable reduction of the intensity. Also for triple axis spectrometers this is a very unfavourable energy range, as the reflectivities of the crystal monochromators are low and narrow beam collimation is necessary in order to obtain the necessary resolution in energy.



It might be worthwhile to study other moderator concepts which improve the performance of TOF spectrometers in this energy range. A possible solution might be a moderator of liquid methane with a rethermalization layer on one of the surfaces. TOF instruments will be placed at beam tubes looking on the methane moderator, whereas stationary experiments extract their beams from the rethermalization layer.

In this report only those experiments were studied, which use neutron beams from the fast moderator. Due to the long lifetime of neutrons in the D<sub>2</sub> cold source TOF spectroscopy is unfavourable for studies of collective excitations in single crystals.

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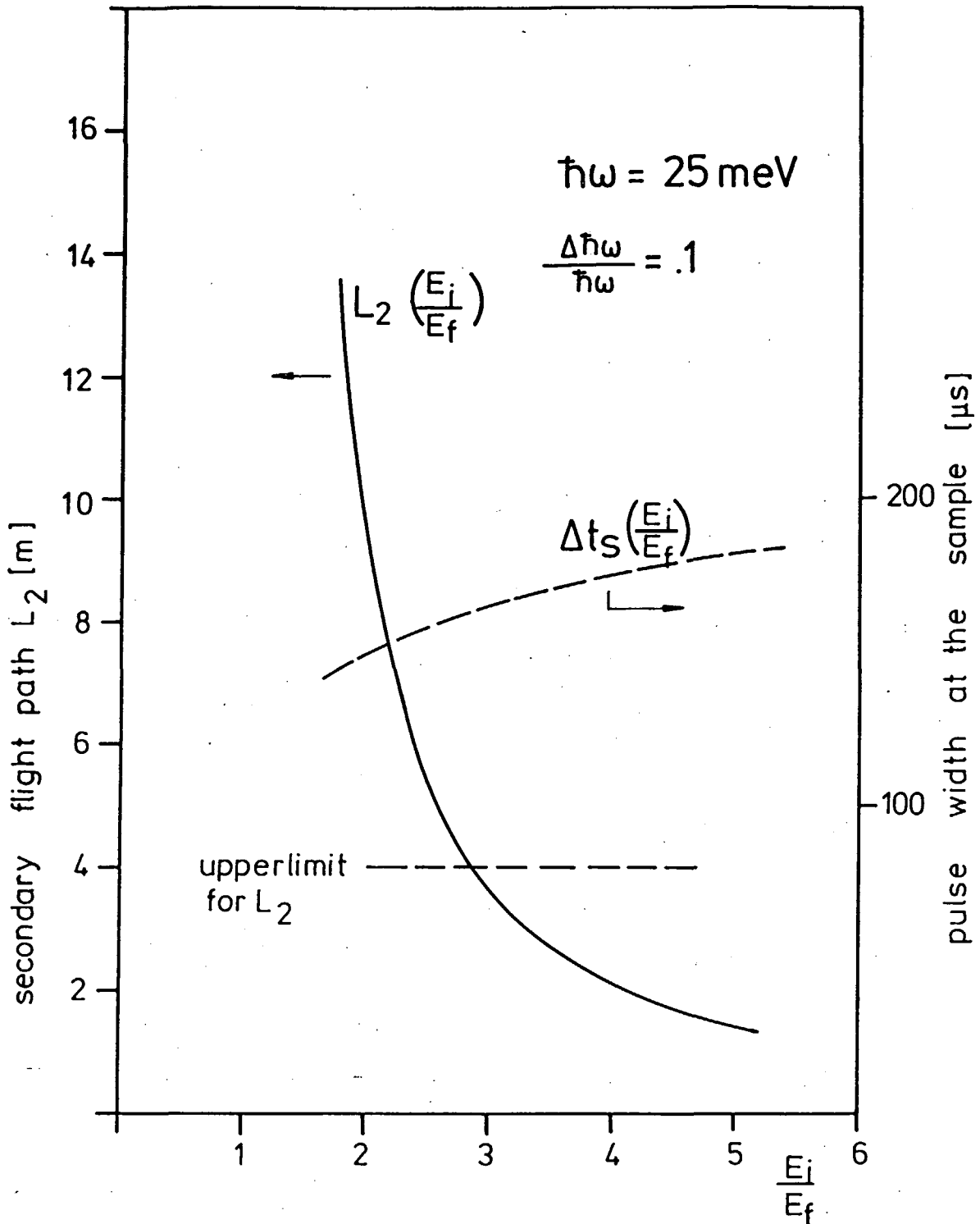


Fig.1: Length of the secondary flight path as a function of  $E_i/E_f$  for a TOF spectrometer of type X-TOF at the thermal moderator of the SNQ if  $\hbar\omega = 25 \text{ meV}$  and  $\frac{\Delta\hbar\omega}{\hbar\omega} = 10\%$   
 ----- pulse width at the sample

# Multi-arm Crystal Analyzer TOF Spectrometer (TOF-XXX)

H.Rauh and R.Geick

Physikalisches Institut der Universität, Röntgenring 8, 8700 Würzburg

## 1. The concept

A neutron spectrometer of inverted geometry type for the measurement of collective excitations in single crystals with excitation energy up to about 300 meV is described. Its principle, shown in Fig. 1, is similar to that of the multi-analyzer crystal spectrometer MAX /1/ at the KENS pulsed spallation neutron source: A white beam of incoming neutrons is collimated onto the sample. Scattered neutrons of fixed energy  $E_f$  are selected by analyzer crystals and registered as a function of their time-of-flight (TOF). The scattering diagram (of a single detector arm ) is shown in Fig. 2. During a scan,  $k_f$  ( and therefore  $Q_{\perp}$  , the component of the momentum transfer  $Q$  perpendicular to  $k_i$  ) is kept constant, while  $k_i$  varies with

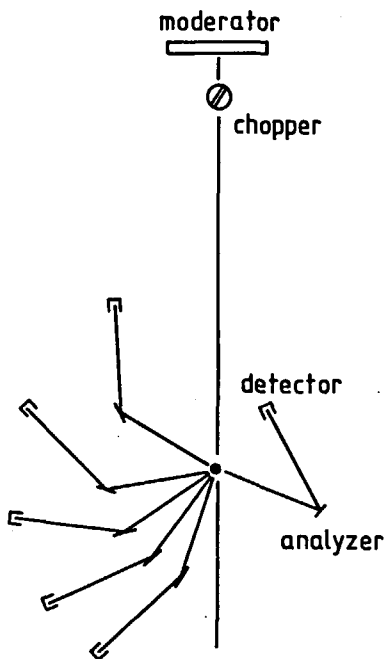


Fig.1: Principle layout of the TOF-XXX spectrometer

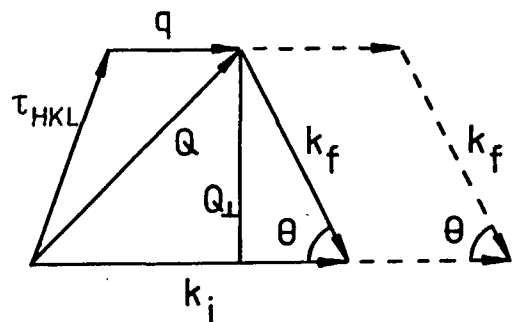


Fig.2: Scattering diagramm (at two different TOF)

TOF. Let us write  $Q$  as sum of a reciprocal lattice point  $\tau_{\text{HKL}}$  of the sample, and  $q$  ( $q$  is not necessarily inside the Brillouin zone around  $\tau_{\text{HKL}}$ ). If  $Q_{\perp} = (\tau_{\text{HKL}})_{\perp}$ ,  $q$  is parallel to  $k_i$  at all TOF. Therefore every single detector arm performs a TOF scan along a constant direction  $q$ . Note that  $q$  can be aligned parallel to any desired direction in the first Brillouin zone of the sample simply by rotating the sample ( but this does not imply that any desired direction of  $q$  starting from any  $\tau_{\text{HKL}}$  can be aligned, see below ). The path of a detector arm in the  $(\hbar\omega - Q)$  plane is given by

$$\hbar\omega = \hbar^2/2m [(Q_{\parallel} + k_f \cos \theta)^2 - k_f^2] \quad (1)$$

$$Q_{\perp} = -k_f \sin \theta$$

where  $\perp$  and  $\parallel$  refer to the direction of the incoming neutron beam.

The multi-arm equipment may be used for survey measurements. If all analyzer crystals are aligned at the same value of  $Q_{\perp}$ , the various spectrometer paths cut neighbouring Brillouin zones of the sample at different value of the energy transfer  $\hbar\omega$ , cf. Fig. 3(a). Thus the complete excitation spectrum can be measured simultaneously. As an example, Fig. 4 shows the result of a survey measurement of phonons in  $\text{TiO}_2$  ( which was optimized for high frequency branches ) with the spectrometer MAX /2/ ( solid lines: spectrometer paths; broken lines: phonon dispersion ( selected branches ) in (010) direction and Brillouin zones ). 18 phonons with energy above about 40 meV ( circles in Fig. 4 ) have been observed in a measuring time

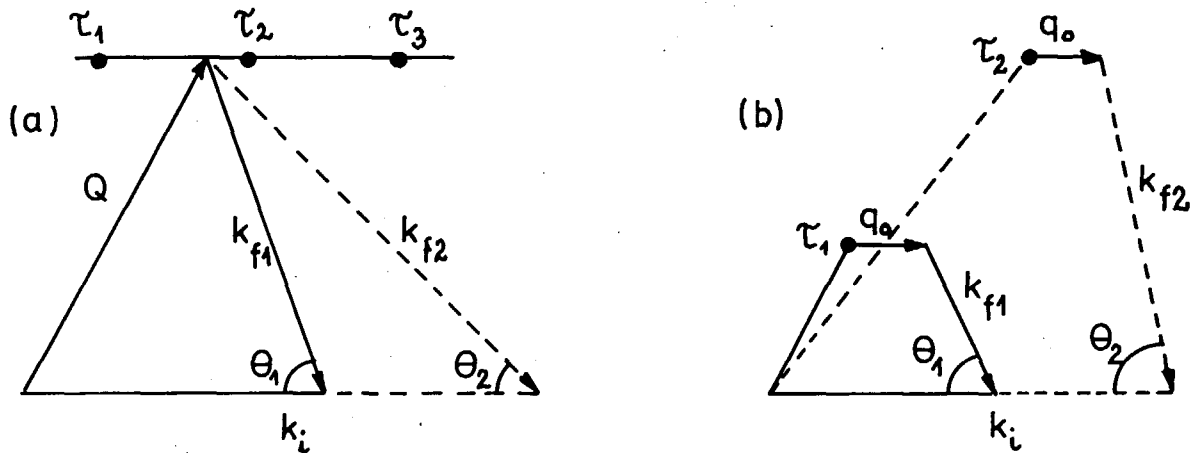


Fig.3a: Survey measurement of the complete excitation spectrum (a) or of a single excitation at different  $\tau_{\text{HKL}}$  (b).

of about 2 days, which corresponds to a few minutes at the SNQ (scaled by the peak flux ratio). For survey measurements one would like to lay a net of spectrometer paths as dense as possible through the  $(\hbar\omega - Q_{||})$  plane. This has the consequence that the analyzer crystals and detectors practically cannot be moved, and that an adjustment of the instrumental resolution /3/ to the experiment is nearly impossible. For many experiments it

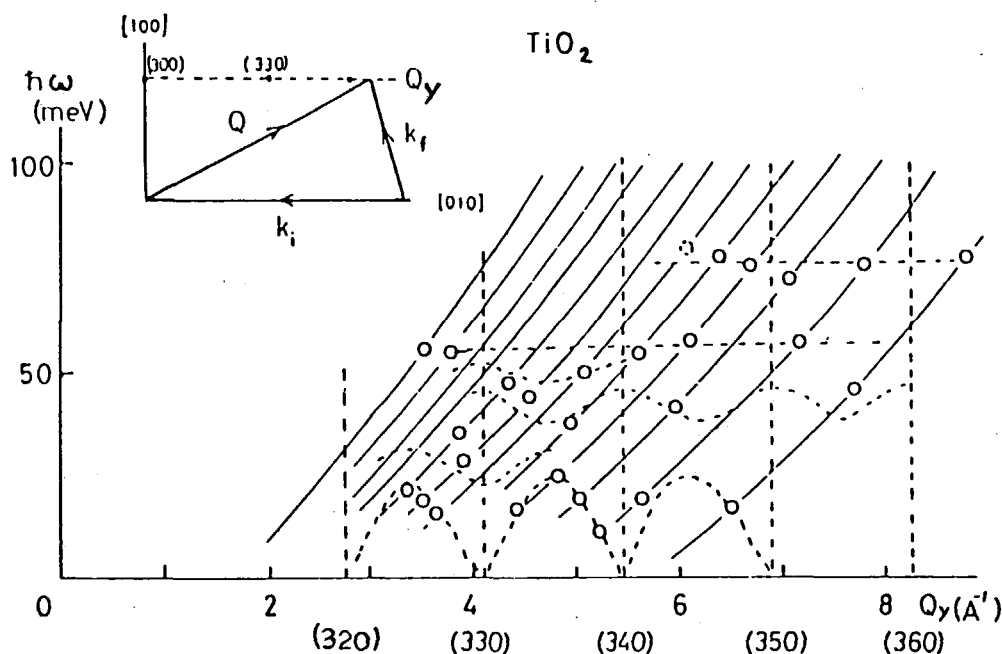


Fig. 4: Survey measurement of phonons in  $\text{TiO}_2$  with MAX at KENS /2/.

may be important to keep the freedom of adjusting the resolution by using only a few detector arms, say up to 10, and increase the density of spectrometer paths in the  $(\hbar\omega - Q_{||})$  plane consecutive in time (i.e. by a number of independent measurements at different scattering angles) instead of simultaneously. Some flexibility in the adjustment of the instrumental resolution can be gained by "vertical scattering" at the analyzer crystal as indicated in Fig. 5. For intensity reasons one would like to avoid the use of vertical collimators. In the case of very small samples, an image of the sample can be focussed monochromatically on the detector by using curved analyzers and placing the detector at  $L_D = L_A$ . However, shielding the secondary neutron flight path seems to be very complicated in the case of vertical scattering at the analyzer.

The various detector arms may also be used to measure a certain excitation in different Brillouin zones (which differ from each other in  $Q_{\perp}$ )

simultaneously, cf. Fig. 3(b).

Beside survey measurements this instrument should be useful for the detailed study of a certain excitation, where the adjustment of the instrumental resolution is mandatory. In order to allow even for high-resolution experiments, quasi-back scattering from the analyzer crystal should be possible. Therefore one side of the scattering plane should be reserved for a single detector-arm version of the instrument as indicated in Fig. 1.

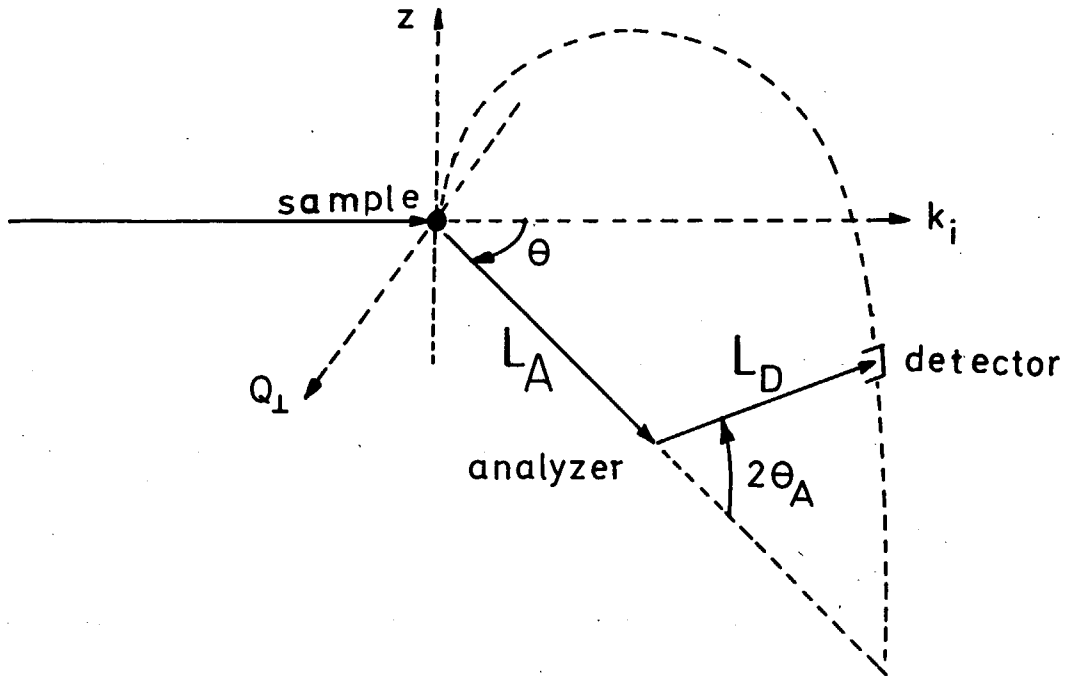


Fig. 5: "vertical scattering" at the analyzer crystal.

#### Scan polarization

The measurement of purely transverse ( $q \perp \tau_{\text{HKL}}$ ) (where  $\tau_{\text{HKL}}$  is a reciprocal lattice point) or longitudinal scans ( $q \parallel \tau_{\text{HKL}}$ ) is only possible consecutive in time, but not simultaneously. A special situation occurs at those time-channels where  $Q_{\parallel} = 0$ . There, the energy transfer  $\hbar\omega =: -\hbar\omega_0 = -\hbar^2/2m Q_{\perp}^2$  is independent of the scattering angle  $\theta$ . If  $n$  detector arms are aligned at the same value of  $Q_{\perp}$ , the fixpoint  $(-\hbar\omega_0, 0)$  is simultaneously  $n$ -fold illuminated by. Provided the resolution of the different detector arms at  $(\hbar\omega_0, 0)$  is approximately the same, a longitudinal scan can be constructed from a series of fixpoints. Such a scan may be useful to study excitations in very small samples.

Scan modus "Const.-E" or "Const.-Q"

Such scans may be constructed by re-arrangement of the data from a survey measurement without any additional measuring time. Furthermore, the re-arrangement can be done in such a way that the resulting scan cuts the dispersion curve perpendicularly. It is also possible to perform (quasi) const.-Q scans directly by using vibrating analyzer crystals and collimators /4/ or samples /5/.

### 3. An example: Measurement of high-energy magnetic excitations

In those experiments the momentum transfer is restricted to "small values", say  $Q \leq Q_{\max} = 6 \text{ \AA}^{-1}$ . Consequently the scattering angle  $\theta$  is "small" and the final neutron energy  $E_f$  "large". Fig. 6 shows some detector paths at various  $\theta$  and  $Q_{\perp}$ . The vertical lines represent the limitation in  $Q$

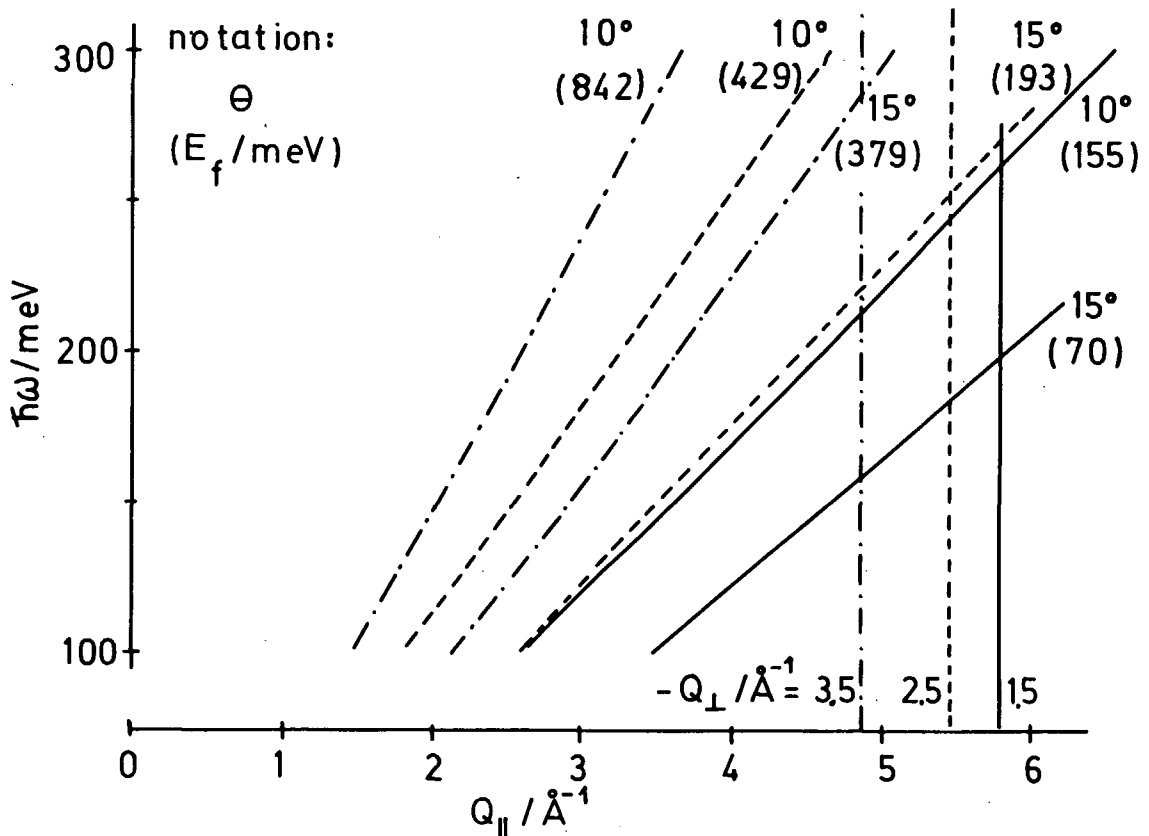


Fig. 6: Detector paths at for the measurement of high energy excitations at low momentum transfer.

( $= Q_{\max}$ ). At fixed  $Q_{\perp}$ ,  $E_f$  has a minimum for  $\theta = 90^\circ$  and increases rapidly with decreasing  $\theta$ , cf. equ.(1); note that  $E_f$  does not depend upon the energy transfer). At fixed  $\theta$ ,  $E_f$  decreases with decreasing  $Q_{\perp}$ . However, a decreasing value of  $Q_{\perp}$  (which at fixed  $\theta$  improves the energy resolution) decreases the accessible range of the energy transfer. As a consequence, analyzer crystals with small d-spacing and high reflectivity are absolutely necessary for the study of high-energy magnetic excitations. As an example, let us measure a magnon at  $Q_{\perp} = -2.5 \text{ \AA}^{-1}$ ,  $Q_{\parallel} = 5.45 \text{ \AA}^{-1}$  ( $Q = Q_{\max}$ ) and  $\hbar\omega = 300 \text{ meV}$ . The energy resolution should be 10%, and the collimation  $0.5^\circ$ . With equ.(1) one finds  $\theta = 12.7^\circ$ ,  $E_f = 270 \text{ meV}$  and consequently  $E_i = 570 \text{ meV}$ . Assuming that the energy resolution is dominated by collimators ( $\theta_A = 0.5^\circ$ ) and the Bragg angle  $\theta_A$  of the analyzer crystal, we find  $\theta_A = 9.9^\circ$  and  $d_A = 1.6 \text{ \AA}$  ( $d_A(\text{PG004}) = 1.67 \text{ \AA}$ ). This example shows that the measurement of magnetic excitations with energy up to about 300 meV is indeed possible with a TOF-XXX instrument.

### 3. Comparison with competing concepts

#### a) Triple-Axis Spectrometer (TAS)

The performance of a TAS at the SNQ in comparison to a X-TOF instrument has been discussed in detail in separate papers [6,7]. Similar conclusions hold also for the TOF-XXX spectrometer: With a TAS, any scan is performed in a step-by-step procedure, i.e. consecutive in time, and not simultaneously like with a TOF-XXX. On the other hand, the instrumental resolution of a TAS can be adjusted continuously within rather wide limits, but only step-wise (by changing the reflection plane or even the analyzer crystal) in the case of TOF-XXX. Therefore we conclude that a TOF-XXX is superior compared to a TAS for survey measurements, but not for detailed studies. An exception are high energy excitations ( $\hbar\omega > 100 \text{ meV}$ ), especially magnons, where the small reflectivity of the TAS monochromator crystal at high incoming neutron energies may lead to unacceptably long measuring times. Finally we note that the TOF-XXX concept is mechanically more simple than the TAS concept as there are no moving parts during a scan.



## b) Constant-Q Spectrometer (CQS)

The concept of a CQS is specialized for the performance of a special scan, the "constant-Q" scan. It is therefore much more restricted in its application than the TOF-XXX. Furthermore, scattering angles around  $90^\circ$  (important for phonon measurements) are not accessible with a CQS. The large analyzer crystal in a CQS guarantees a continuous variation of the scattering angle, but prohibits the use of collimators in the scattered beam. This fact, in addition to the coupling between analyzer Bragg angle and scattering angle, leads to an inflexible instrumental resolution of the CQS. Noting that const.-Q scans can be performed with a TOF-XXX by data rearrangement, we conclude that TOF-XXX is the superior concept.

4. Neutron pulse and  $E_i$ -resolution

While the  $E_f$ -resolution is mainly determined by the analyzer crystals, the  $E_i$ -resolution  $\Delta E_i$  is governed by the finite width  $\Gamma_p$  and TOF of the incoming neutron pulse:

$$\Delta E_i/E_i = 2 \Gamma_p / \text{TOF} \quad (2)$$

In principle is possible to improve the  $E_i$  resolution by making the incoming neutron flight path longer, but intensity considerations prevent us from going that way. Therefore the  $E_i$  resolution should be controlled through  $\Gamma_p$ . The time-spread of the proton bunch at the spallation target,  $\gamma_p$ , is a lower limiting value for  $\Gamma_p$ . The additional broadening is caused by the moderation time  $\Gamma_p^0$  (which is determined by the properties of the moderator like material, geometry, temperature, poisoning, etc.).  $\Gamma_p$  may be controlled by pulse-forming devices like choppers. Note that both moderator poisoning and the use of a chopper must be paid by neutron beam intensity. In order to give some numerical examples for minimum pulse-widths, let us assume  $l_i = 8$  m and  $\Delta E_i/E_i = 3$  %. Then  $\Gamma_p(50 \text{ meV}) = 38.8 \text{ } \mu\text{s}$ ,  $\Gamma_p(300 \text{ meV}) = 15.8 \text{ } \mu\text{s}$  and  $\Gamma_p(1000 \text{ meV}) = 8.7 \text{ } \mu\text{s}$ . We compare these values to the SNQ design goals. Without compressor ring ( $\Gamma_p \geq \gamma_p = 250 \text{ } \mu\text{s}$ ), the SNQ pulse-width is much larger than these values, and pulse-forming choppers are absolutely necessary for a TOF-XXX spectro-

meter. The compressor ring reduces the proton pulse-width to about 0.6  $\mu$ s. Then the contribution of the neutron pulse-width to the  $E_i$  resolution at neutron energies in the slowing-down region of the spectrum is only 0.7 %, but this value increases strongly in the Maxwellian region. (Set  $E_i = 25$  meV and  $\Gamma_p = 120$   $\mu$ s, then  $\Delta E_i/E_i = 6.6$  %.) Pulse-forming choppers for thermal neutrons are of standard technology and allow for an adjustment of the  $E_i$ -resolution in rather wide limits. However, they must be installed as close as possible to the moderator and are therefore not easy to handle in the case of malfunction. Frame-overlap considerations specify the repetition rate  $r$  of the optimum source. Assuming  $l_i + l_f = 10$  m and  $E_i = E_f \geq 10$  meV, we find  $r = 138$  Hz as optimum value for such an instrument.

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TOF-TOF Spectrometers atPulsed Neutron Sources and at Steady-State Reactors

R. E. Lechner

Hahn-Meitner-Institut für Kernforschung  
Bereich Kernchemie und Reaktor  
Glienicke Straße 100, D-1000 Berlin 39

General formulae for the determination of intensity and energy resolution of a TOF-TOF spectrometer employing disk choppers are derived in terms of the neutron wavelength, the chopper-chopper and sample-detector distances and the chopper burst times. Aspects of spectrometer optimization on the basis of these formulae are considered, including a discussion of the restrictions encountered at a pulsed source due to fixed pulse width and pulse rate of the source. It is found that a TOF-TOF spectrometer can be optimized for a given incident neutron wavelength, so that the peak flux of the pulsed source is fully used, and the intensity ratio pulsed-source/steady-state-reactor for the same spectrometer is directly given by the pulsed-source flux averaged over the burst time  $\tau_1$  of the first chopper, divided by the flux of the steady-state reactor. Numerical results from the evaluation of the intensity and resolution formulae are presented for a number of different spectrometer configurations. We show graphically for a few typical examples, in which way intensity and resolution are influenced by the four essential spectrometer variables.

## 1. Introduction

The TOF-TOF method employing time-of-flight techniques for neutron monochromatisation as well as for energy analysis (as opposed for instance to instruments where partially or exclusively crystal reflection techniques are used) is one of the methods that are widely applied in inelastic neutron scattering experiments. It is particularly appropriate for quasielastic scattering studies, where great flexibility in the choice of the energy resolution  $\Delta E$  is required. Since the latter is closely related to the intensity of the experiment, both quantities must be considered when the spectrometer configuration is optimized.

In the present paper intensity and energy resolution of a TOF-TOF spectrometer are discussed as a function of the essential instrument variables defining the spectrometer configuration. Because of its conceptual simplicity we have chosen to treat a double-chopper spectrometer of the IN5-type [1], where the neutron beam is chopped with disk choppers. In such a system the definition of the incident neutron wavelength  $\lambda_0$  and of the neutron burst times are uncoupled leading to the remarkable flexibility of the instrument with respect to resolution and range of wave vector transfer  $\vec{Q}$ . Since in the present context we are only interested in intensity and resolution the discussion will be limited to the two choppers needed for the creation of monochromatic neutron pulses. In practice a TOF-TOF spectrometer contains additional choppers for the purpose of i) avoiding frame overlap of incident neutrons and ii) reducing the frame overlap of scattered neutrons. In case i) there is no influence on intensity and resolution; in case ii) the resolution is not affected, but the intensity is, because such anti-frame-overlap choppers are used to define the period  $P$  of the spectrometer. This effect will be taken into account by introducing the period into the intensity formula.

In the following we will derive theoretical expressions for intensity and resolution of the instrument. These formulae will then be used for a numerical evaluation of the spectrometer performance as a function of the instrument parameters. Special attention will be paid to the use of TOF-TOF spectrometers at pulsed neutron sources.

## 2. The Intensity of the Spectrometer

The principle of a double-chopper spectrometer is shown in Fig.1 with the aid of a flight-path/time-of-flight diagram. Two choppers connected by a neutron guide are set up at a distance  $L_{12}$  from each other, the sample is at a distance  $L_{2S}$  from chopper 2, and the detectors are mounted at a distance  $L_{SD}$  away from the sample. The "white" continuous neutron beam incident on chopper 1 is chopped into pulses with widths  $\tau_1$  in time;  $P$  is the period,  $P^{-1}$  the pulse rate. Chopper 2 has an opening time  $\tau_2$ , during which it selects a narrow wavelength band from the incident neutron pulses. Only the paths of those neutrons which actually can reach the sample are shown in the Figure.

The intensity of an experiment is determined by the number of neutrons arriving at the sample per unit of time. This is proportional i) to the "duty cycle"  $\tau_1/P$ , i.e. to the fraction of the time during which chopper 1 is open and ii) to the incident neutron flux per unit of energy,  $d\phi/dE_0$ , multiplied by the energy band width  $\Delta E_0$  selected during the chopper burst time  $\tau_2$ :

$$I \propto (\tau_1/P) \cdot (d\phi/dE_0) \cdot \Delta E_0 \quad (1)$$

In an actual experiment the period is equal to the maximum neutron time-of-flight (TOF) to be measured. Usually this must be large enough to enable neutrons of wavelengths somewhat larger than  $\lambda_0$  to be measured. As a rule of thumb we may set

$$\text{TOF}_{\text{max}} = P[\mu\text{s}] = 1.5 \cdot \lambda_0[\text{\AA}] \cdot L_{SD}[\text{m}] \cdot 252.77 \quad (2)$$

The energy width  $\Delta E_0$  is given by

$$\Delta E_0 = \text{const.} \cdot \tau_2 / (L_{12} \lambda_0^3) \quad (3)$$

From eqs. (1) to (3) we obtain

$$I \propto (d\phi/dE_0) \cdot \tau_1 \tau_2 / (\lambda_0^4 \cdot L_{12} \cdot L_{SD}) \quad (4a)$$

or

$$I \propto (d\phi/d\lambda_0) \tau_1 \tau_2 / (\lambda_0 \cdot L_{12} \cdot L_{SD}) \quad (4b)$$

It is seen that for a given incident wavelength  $\lambda_0$  the experimental intensity is governed by the simple factor

$$F = \tau_1 \tau_2 / (L_{12} L_{SD}) \quad (5)$$

Obviously the spectrometer intensity is also influenced by attenuation factors due to losses in the neutron guide, as well as neutron absorption and scattering by material encountered along the flight path, such as gas, windows, sample holder and detector walls. Such factors will however not be discussed in detail in this paper.

### 3. The Resolution of the Spectrometer

In this section we derive the apparent energy resolution  $\Delta h\nu$  of the spectrometer. This is the full-width-at-half-maximum (FWHM) in energy of the spectrometer's response to a  $\delta$ -function peak in the double-differential neutron scattering cross-section. We proceed by first calculating the time spread  $\Delta t_s$  of a neutron pulse arriving at the sample. By inspection of Fig.1 it is seen that this can be obtained as the width of the convolution of two distributions in time at the sample position. One is the distribution of all neutrons having passed chopper 2 during the time interval  $\tau_2$ , after having passed chopper 1 just in the middle of its burst (of duration  $\tau_1$ ). The other is the distribution of all neutrons which have passed chopper 2 at a given instant within the time interval  $\tau_2$  after having

passed chopper 1 at any instant during the time interval  $\tau_1$ . For simplicity we assume that these distributions are Gaussians, so that we can write (see Fig.1):

$$\Delta t_s = (\tau_{1s}^2 + \tau_{2s}^2)^{1/2} \quad (6)$$

where

$$\tau_{1s} = \tau_2 \cdot (L_{12} + L_{2s})/L_{12} \quad (7a)$$

and

$$\tau_{2s} = \tau_1 \cdot L_{2s}/L_{12} \quad (7b)$$

Similarly we obtain for the time spread at the detector:

$$\Delta t_d = (\tau_{1d}^2 + \tau_{2d}^2)^{1/2} \quad (8)$$

For elastic scattering one simply finds

$$\tau_{1d} = \tau_2 (L_{12} + L_{2s} + L_{sd})/L_{12} \quad (9a)$$

$$\tau_{2d} = \tau_1 (L_{2s} + L_{sd})/L_{12} \quad (9b)$$

In the case of inelastic scattering  $L_{sd}$  must be replaced by  $L_{sd} \cdot \lambda^3/\lambda_0^3$  in eqs. (9a) and (9b).

A further contribution to the time spread at the detector is due to the uncertainty  $\delta L$  in the length of the flight path, caused by beam divergence, finite sample size and detector thickness. We therefore write instead of (8):

$$\Delta t_d = (\tau_{1d}^2 + \tau_{2d}^2 + \tau_{3d}^2)^{1/2} \quad (10)$$

where

$$\tau_{3D}[\mu s] = \oint L[m] \cdot \lambda[\text{\AA}] \cdot 252.77 \quad (11)$$

Thus we obtain

$$\begin{aligned} \Delta t_d = [ & (L_{12} \cdot \oint L \cdot \lambda \cdot 252.77)^2 + \tau_1^2 (L_{2S} + L_{SD} \cdot \lambda^3 / \lambda_o^3)^2 \\ & + \tau_2^2 (L_{12} + L_{2S} + L_{SD} \cdot \lambda^3 / \lambda_o^3)^2 ]^{1/2} / L_{12} \end{aligned} \quad (12)$$

This may be converted into an effective energy spread using the relations

$$\Delta E/E = 2 \Delta \lambda / \lambda \quad (13)$$

and

$$\Delta \lambda_d = \Delta t_d / (L_{SD} \cdot 252.77) \quad (14)$$

where  $\Delta \lambda_d$  is the apparent wavelength spread at the detector. Setting  $\Delta E = \Delta(\hbar\omega)$  we finally obtain for the effective energy resolution:

$$\Delta(\hbar\omega) [\text{meV}] = 0.6472 \cdot [A^2 + B^2 + C^2]^{1/2} / (\lambda^3 L_{12} L_{SD}) \quad (15)$$

with

$$\begin{aligned} A &= L_{12} \cdot \oint L \cdot \lambda \cdot 252.77 \\ B &= \tau_1 (L_{2S} + L_{SD} \cdot \lambda^3 / \lambda_o^3) \\ C &= \tau_2 (L_{12} + L_{2S} + L_{SD} \cdot \lambda^3 / \lambda_o^3) \end{aligned} \quad (16)$$

where  $L_{12}$ ,  $L_{2S}$ ,  $L_{SD}$ ,  $\oint L$  are given in [m],  $\lambda$  in [ $\text{\AA}$ ] and  $\tau_1$ ,  $\tau_2$  in [ $\mu s$ ].



#### 4. Aspects of Spectrometer Optimisation at a Pulsed Source

Formulae (1) for the intensity and (15) for the energy resolution are quite general, if we disregard some minor factors which are of little interest in our context. They are equally valid for TOF-TOF spectrometers at pulsed sources and at steady-state reactors. We note however that a pulsed source implies two restrictions:

- i) An upper limit for  $\tau_1$  is given by the burst time  $\tau_s$  of the source itself, i.e.  $\tau_1 \leq \tau_s$ . It is of the order of 100 to 500  $\mu\text{s}$  for cold neutrons at the British (SNS) and German (SNQ) spallation source projects, respectively. Obviously this will have some influence on instrument design and performance, but it should not cause any serious limitation concerning intensity and resolution of a TOF-TOF spectrometer.
- ii) A lower limit is imposed on the spectrometer period  $P$  by the period  $P_s$  of the pulsed source, i.e.  $P \geq P_s$ , where  $P_s = 10000 \mu\text{s}$  for the SNQ and  $20000 \mu\text{s}$  for the SNS. According to eq. (2) a period  $P_s$  is well adapted if the desired incident neutron wavelength  $\lambda_0$  is equal to an optimum value

$$\lambda_{\text{opt}} [\text{\AA}] = P_s [\mu\text{s}] / (1.5 L_{\text{SD}} [\text{m}] 252.77) \quad (17)$$

If  $\lambda_0 < \lambda_{\text{opt}}$  or if  $\lambda_0 > \lambda_{\text{opt}}$  the source period  $P_s$  is not well adapted to the incident wavelength and this causes an intensity loss as compared to the adapted situation. Obviously one might vary  $L_{\text{SD}}$ ; but this seems feasible only, when the angular range where neutrons are detected is very limited (as for instance in small-angle scattering), and it would in general require the simultaneous variation of other spectrometer parameters according to eq. (15), in order to achieve the required energy resolution. In Table 1 we list the  $\lambda_{\text{opt}}$ -values corresponding to various sample-detector distances  $L_{\text{SD}}$  for the SNQ and for the SNS, respectively.

Let us illustrate this problem by an example. Suppose we wish to use an incident neutron wavelength of  $\lambda_0 = 8.8 \text{\AA}$  on a

spectrometer with  $L_{SD} = 1.5$  m at SNQ. For this case  $P_S$  is twice the value needed, which corresponds to an intensity loss factor

Table 1 Optimum Wavelength Values

| $L_{SD}$ [m] | $\lambda_{opt}$ [Å]<br>for SNQ<br>$P_S = 10000 \mu s$ | $\lambda_{opt}$ [Å]<br>for SNS<br>$P_S = 20000 \mu s$ |
|--------------|-------------------------------------------------------|-------------------------------------------------------|
| 1.5          | 17.6                                                  | 35.2                                                  |
| 3            | 8.8                                                   | 17.6                                                  |
| 6            | 4.4                                                   | 8.8                                                   |
| 12           | 2.2                                                   | 4.4                                                   |
| 24           | 1.1                                                   | 2.2                                                   |

of 2, as compared to a source with half the value of  $P_S$  at the same peak flux. If  $L_{SD} = 6$  m the source period is too short by a factor 2 leading again to the same loss factor. If however  $L_{SD} = 3$  m the desired incident wavelength is well adapted to  $P_S$  and consequently there is no loss factor. In this ideal case the peak flux of the spallation source is fully used and the intensity ratio pulsed-source/steady-state-reactor for the same spectrometer is directly given by the pulsed-source flux averaged over the chopper burst time  $\tau_1$ , divided by the flux of the steady-state reactor.

## 5. Qualitative Discussion and Numerical Results

Apart from the restrictions just discussed the design optimisation of a TOF-TOF spectrometer on the basis of eqs. (1) (or (4a), (4b)) and (15) represents exactly the same problem for a pulsed source as for a steady-state reactor. In this paper we will not treat this question in much detail, but merely derive a few qualitative conclusions from the formulae. Further information on the general subject can be found in previous

publications about various TOF-spectrometers [2-5]. It is obvious that in many cases the energy resolution can be improved by increasing distances  $L_{12}$  and/or  $L_{SD}$  and by decreasing the chopper burst times  $\tau_1$  and/or  $\tau_2$ . Simultaneously the intensity then usually diminishes. In order to achieve high flexibility, i.e. large ranges of variation for resolution and intensity, it is advantageous

- i) to minimize term A in eq.(15), so that it is kept small as compared to the terms B and C:  $A \ll B, C$ .
- ii) to vary the spectrometer parameters in such a way that the terms B and C remain of the same order of magnitude:  $B \approx C$ .

The distance  $L_{2S}$  between chopper 2 and the sample must be minimized, because - just as the term B - it worsens the resolution without providing any gain in intensity.

The sample-detector distance  $L_{SD}$  will generally be limited to a few meters because of the high cost of neutron detectors in the case of spectrometers with simultaneous data recording over large angular ranges.

In the following we report some results from a numerical evaluation of the above formulae for various spectrometer configurations. These results are shown in Figs. 2 to 7. Each figure presents a "quasi-threedimensional" plot of the energy resolution  $\Delta(\hbar\omega)$  [meV] as a function of  $\lambda_0$  [Å] and  $\lambda$  [Å] (see eq.(15)). The lower left corners of the insets correspond to  $\Delta(\hbar\omega) = 0$  and  $\lambda_0 = 3$  Å for Fig.2,  $\lambda_0 = 6$  Å for Figs. 3 to 7. The number at the bottom of each plot indicates the pertinent spectrometer configuration, described in Table 2. The last column of this Table lists the corresponding intensity factor F defined by eq.(5). In all calculations the same values were used for the parameters  $L_{2S}$  (= 1m) and  $\phi_L$  (= 0.02 m).

Table 2: Spectrometer Configurations

| Nr. | $L_{12}$<br>[m] | $L_{2S}$<br>[m] | $L_{SD}$<br>[m] | $\delta L$<br>[m] | $\tau_1$<br>[ $\mu s$ ] | $\tau_2$<br>[ $\mu s$ ] | F<br>[ $\mu s^2 m^{-2}$ ] |
|-----|-----------------|-----------------|-----------------|-------------------|-------------------------|-------------------------|---------------------------|
| 5   | 48              | 1               | 1.5             | 0.02              | 400                     | 12.5                    | 69.44                     |
| 6   | 24              | 1               | 3               | 0.02              | 200                     | 25                      | 69.44                     |
| 7   | 12              | 1               | 6               | 0.02              | 100                     | 50                      | 69.44                     |
| 8   | 6               | 1               | 12              | 0.02              | 50                      | 100                     | 69.44                     |
| 9   | 3               | 1               | 24              | 0.02              | 25                      | 200                     | 69.44                     |
| 10  | 1.5             | 1               | 48              | 0.02              | 12.5                    | 400                     | 69.44                     |
| 11  | 1.5             | 1               | 1.5             | 0.02              | 12.5                    | 12.5                    | 69.44                     |
| 12  | 3               | 1               | 3               | 0.02              | 25                      | 25                      | 69.44                     |
| 13  | 6               | 1               | 6               | 0.02              | 50                      | 50                      | 69.44                     |
| 14  | 12              | 1               | 12              | 0.02              | 100                     | 100                     | 69.44                     |
| 15  | 24              | 1               | 24              | 0.02              | 200                     | 200                     | 69.44                     |
| 16  | 48              | 1               | 48              | 0.02              | 400                     | 400                     | 69.44                     |
| 18  | 3               | 1               | 1.5             | 0.02              | 25                      | 12.5                    | 69.44                     |
| 19  | 6               | 1               | 1.5             | 0.02              | 50                      | 12.5                    | 69.44                     |
| 20  | 12              | 1               | 1.5             | 0.02              | 100                     | 12.5                    | 69.44                     |
| 21  | 24              | 1               | 1.5             | 0.02              | 200                     | 12.5                    | 69.44                     |
| 22  | 48              | 1               | 1.5             | 0.02              | 400                     | 12.5                    | 69.44                     |
| 23  | 48              | 1               | 3               | 0.02              | 400                     | 25                      | 69.44                     |
| 24  | 48              | 1               | 6               | 0.02              | 400                     | 50                      | 69.44                     |
| 25  | 48              | 1               | 12              | 0.02              | 400                     | 100                     | 69.44                     |
| 26  | 48              | 1               | 24              | 0.02              | 400                     | 200                     | 69.44                     |
| 27  | 48              | 1               | 48              | 0.02              | 400                     | 200                     | 34.72                     |
| 28  | 48              | 1               | 48              | 0.02              | 400                     | 100                     | 17.36                     |
| 29  | 48              | 1               | 48              | 0.02              | 400                     | 50                      | 8.68                      |
| 30  | 48              | 1               | 48              | 0.02              | 400                     | 25                      | 4.34                      |
| 31  | 48              | 1               | 1.5             | 0.02              | 400                     | 12.5                    | 69.44                     |
| 32  | 48              | 1               | 1.5             | 0.02              | 400                     | 25                      | 138.9                     |
| 33  | 48              | 1               | 1.5             | 0.02              | 400                     | 50                      | 277.8                     |
| 34  | 48              | 1               | 1.5             | 0.02              | 400                     | 100                     | 555.6                     |
| 35  | 48              | 1               | 1.5             | 0.02              | 400                     | 200                     | 1111                      |
| 36  | 48              | 1               | 1.5             | 0.02              | 400                     | 400                     | 2222                      |

| Nr. | $L_{12}$<br>[m] | $L_{2S}$<br>[m] | $L_{SD}$<br>[m] | $\mathcal{S}L$<br>[m] | $\tau_1$<br>[μs] | $\tau_2$<br>[μs] | F<br>[μs <sup>2</sup> m <sup>-2</sup> ] |
|-----|-----------------|-----------------|-----------------|-----------------------|------------------|------------------|-----------------------------------------|
| 57  | 6               | 1               | 1.5             | 0.02                  | 25               | 25               | 69.44                                   |
| 58  | 6               | 1               | 1.5             | 0.02                  | 50               | 25               | 138.9                                   |
| 59  | 6               | 1               | 1.5             | 0.02                  | 100              | 25               | 277.8                                   |
| 60  | 6               | 1               | 1.5             | 0.02                  | 200              | 25               | 555.6                                   |
| 61  | 6               | 1               | 1.5             | 0.02                  | 400              | 25               | 1111                                    |

Figs. 2 to 5 show the resolution for a series of spectrometer configurations, where the intensity factor F has been kept constant. In Fig.2 the distances and burst times have been varied such that  $L_{12} \cdot L_{SD} = \text{const.}$ ,  $\tau_1 \cdot \tau_2 = \text{const.}$  and  $L_{12} / L_{SD} = \tau_1 / \tau_2$ . Best resolution is obtained in example Nr.7. The resolution is worst in configuration Nr.10 (see Table 2). This is a demonstration of the fact that poor resolution does not necessarily warrant high intensity.

Fig.3 corresponds to a number of configurations in which the four parameters  $L_{12}$ ,  $L_{SD}$ ,  $\tau_1$ ,  $\tau_2$  have been scaled simultaneously, whereby within the present approximation the intensity remains constant. This indicates that increasing the spectrometer "size" (at constant values of  $L_{2S}$  and  $\mathcal{S}L$ ) generally improves the energy resolution. Fig.4 shows that the same effect is obtained to a lesser extent, if only the dimensions of the primary spectrometer are scaled.

In Fig.5 only  $L_{SD}$  and  $\tau_2$  are varied, with the condition  $\tau_2 / L_{SD} = \text{const.}$ , i.e. the secondary spectrometer is scaled while the primary spectrometer is kept constant. In this case the largest variation of the energy resolution is observed for elastic scattering and for energy gain scattering at long incident neutron wavelength.

Fig.6 shows the variation of the resolution with  $\tau_2$ , if  $L_{12}$  is large,  $L_{SD}$  is small,  $\tau_1$  is large and the latter three parameters are kept fixed; clearly in this case the smallest value of  $\tau_2$  is best adapted to the short distance  $L_{SD}$ . It yields best reso-

lution, although lowest intensity. Finally Fig.7 is an example of the effect of varying  $\tau_1$  at fixed values of  $L_{12}$ ,  $L_{SD}$  and  $\tau_2$ .

## 5. Conclusion

We have presented formulae for intensity and resolution of a TOF-TOF spectrometer employing disk choppers, which can be used to optimize the spectrometer design for given applications. It has not been our aim to actually present such an optimization, but we have shown numerical results for a number of spectrometer configurations, giving an idea of the way in which the essential variables of the spectrometer ( $L_{12}$ ,  $L_{SD}$ ,  $\tau_1$ ,  $\tau_2$ ) together with the wavelengths of incident and scattered neutrons govern its performance. A discussion of the restrictions encountered at a pulsed neutron source regarding the pulse width and rate of the source has shown that conditions can be found under which the peak flux of the source can be fully used, leading to the maximum possible gain factor over a steady-state reactor with the same time-averaged flux for the same type of spectrometer. In certain cases where very specific requirements of the experiment must be fulfilled, the gain factor will be somewhat lower.

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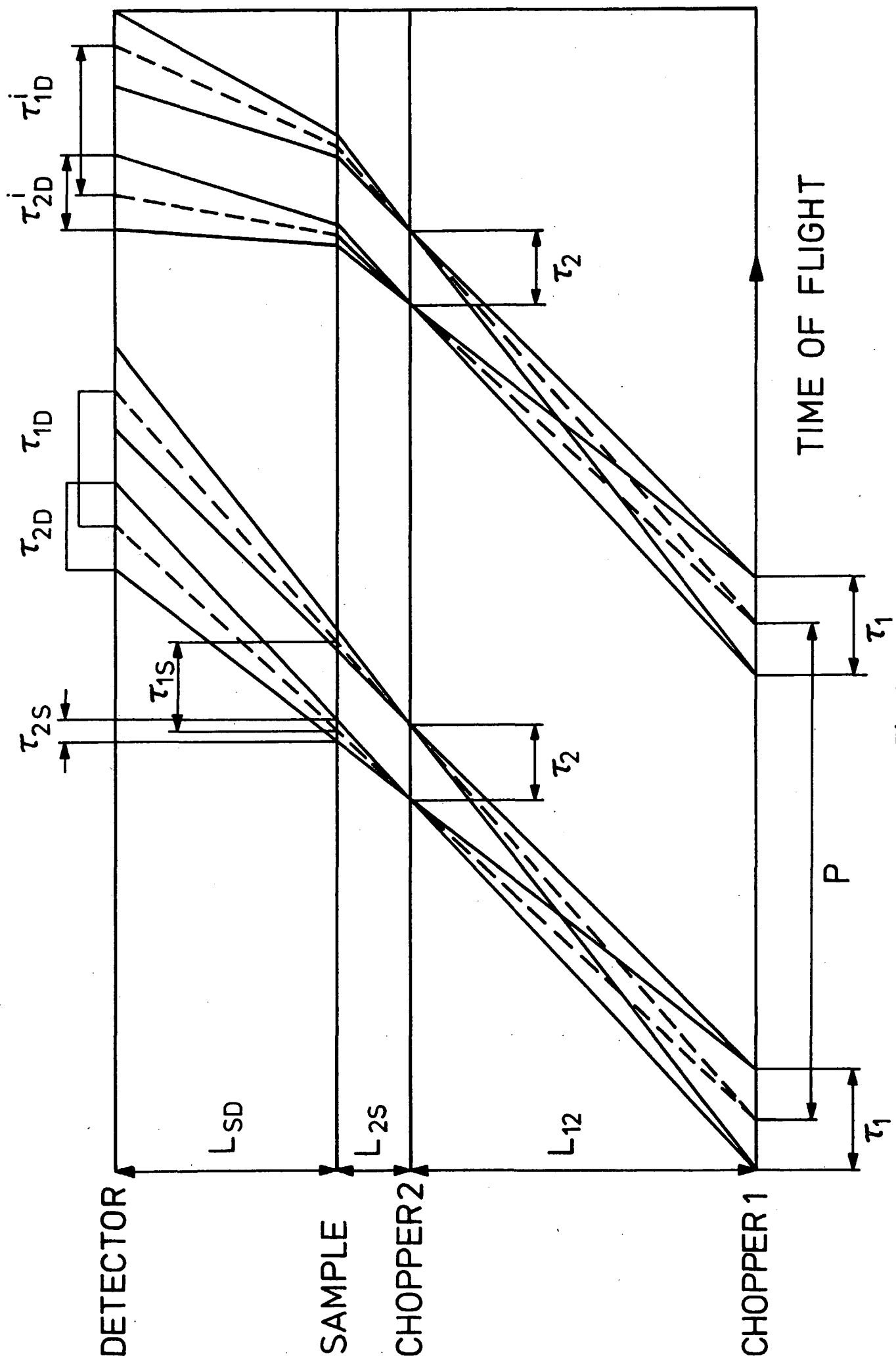


Fig. 1

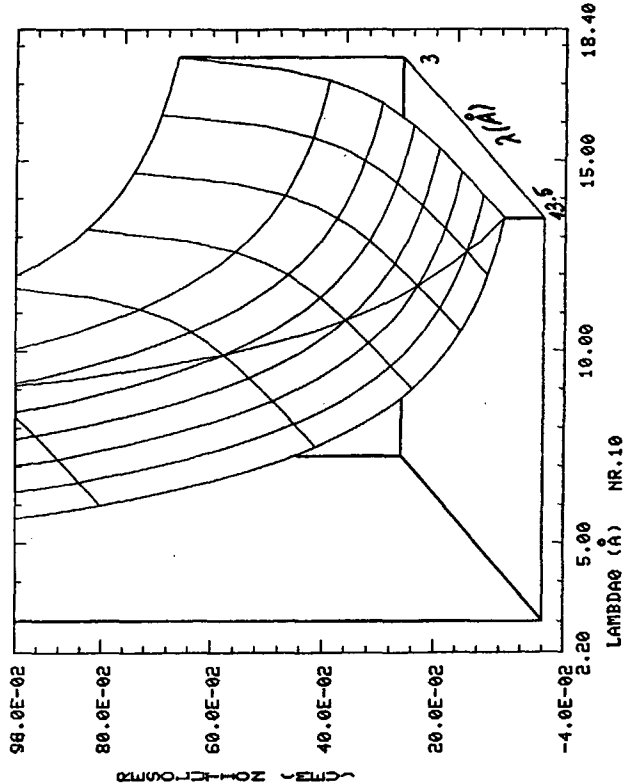
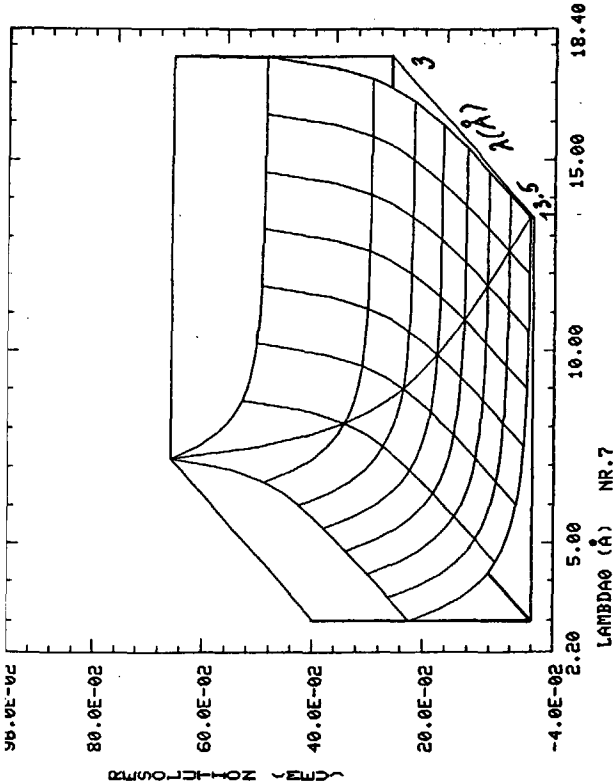
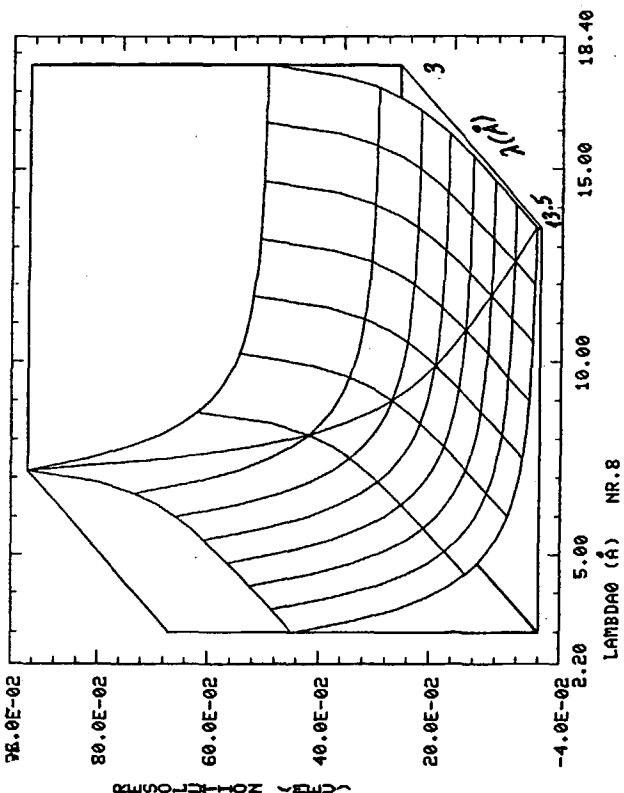
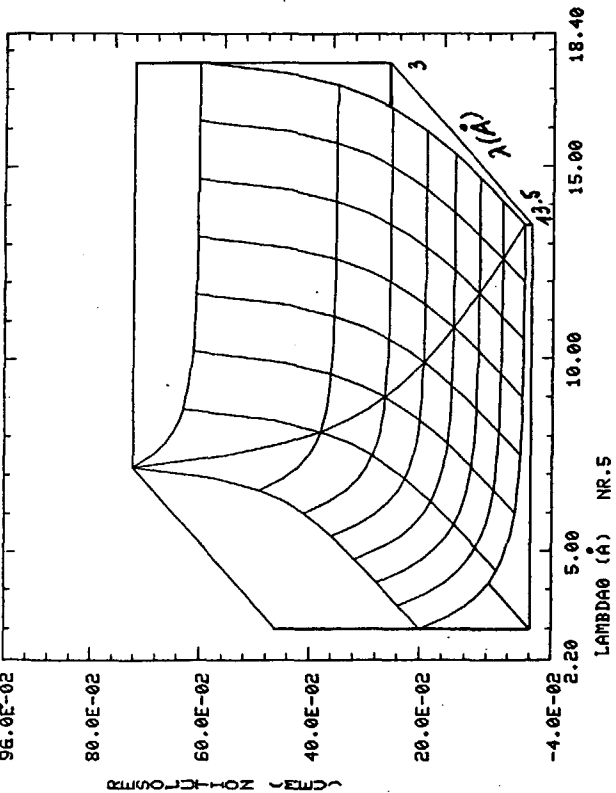


Fig. 2





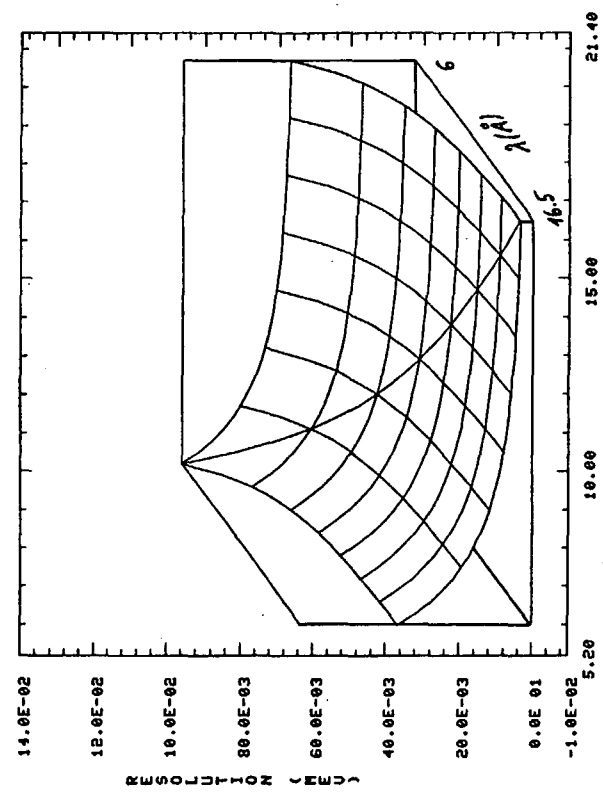
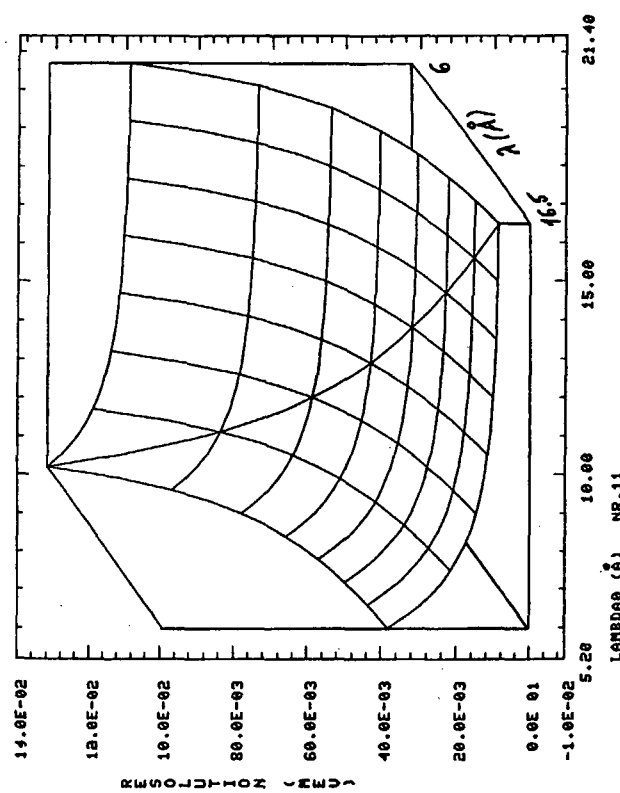
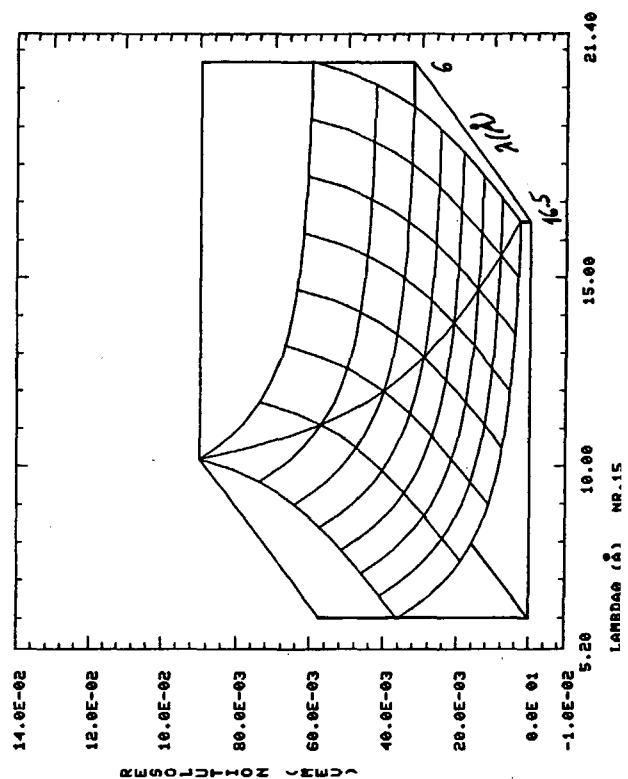
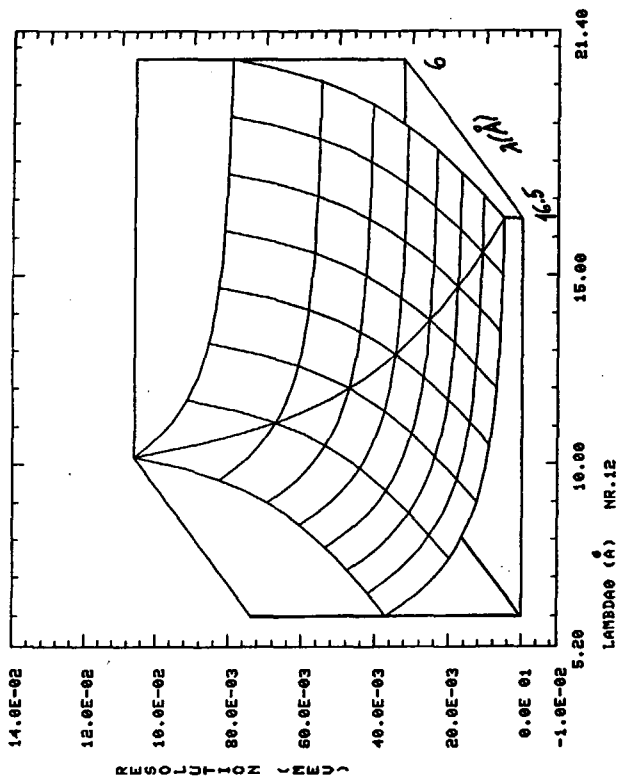


Fig.3

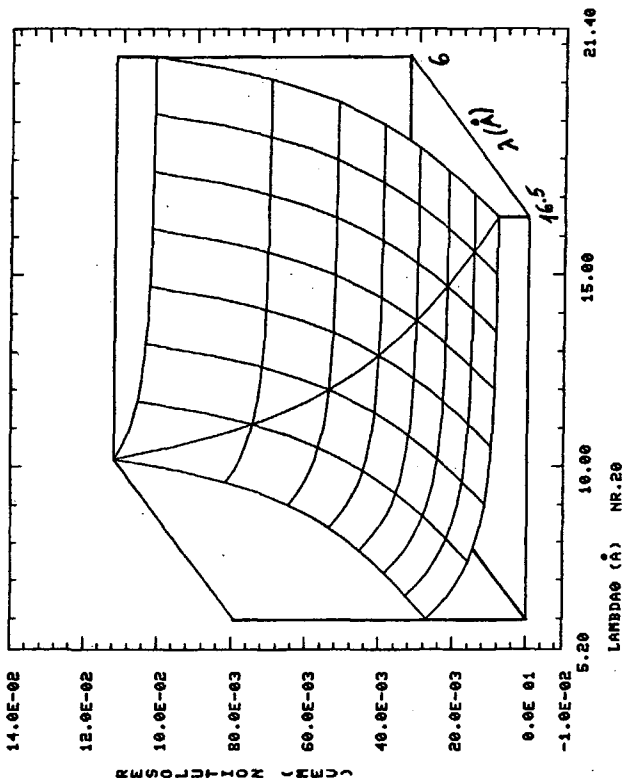
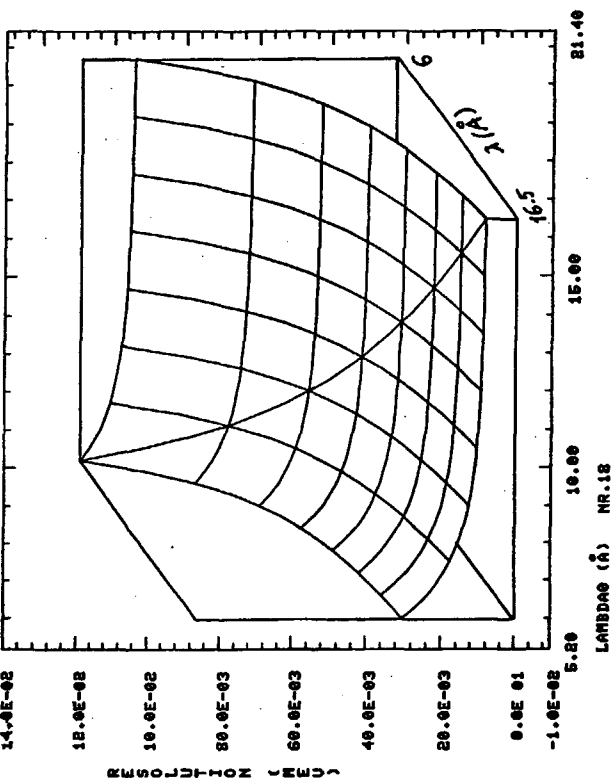
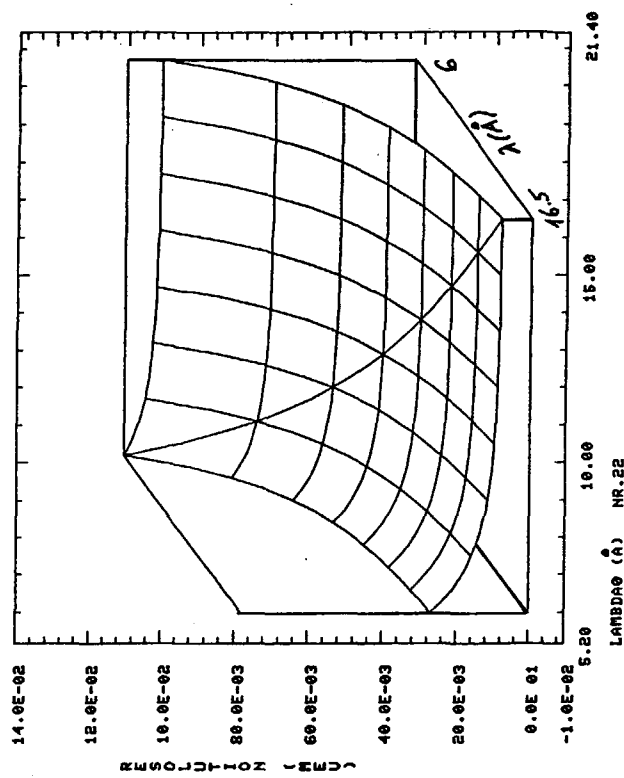
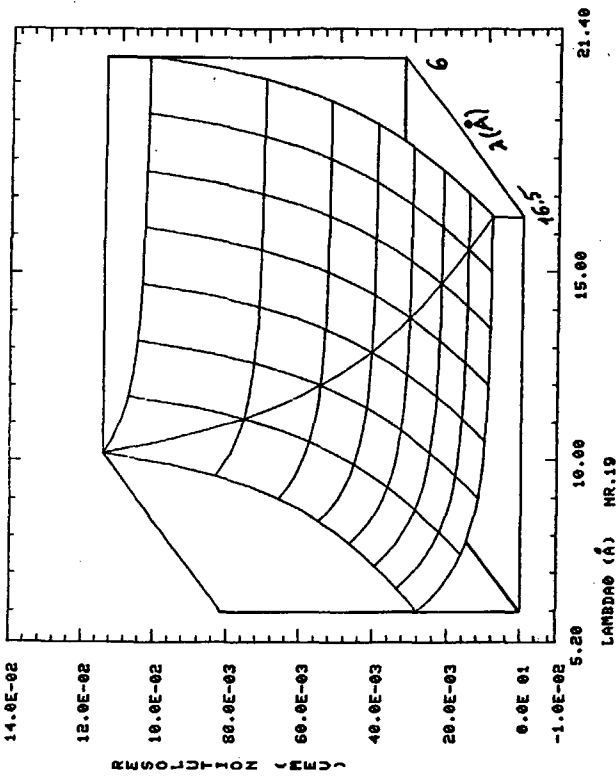
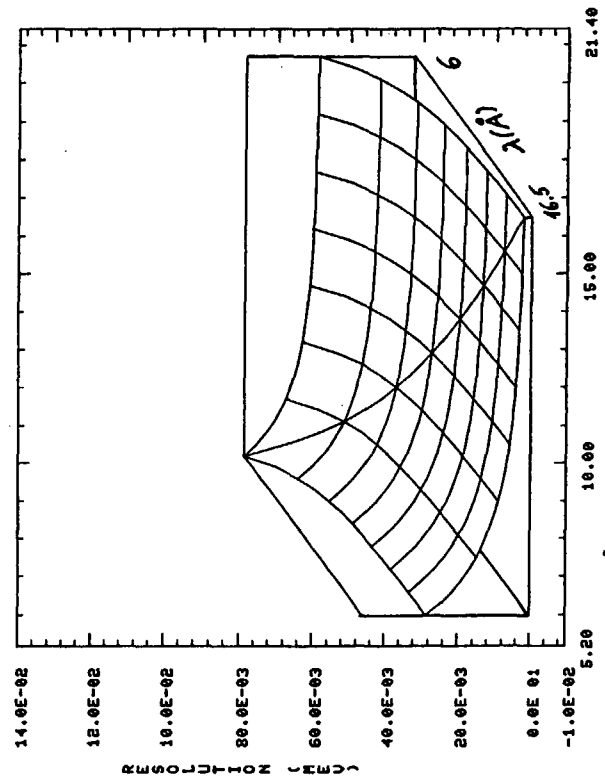
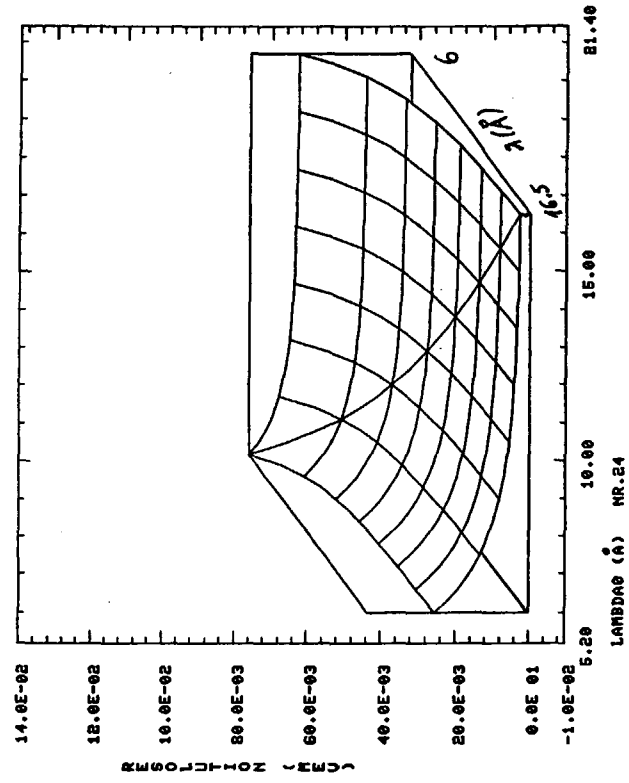
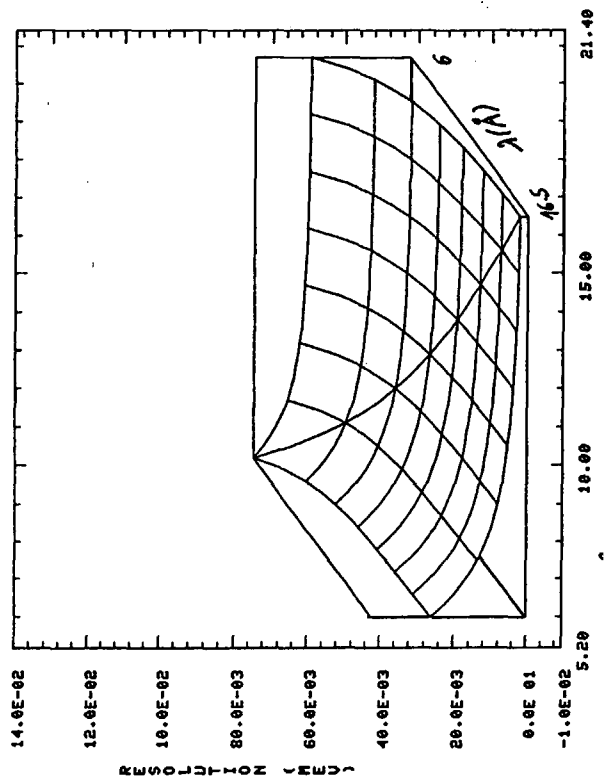
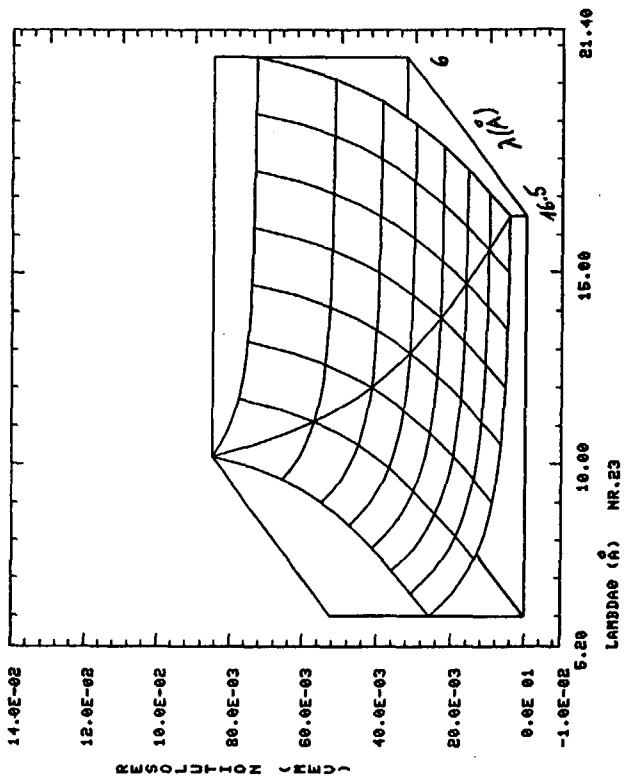


Fig. 4



5. File



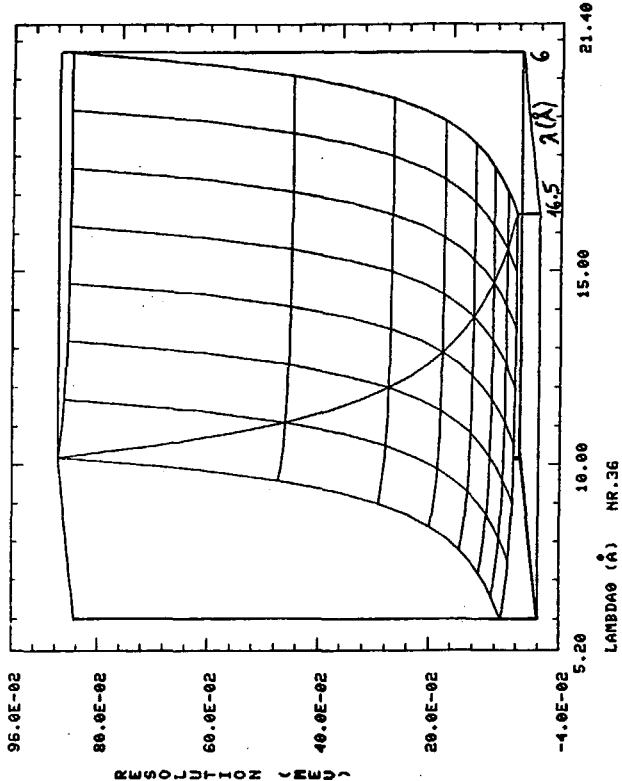
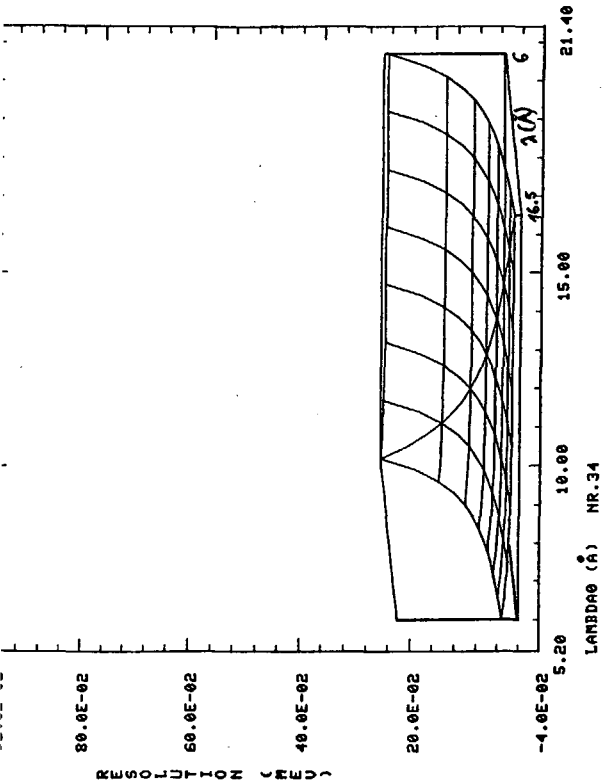
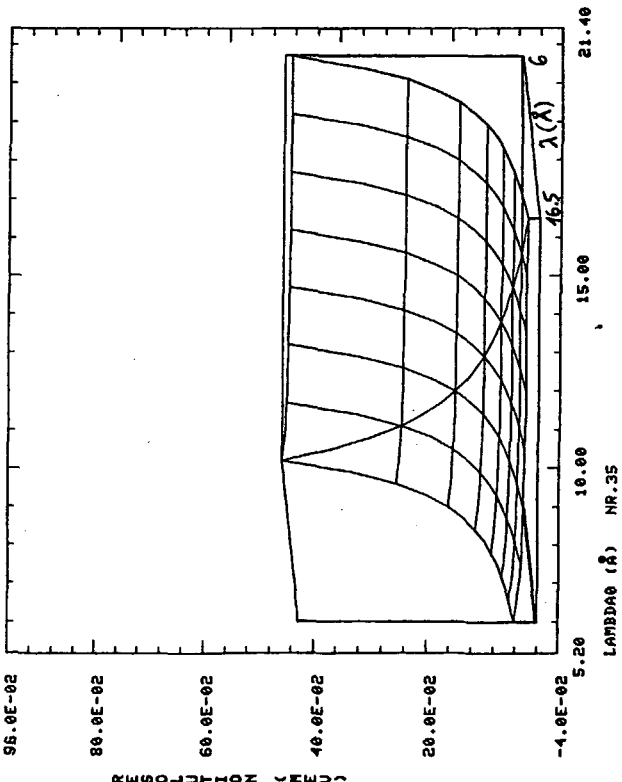
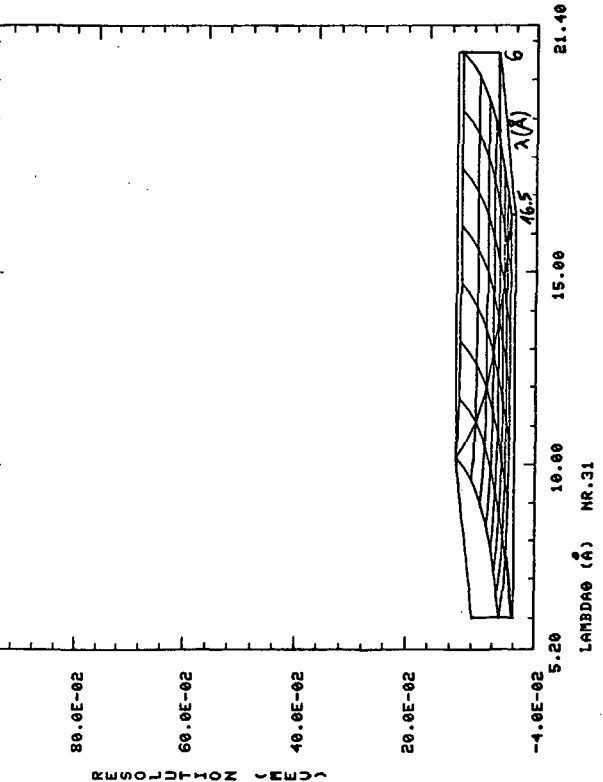


Fig. 6



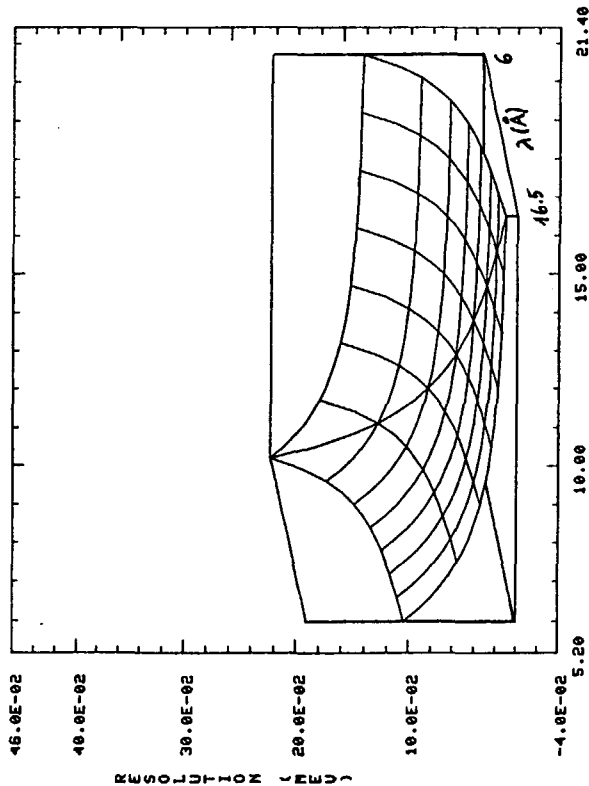
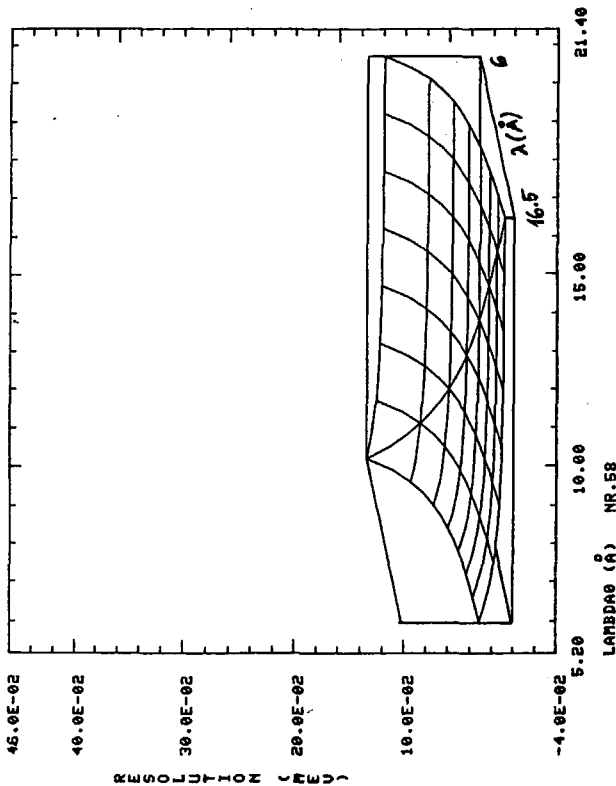
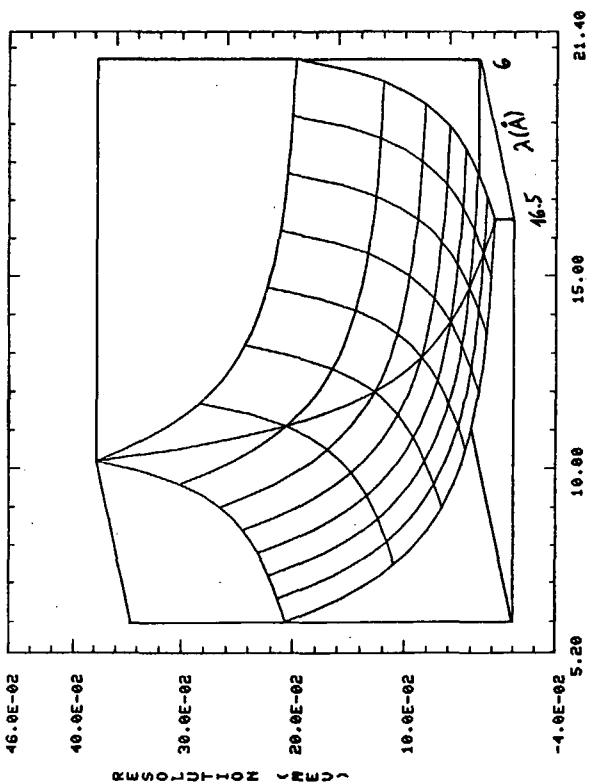
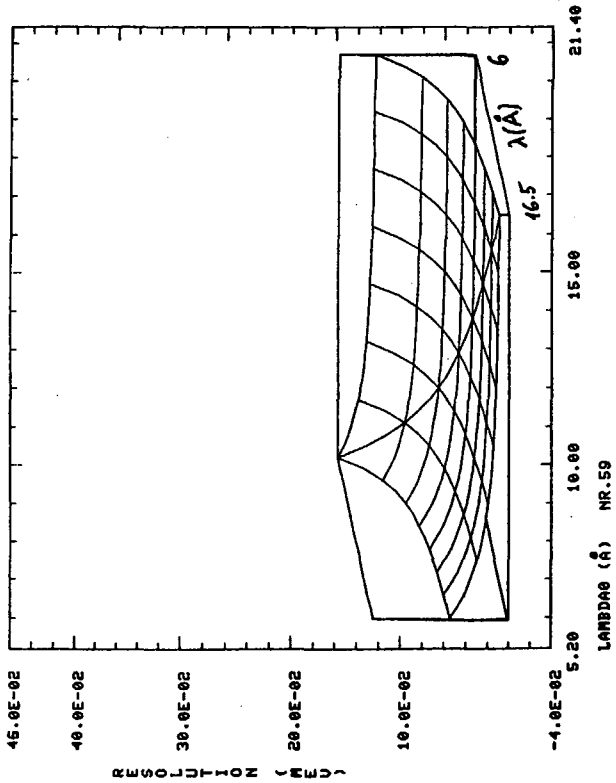


Fig.7

## "High Energy Neutron Scattering"

M. Loewenhaupt, Institut für Festkörperforschung, KFA Jülich

H. Rauh, Physikalisches Institut der Universität Würzburg

V. Wagner, Physikalisch-Technische Bundesanstalt, Braunschweig

### Abstract

Inelastic neutron scattering experiments beyond cold and thermal neutron energies have become feasible since the installation of the hot source at the ILL reactor and even more so since the operation of pulsed spallation neutron sources. In this paper we will briefly review the possible scientific problems in this "high energy area" and discuss the different types of spectrometers which are necessary to cover this field.

For the construction of any of these instruments it is very important that the neutron source offers short pulses. This can only be accomplished by the SNQ with compressor ring and appropriate moderators.

## 1. Introduction

A simple but general rule for inelastic neutron scattering is to do the experiment with the lowest possible incident neutron energy. Increasing neutron energies mean increasing background problems and worse absolute energy and momentum resolution. Therefore the overwhelming amount of inelastic neutron scattering experiments has been performed with cold and thermal neutrons. If however, the physical, chemical or biological sample under investigation has spectral intensity

- (i) at energies far beyond thermal energies, or
- (ii) at moderate energies but low  $Q$  values, or
- (iii) at very large  $Q$  values,

the use of neutrons with high incident energies is required.

For this purpose the hot source of the ILL reactor had been installed extending the offer of neutrons to incident energies around 500 meV, delivered to two TAS (IN1, D5) and two diffractometers (D4, D9). A time-of-flight machine was not considered because of the therefore necessary disadvantageous short pulses (few  $\mu\text{sec}$ ) and long flight paths (several m), leading to a low duty cycle and low intensity compared to the steady state type instruments. A different approach, however, to produce a large amount of neutrons beyond thermal energies has been realised through the operation of pulsed spallation neutron sources (IPNS, Argonne, WNR, Los Alamos; KENS, Japan). If the proton pulse is sufficiently short ( $< 1 \mu\text{sec}$ ) and an appropriate moderator is used then the pulse width  $\tau$  of neutrons from the slowing down region of the spectrum with incident energies  $E_0$  ( $> 250 \text{ meV}$ ) is approximately given by

$$\tau = \frac{2 \mu\text{s}}{\sqrt{E_0/\text{eV}}} \quad (1)$$

This leads to a constant relative energy resolution (independent of incident energy) for the primary flight path  $L_0$  (neglecting all other resolution elements) of:

$$\frac{\Delta E_0}{E_0} = \frac{2\tau}{t_0} = \frac{5.5 \%}{L_0/\text{m}} \quad (2)$$

This gives for incident flight paths of  $\sim 10 \text{ m}$  a reasonably good energy resolution of  $< 1 \%$ . The big gain, however, lies in the fact, that all neutrons produced by the source in the corresponding energy interval and impinged on the sample can be used in the scattering experiment. To give an example:

for  $E_0 = 1$  eV the counting rates of an arbitrary inelastic experiment on the SNQ with compressor ring to SNQ without ring to a reactor with same mean flux at 1 eV scale as

$$5000:40:1$$

assuming the same experimental set-up (same primary and secondary resolution, same repetition rate)! From this it becomes immediately clear that the SNQ with compressor ring will be a unique source for "high energy neutron scattering". The loss factor of 125 ( $= 5000:40 = 250 \mu s:2 \mu s$ ) for the version without compressor ring will settle the SNQ below the SNS, roughly at the level of the IPNS.

## 2. Scientific problems

Due to the fact that "high energy" inelastic neutron scattering has only recently been made possible there is only a small number of established scientific realms compared to conventional thermal and cold neutron scattering. A large part of this field was covered by the workshop on "High Energy Excitations in Condensed Matter" (HEECM) held at Los Alamos National Laboratory 13 - 15 February, 1984 [1,2]. Taking this as guide line we will give a brief review on the interesting subjects:

- A. Magnetism in transition metal (3d, 4d), rare earth (4f) and actinide (5f) systems
- B. Excitations in liquids, gases and amorphous materials
- C. Chemical spectroscopy including localized modes of light atoms in metals
- D. Momentum Distributions (Neutron Compton Spectroscopy) e.g. of quantum liquids and ions in solids.

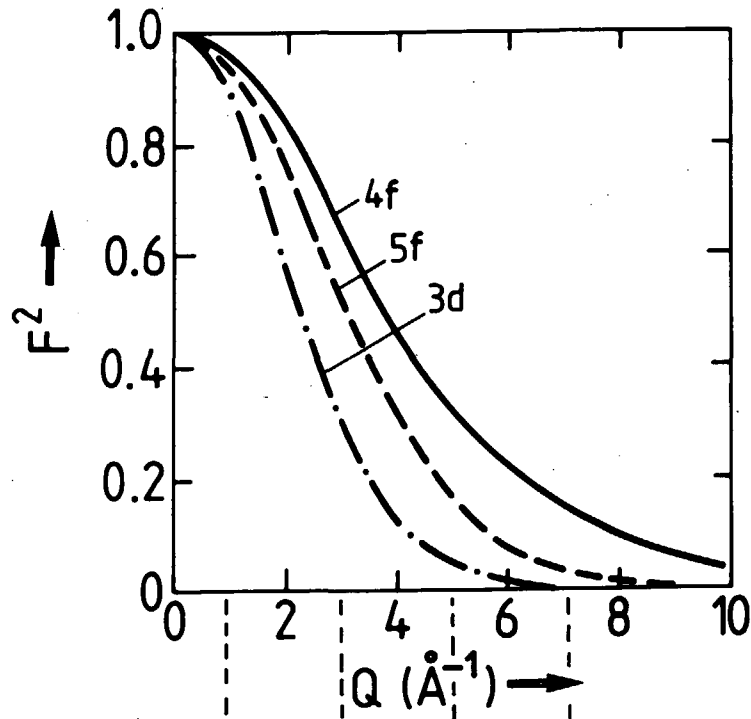
Most of the already performed or proposed experiments in the above specified fields are extensions in energy and momentum transfer into regions which cannot be reached by conventional neutron scattering experiments on reactors. Of more speculative nature are ideas e.g. to investigate electronic excitations in semiconductors. Calculations for the classical semiconductors Si and Ge, however, indicate that the experimental conditions (low cross-section, very high energy transfer of several eV at low Q) will make these investigations very difficult if not impossible.



### A. Magnetism and some considerations concerning counting rates

From many investigations of magnetic excitations in the last two decades in the thermal and cold region it became clear that in most systems the magnetic response has either long tails or additional inelastic intensity extending far into the high energy region. A complete understanding of magnetic phenomena requires the knowledge of the whole magnetic excitation spectrum on an absolute intensity scale. The existence of the high energy part in the magnetic response is due to the fact that magnetism in condensed matter is produced by unpaired electrons. Strong exchange interactions as e.g. in 3d-systems lead to steep collective excitations (magnons) merging into a broad particle-hole continuum (Stoner excitations). Stable moments in f-electron systems, on the other hand, show a sharp single particle excitation spectrum (crystal-field- and spin-orbit-splitting) ranging from  $\mu\text{eV}$  to eV, modified by interactions between different ions in concentrated systems. Stable moments become unstable through a strong hybridization of local (f) and delocalized (conduction) electrons exhibiting a broad and far reaching magnetic response (Kondo effect, intermediate valence phenomena, heavy fermions). Because of the finite spatial extent of the wave functions of the "magnetic electrons" the envelope of all magnetic scattering intensity falls off with increasing  $Q$  like  $f^2(Q) = e^{-\alpha Q^2}$  with  $\alpha$  typically between 0.04 and 0.08 (Fig. 1). Thus the main magnetic scattering intensity is confined to  $Q \leq 4 \text{ \AA}^{-1}$ . If large energy transfers are desired, the kinematics of the scattering process require small scattering angles and rather large incident neutron energies of several times the desired energy transfer (see Fig. 2 and table underneath Fig. 1). These diagrams reveal two rather unfavorable things:

- (a) while for lower incoming energies ( $E_0 < 100 \text{ meV}$ ) the energy transfer in a magnetic scattering experiment can be nearly as large as  $E_0$ , this is not the case for higher  $E_0$ : maximum attainable energy transfers (at  $\theta = 0^\circ$ ) are only 33 % ( $E_0 = 1 \text{ eV}$ ), 11 % ( $E_0 = 10 \text{ eV}$ ) and 3.6 % ( $E_0 = 100 \text{ eV}$ ) of the incident energies (taking e.g.  $Q = 4 \text{ \AA}^{-1}$  as desirable momentum transfer). The necessary incoming energies are proportional to the square of the maximum energy transfer divided by the desired momentum transfer.



|           |                       |                       |      |      |
|-----------|-----------------------|-----------------------|------|------|
| 0.10      | 0.03                  | 14.9                  | 35.1 | 55.2 |
| 1         | 0.15                  | 2.8                   | 9.8  | 15.7 |
| 10        | 0.5                   | 0.8                   | 3.0  | 4.8  |
| 100       | 1                     | 0.4                   | 1.0  | 1.5  |
| 100       | 2                     | —                     | 0.9  | 1.4  |
| E<br>(eV) | $\hbar\omega$<br>(eV) | $\theta$<br>(degrees) |      |      |

**Fig. 1:** Typical  $Q$  dependence of the square of the magnetic formfactor for 3d, 4f and 5f moments (and hence of the magnetic scattering intensity). The table underneath gives the scattering angles  $\theta$  for different sets of  $E_0$  and  $\hbar\omega$  at momentum transfers of 2, 4 and 6  $\text{\AA}^{-1}$ , respectively.

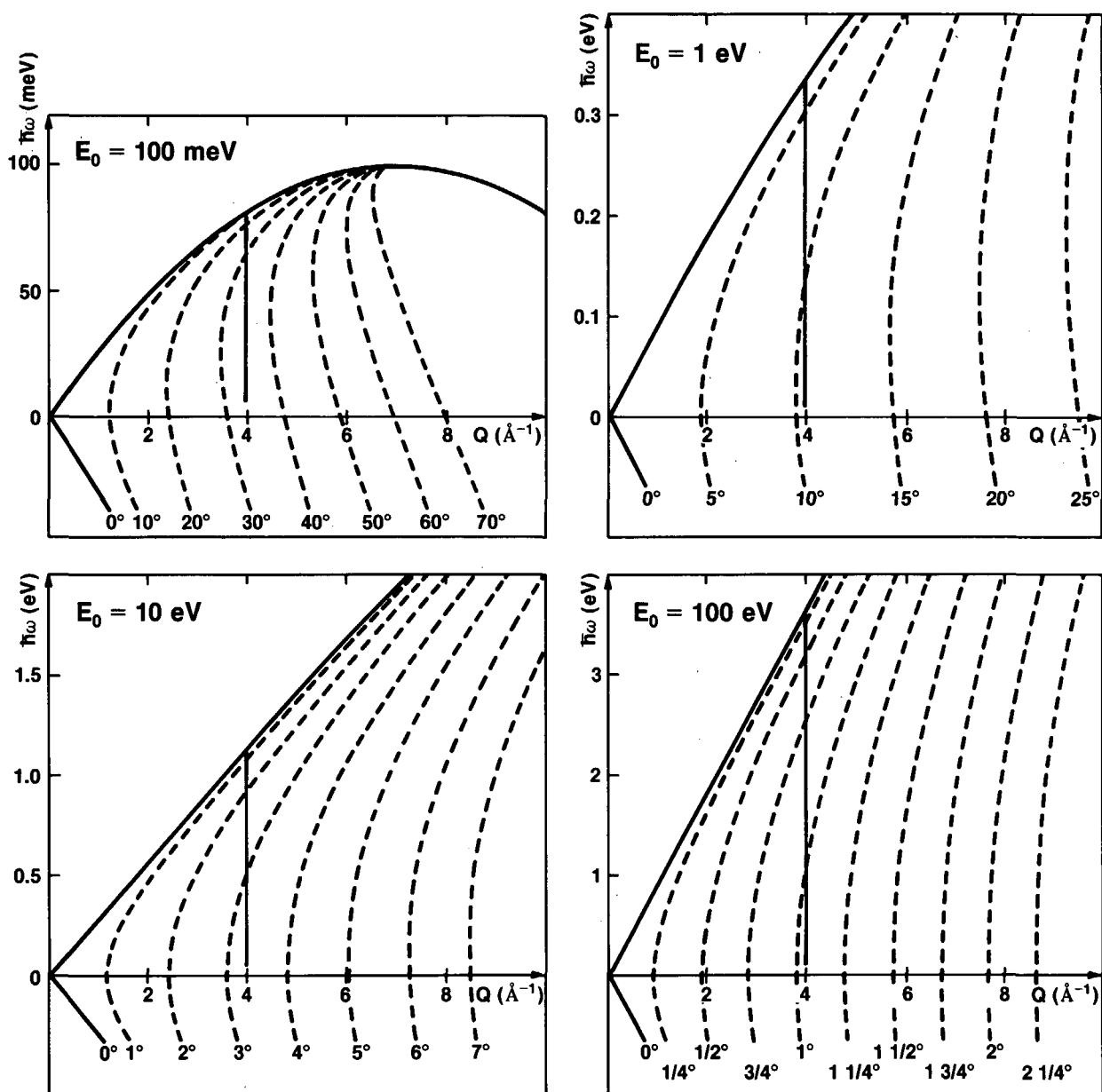


Fig. 2: Diagrams of the accessible  $Q$ - $\omega$ -regions for different incoming neutron energies  $E_0$  in the for magnetic neutron scattering interesting low  $Q$  range. The slope of the  $\theta = 0^\circ$  line at  $(0,0)$  is given by  $\omega/Q \sim 2k_0$ . It determines the necessary incoming energy  $E_{\min} \sim (\hbar\omega_{\text{ex}}/Q_{\text{ex}})^2$  if one wants to measure an excitation with energy  $\hbar\omega_{\text{ex}}$  at momentum transfer  $Q_{\text{ex}}$ .

- (b) the scattering angles become drastically smaller for increasing energy transfers; the step from a few degrees ( $\hbar\omega \approx 0.5$  eV) to only a few tenths of a degree ( $\hbar\omega \approx 2$  eV) will of course have severe consequences for the design of any such instrument.

To illustrate the rapidly increasing difficulty in performing high quality high  $\hbar\omega$  measurements at low  $Q$  we will give a general expression for the counting rate in the detector

$$\dot{C} \sim \dot{S} \cdot f_0 \cdot d\Omega_0 dE_0 \left[ N \frac{k_1}{k_0} S(Q, \omega) \right] d\Omega_1 dE_1 f_1 \quad (3)$$

where the different factors characterize:

- (a) the source where  $\dot{S}$  is the flux per solid angle and energy interval; in the slowing down region this is proportional to  $1/E$ .
- (b) the incoming and outgoing collimation which is chosen to be about equal and mainly given by the perpendicular components of the momenta; these in turn determine the  $Q$ -resolution (because of the small scattering angles we have to assume a point geometry instead of the usually used slit geometry where the collimation perpendicular to the scattering plane may be more relaxed than in the plane).

$$d\Omega_0 \approx d\Omega_1 = \left( \frac{k^\perp}{k} \right)^2 \approx \left( \frac{dQ^\perp}{k} \right)^2 \quad (4)$$

- (c) the incident and final energy resolution  $dE_0$  and  $dE_1$  which again should be equal and of the order of  $dE \approx 0.1$  eV or better (this requires  $dE/E < 10^{-3}$  for  $E_0 = 100$  eV or  $L_0 > 55$  m!).
- (d) attenuation of the beam before and after the scattering; the factor  $f_0$  may not depend on the neutron energy;  $f_1$  contains among others the counter efficiency  $\varepsilon$ .
- (e) the scattering law  $S(Q, \omega)$  and the number  $N$  of scatterers in the beam; for the case of high energies we have  $k_1 \approx k_0$  and the expression in the brackets independent of the neutron energy.

Counting all energy or momentum dependent factors together we obtain (with  $E_0 \approx E_1 \approx E \sim k^2$  and  $E \sim (\frac{\omega}{Q})^2$  as mentioned before).

$$\dot{C} \sim (E^{-3} \text{ or } k^{-6} \text{ or } (\frac{Q}{\omega})^6) * \varepsilon * (dE)^2 (dQ^\perp)^4 \quad (5)$$

This strong dependence of the counting rate on the neutron energy makes any significant increase in energy transfer at low  $Q$  or decrease of  $Q$  at high  $\hbar\omega$  immediately to a severe intensity problem, unless relaxed  $Q$  and  $\hbar\omega$  resolutions may be tolerated.

From the experience of experiments on existing spallation sources we estimate an upper limit of 0.5 to 1 eV of energy transfers at momentum transfers around 2 to 5  $\text{\AA}^{-1}$  as achievable in inelastic neutron scattering using the SNQ with compressoring.

#### B. Excitations in liquids, gases and amorphous materials

Because of the disordered nature of these systems any sharp excitations are confined to low values of the total momentum transfer (exception: superfluid He). Though the typical energy transfers are quite moderate high incident energies are required because of the kinematical restrictions if low  $Q$  values are desired.

A similar consideration concerning the counting rates as demonstrated for high energy magnetic scattering applies for the above experiments.

#### C. Chemical Spectroscopy

In this field competition with optical spectroscopy methods is heavy but neutrons take advantage in many cases because

- (i) there are different selection rules
- (ii) special atoms can be labelled by appropriate D/H substitution
- (iii) in-situ experiments can be performed because neutrons see e.g. through even complicated sample containers or catalyser supporting material.

Furthermore inelastic neutron scattering is a unique tool in the investigation of local vibrations of H in metals because of the high incoherent cross-section of H for neutrons. The typical energy transfers from molecular vibrations or H-oscillations in metals are below 0.5 eV (H - H vibrations). In some cases, where the determination of higher harmonics is of interest, also higher energy transfers may be desirable. The formfactor  $Q^2 e^{-\langle u^2 \rangle Q^2}$  of these vibrations are maximal for  $Q$ -values between 5 and 8  $\text{\AA}^{-1}$  (50 to 130 meV). Thus, in general, large energy transfers together with large momentum transfers are suitable. These requirements are met best by inverse filter or crystal spectrometers which in addition yield high intensity (Be,

BeO filter or crystal analyser CAT, KENS). For special cases like line width measurements, where multiphonon contributions are important, also small  $Q$  high energy transfer measurements would be interesting which could be achieved best by chopper instruments.

#### D. Momentum Distributions

In the impulse approximation (large  $Q$  values where recoil energy  $\gg$  effective temperature of the system) the scattering function reveals directly the momentum distribution  $n(\vec{p})$  of the scattered nuclei ("Neutron Compton Scattering"). The first experiments done in this field [1] comprehend quantum liquids (superfluid  $^4\text{He}$ ), light atoms (pyrolytic graphite), light and heavy water. At present experimentalists concentrate on the determination of the Bose condensate fraction  $n_0$  in liquid  $^4\text{He}$ . First results, namely  $n_0(T)$  at constant pressure, have been gained, but many interesting problems are still open, for example the dependence of  $n_0(T)$  upon pressure or upon the particle density (dilution with  $^3\text{He}$ ), interaction between Bose and Fermi gas ( $^4\text{He}$   $^3\text{He}$ ), and others. Furthermore it should be noted that the standard example  $^4\text{He}$  is not the only existing Bose system, and new features may arise by studying new systems like e.g. monoatomic hydrogen gas. Not only  $n(\vec{p})$  itself is of interest: Noting that  $n(\vec{p})$  is an ensemble average over the Fourier transform of the one-particle wave functions, Schrödinger's equation may be used to calculate the single particle potential from  $n(\vec{p})$  [G. Reiter in Ref. 1]. This is of particular interest for H in doublewell potentials where diffraction cannot distinguish between static and dynamic disorder.

Up to now, high resolution work at  $Q$  values corresponding to the lower edge of the impulse approximation regime have been performed at IPNS using a chopper spectrometer, and lower resolution work at  $Q$  values well within the impulse approximation have been performed at KENS and Los Alamos using a resonance detector and resonance filter instrument, respectively, both in inverse geometry.

### 3. "High Energy" Spectrometers

To cover the field of high energy neutron scattering we propose to consider three types of spectrometers:

- (i) a direct geometry chopper spectrometer with  $E_0$  up to 1 eV, possibly up to 2 or 3 eV, (A,B,C,D)
- (ii) an inverse geometry high intensity, Be, BeO filter or crystal analyser spectrometer (C)
- (iii) an inverse geometry resonance filter or resonance detector spectrometer (A,D).

#### Chopper instruments

Chopper time-of-flight spectrometers in direct geometry appear to become the working horses of high energy neutron scattering on pulsed sources (LRMECS, HRMECS at IPNS). To define the incident energy a chopper in front of the sample has to be tuned in phase to the source pulse. This can be done quite easily (chopper and source should have the same master clock) and it is superior to a solution with monochromator crystals because of low crystal reflectivity and because no parts of the secondary spectrometer have to be moved to change  $E_0$ . The final energy is determined from the time-of-flight from sample to detectors. A detailed description for such an instrument for low angle scattering is given by W. Reichard [3]. A more general instrument, however, should also have detectors at larger scattering angles to cover a larger  $(Q, \omega)$ -space e.g. for experiments on momentum distributions of light nuclei. Even in the case that the physics of interest is confined to the low angle region it is often important to have information about the scattering pattern at high  $Q$  values (e.g. to control sample induced background from high  $Q$  multiple scattering; control of phonon scattering and magnetic scattering a.s.o). Concerning the design of the instrument, similar considerations as in the case of thermal time-of-flight machines [4] apply here. We need:

- a. a pulse cutter, as near as possible to the source
- b. a background chopper
- c. an incident energy defining chopper as near as possible to the sample
- d. detectors in different groups
  - $\alpha$  . at low angles ( $1^\circ - 15^\circ$ ) but large distance from the sample covering

as much as possible of the available solid scattering angle, and  $\beta$  at higher angles ( $15^\circ - 120^\circ$ ) but shorter distance from the sample.

A sketch of such an instrument is given in Fig. 3.

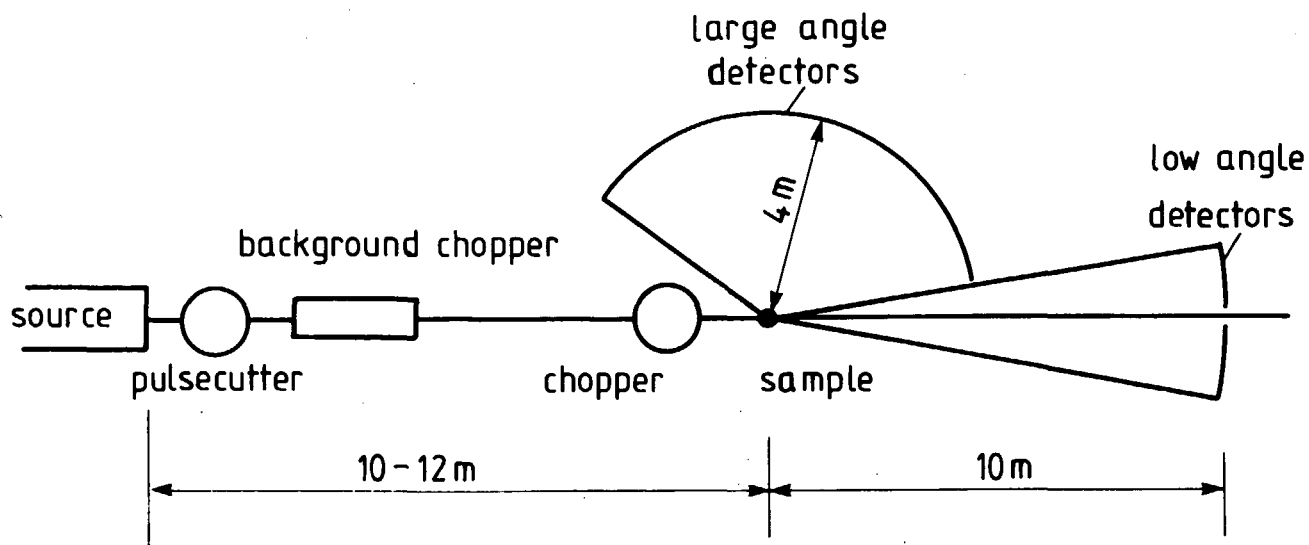


Fig. 3

#### Filter or crystal analyser spectrometers

Inverse geometry instruments are specially well suited for pulsed neutron sources (small dimensions of secondary spectrometer; use of all incident  $E_0$ 's in the selected energy band). The incident energy resolution is given by the distance source (or pulse cutter) to sample and its relative value is independent of  $E_0$  for epithermal neutrons (see Introduction, assuming a negligible short proton pulse). In a filter spectrometer, the secondary energy resolution is given by the cut-off energy of the filter (Be: 5.22 meV, BeO: 3.76 meV). Only neutrons with  $E_1$  smaller than the cut-off energy will be seen by the detector. Problems arising from large uncertainties in the secondary TOF (and hence exact determination of  $E_0$ ) may be overcome using a Be/BeO difference method [5].



A sketch of a filter spectrometer is given in Fig. 4.

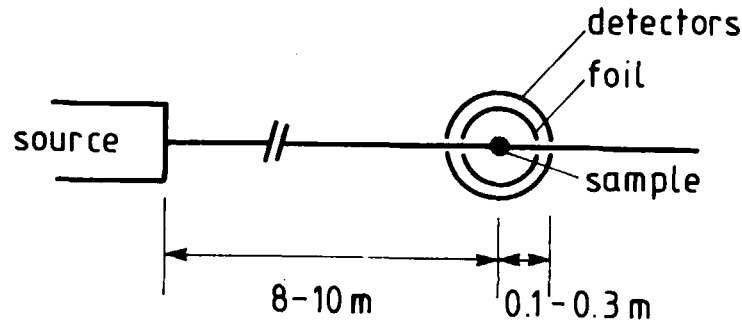


Fig. 4

Instead of a filter one can also use a crystal (or a set of crystals) to define  $E_1$  (CAT, KENS, Fig. 5; CAS, IPNS). A spectrometer with many independent crystal analysers is not considered here. It will be discussed separately for the case of collective excitations by H. Rau and R. Geick [6]. There is one common feature in the above described instruments:

$$E_1 \ll E_0 \approx \hbar \omega \quad \text{and thus } Q \approx k_0 = \sqrt{\frac{2m}{\hbar^2}} \omega$$

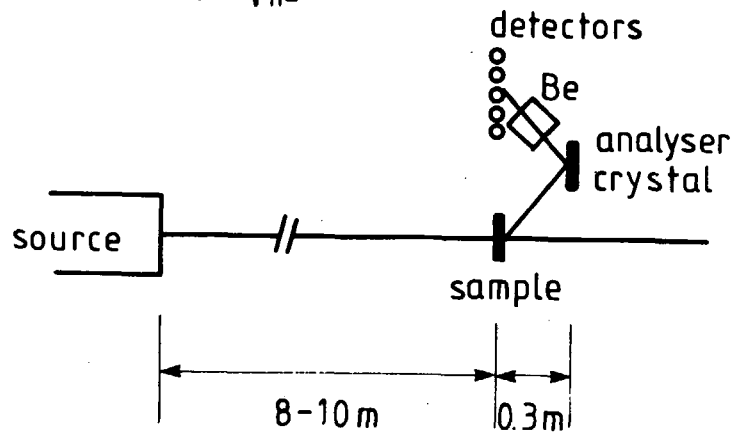


Fig. 5

#### Resonance Detector (RDS) and Resonance Filter Spectrometers (RFS)

Again inverse geometry must be used (see preceding discussion). Resonances occur at high energies (0.1 eV up to 100 eV, [ 7 ] ). The resonance energy determines  $E_1$  if the resonance foil is placed between sample and detector (Fig. 6a).

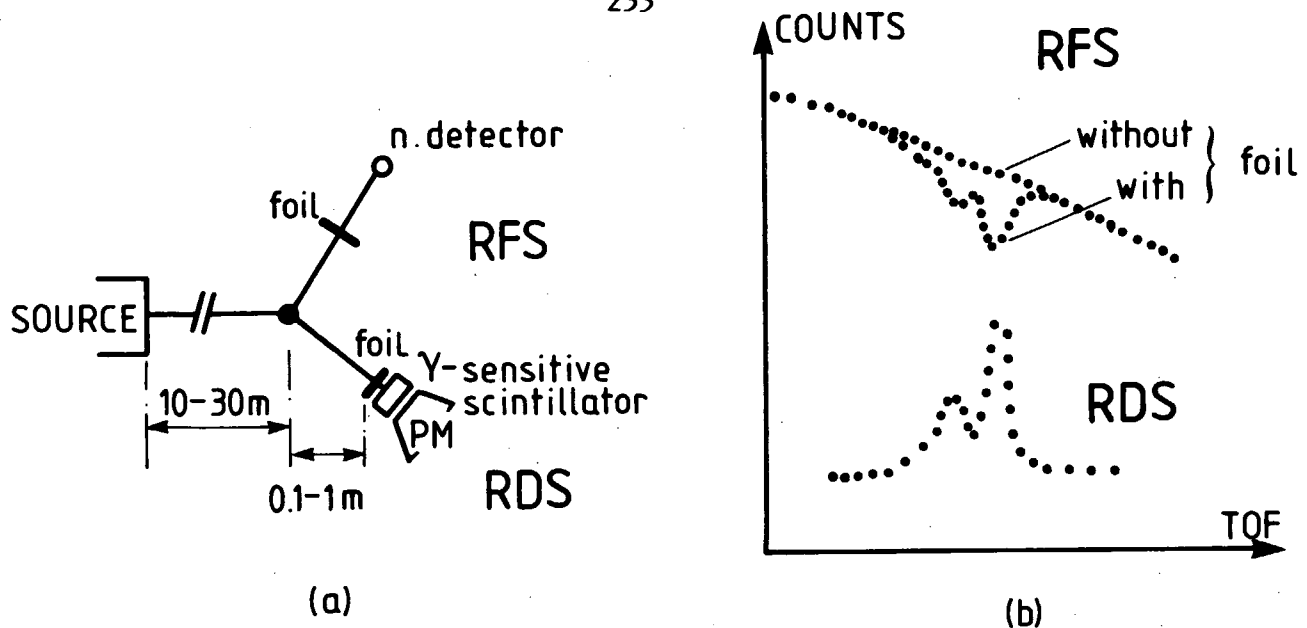


Fig. 6

In a resonance detector the  $\gamma$ 's after a neutron capture are registered while in a resonance filter spectrometer all scattered neutrons are recorded in a neutron detector. In the first case the spectrum is obtained directly while in the second case the spectrum is obtained from the difference of measurements with and without filter (Fig. 6b).

As a direct consequence we note that an RDS is the superior concept, especially for line-profile analysis measurements like the determination of momentum distribution (direct measurement: low statistical error at low counting rates!). The absolute energy resolution is mainly determined by the pulse width (Equ. 1) and the resonance width of the filter. The energy resolution may be improved by cooling the filter (best experimental value: 53 meV for  $^{238}\text{U}$  at 25K) or removing the Lorentzian wings of the resonance line via difference spectra of two foils of different thickness or temperature [8,9]. Up to now, successful experiments have been performed in (high  $Q$ )-scattering. (Low  $Q$ , high  $\hbar\omega$ )-experiments still suffer from background problems (IPNS, RAL).

Because of the large  $E_1$ , in principle any region of the available ( $Q$ ,  $\omega$ ) space can be reached. Thus with resonance absorbers we do not encounter the kinematical problems of the  $E_1 \approx 0$  filter spectrometers.

#### 4. Conclusion

There is already now and there will be even more interest in the future to explore the scattering law  $S(Q, \omega)$  at higher energy transfers and/or larger momentum transfers compared to conventional inelastic neutron scattering at reactors using the unique features of neutron spallation sources. We discussed the possibilities and limitations of various fields of scientific research and proposed three different types of instruments to cover these fields. All proposed instruments need short neutron pulses from the source which can only be accomplished by the SNQ with compressor ring and appropriate moderator. A repetition rate of  $f_s = 100$  Hz is quite reasonable and the performance of the instruments is practically not affected by a variation of  $f_s$  by a factor of 0.1 to 3 (assuming a constant average proton beam current). The pulse width of the SNQ without compressor ring ( $> 250$   $\mu$ s) is in any case too long to be taken as energy defining component.

#### 5. Miscellaneous

Finally some general remarks concerning experiments with "high energy" neutrons:

1. Cd and Gd are no longer useful as shielding materials; one has to use  $B^{10}$  enriched materials ( $B_4C$ , BN..) which are more difficult to handle and often a source of background itself.
2.  $^3He$  counter efficiencies drop with increasing neutron energy (typical: black for cold and thermal neutrons, 10 to 20 % for 1eV); this may be compensated by higher pressure of the  $^3He$  gas, thicker counters or an array of counters. Above 1 eV resonance foils or  $^6Li$  based glass should be used.
3. As already mentioned in the introduction background conditions become worse with increasing neutron energy. This is especially annoying for small scattering angles where the signal is usually rather weak and dominated by sample induced background. Sample induced background originates from multiple scattering processes at higher  $Q$  values. Buyers in [1] estimated that this background increases with  $k_o^6$  or  $E_o^3$ .
4. Doing an experiment with high energy neutrons one should always find an optimum between
  - a. sufficient scattering intensity (e.g. drop of magnetic intensity with increasing  $Q$ )

- b. lowest possible incident energy to reduce background and increase resolution.

Therefore it is important that the incident energy of a spectrometer can be easily changed.

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NIM = Nucl. Inst. Methods in Physics Research

Computer Simulations of Phonon Measurements on a Multi Detector  
Time-of-Flight Spectrometer

W. Reichardt

Kernforschungszentrum Karlsruhe

Institut für Nukleare Festkörperphysik

P.O.B. 3640, D-7500 Karlsruhe, FRG

ABSTRACT

In order to help discussions about the value of single crystal measurements on a TOF-spectrometer with a multi-detector we have simulated such experiments on a computer. Some results are given for a measurement of the phonon-dispersion relation in  $\text{ZrFe}_2$  on a direct geometry TOF-spectrometer. In addition, general problems connected with TOF-spectrometry for measurements of collective excitations in single crystals are discussed.

1. There is no possibility for an optimized measuring program in order to select those phonons which are of primary importance for an understanding of the lattice dynamics of the system. One has to take those data one can get, among which there may be many which are insignificant or redundant.
2. The cross-section for one phonon scattering varies strongly as a function of both the momentum transfer  $\vec{Q}$  and the energy transfer  $\hbar\omega$ . Thus there may be many detectors where the statistical accuracy is insufficient.
3. In most systems the separation of neighboring phonon branches is smaller than the experimental resolution. Therefore often superpositions of several phonon groups will be observed.
4. The resolution function is the same for all scattering angles. Furthermore there may be many cases, where the measuring path in the  $\hbar\omega$ - $\vec{Q}$ -space cuts unfavorably through the dispersion surface. This will be explained in section 2.

In order to help discussions about the value of phonon data obtained with a multidetector time-of-flight spectrometer we have written a computer program which simulates such experiments. First results obtained with this program will be presented in this report.

## 2. PHONON MEASUREMENTS BY TIME-OF-FLIGHT

The measurement of a phonon by a direct geometry time-of-flight spectrometer is explained in Fig. 1. The angle  $\psi$  between the coordinate system of the sample and the incident beam, the magnitude of  $k_i$  and the scattering angle  $\phi$  determine the neutron measuring path in the  $\hbar\omega$ - $\vec{Q}$  space. A special phonon is recorded at the intersection of the neutron measuring

path with the dispersion curve  $\hbar\omega(\vec{Q})$  along the direction of  $\vec{k}_f$ . The energy and the wave-vector of the phonon are not decoupled. In a time-of-flight experiment  $\vec{Q}$  can only be chosen to lie on a straight line in the reciprocal lattice, both the position on this line and the phonon energy are determined simultaneously by the experiment.

As is shown in Fig. 2 there are favorable measuring conditions yielding sharp neutron groups, if the two curves intersect close to  $90^\circ$ . If the two curves have similar slopes (Fig. 2b) broad distributions will be obtained.

### 3. CALCULATIONAL METHOD

In order to simulate measurements of phonon dispersion surfaces by the time-of-flight technique a special subroutine was incorporated into our general lattice dynamical computer program "GENAX1". Input data are the definition of the scattering plane, the orientation of the sample relative to the incident neutron beam  $\psi$ , the scattering angles  $\phi$ , the primary energy  $E_i$  and the parameters determinating the energy resolution of the experiment. The phonon frequencies and the dynamical structure factor are calculated at many points along the direction of  $\vec{k}_f$  corresponding to equidistant  $\hbar\omega$ -values. The distribution of the scattered neutrons is determined by sampling the dynamical structure factors within a band given by the experimental resolution in energy. For simplicity the resolution function is assumed to be triangular. The full width-half-maximum of this triangular function is given by

$$\Delta\hbar\omega = 8.747 \cdot 10^{-4} \left\{ \Delta t_P^2 \left( \frac{E_i^{3/2}}{L_1} + \frac{E_f^{3/2}}{L_2} \frac{L'_1}{L_1} \right)^2 + \Delta t_{ch} \left( \frac{E_i^{3/2}}{L_1} + \frac{E_f^{3/2}}{L_2} \frac{L'_1}{L_1} + \frac{E_f^{3/2}}{L_2} \right)^2 \right\}^{1/2}$$

$t_p, t_{ch}$ : time resolution of source pulse and chopper pulse, respectively (in  $\mu s$ ).

$L_1, L'_1, L_2$ : distances source-chopper, chopper-sample, sample-detector, respectively (in m).

#### 4. RESULTS FOR $ZrFe_2$

As an example we have chosen the Laves phase compound  $ZrFe_2$  for which a good lattice dynamical model had been obtained from recent measurements on a triple axis spectrometer (1). We chose this compound because the dispersion curves are of intermediate complexity. The lattice is cubic, there are 6 atoms in the elementary cell. Both Zr and Fe are good scatterers. The dispersion curves calculated from the model are shown in Fig. 3. The maximum phonon energy in  $ZrFe_2$  is about 40 meV. In all calculations we have set the temperature of the sample equal to zero. In most cases the resolution was determined from the following parameters:  $L_1 = 10$  m,  $L_2 = 4$  m,  $L'_1 = 0$ ,  $\Delta t_p = \Delta t_{ch} = 30 \mu s$ . The width in time of the source pulse  $\Delta t_p = 30 \mu s$  requires the application of a tail-cutter. The resulting resolution  $\Delta E$  is plotted in Fig. 4. It is comparable to resolutions used in today's measurements on triple axis spectrometers. In Figs. 5.1 - 5.14 and 6.1 - 6.12 we present results for two different scattering configurations, where we have selected the 110-plane and the 001-plane, respectively, as scattering planes. This restricts the number of observable phonon branches to 11 for the 110-plane and to 9 for the 100 plane. The scattering angle ranged from  $70^\circ$  to  $130^\circ$ . This is the most valuable range of scattering angles for phonon measurements on a triple axis spectrometer. The primary energy  $E_i$  was 60 meV.

In the first case the orientation of the sample ( $\psi=20^\circ$ ) was chosen in such a way that the highest dispersion sheet near 40 meV can be measured with a favourable cross-section at the 822 reciprocal lattice point. The corresponding scattering angle  $\phi$  is about  $122^\circ$ . Results are shown for a dense mesh of scattering angles



around  $122^\circ$  ( $\Delta\phi=1^\circ$ ) in order to investigate the range in which useful information can be obtained for the highest dispersion sheet. A coarser mesh between  $70^\circ$  and  $130^\circ$  demonstrates which additional phonon data can be picked up in other Brillouin zones.

In the second "experiment" we aimed at an optimum cross-section for the second highest dispersion sheet at the 660 reciprocal lattice point ( $\psi=70^\circ$ ,  $\phi=116^\circ$ ).

In some figures we have marked by arrows those positions, where peaks should be expected in case of no broadening due to resolution. There is a great number of peaks arising from contributions of two or more branches. However, there are several cases, where the broadening of the peaks is caused by an unfavourable cut of the neutron path through the phonon branch. This may lead to a wrong determination of the phonon energy (see Fig. 5.2).

## 5. SUMMARY AND CONCLUSIONS

It was the purpose of our investigations to demonstrate for a specific example the performance of phonon measurements on a direct geometry time-of-flight spectrometer with a multi-detector. In the computer simulation realistic instrument parameters were used and the "experiment" was performed under conditions, which may be a reasonable recipe for a real experiment. Those conditions were:

1.  $E_0 \approx 1.5 \hbar\omega_{\max}$
2. range of scattering angles:  $70^\circ \leq \phi \leq 130^\circ$
3. use of high symmetry planes as scattering planes. This reduces the number of dispersion sheets that contribute to the scattering
4. the orientation of the sample relative to the primary beam was chosen in such a way, that the optimum intensity was obtained for selected parts of the dispersion surface, which are the hardest to be measured. (Those are the highest phonon branches in most cases). In this way many

additional data can be picked up, which, although not measured with an optimum cross-section, have comparable statistical accuracy.

The latter condition differs from the approach that is usually taken on a triple axis spectrometer. Here the easy parts of the experiment are done first and, exploiting the information already obtained, one proceeds successively to the harder parts of the experiment.

Our calculation for  $\text{ZrFe}_2$  have demonstrated that from nearly every detector a uniquely determined phonon can be extracted. In a further step their value for an understanding of the lattice dynamics of the system has to be investigated. This may be done by using these data to reconstruct the model from which we started.

The analysis has also shown that there is a great number of phonon groups, which cannot be separated from each other due to insufficient resolution. Quite clearly the amount of information that one can extract from such an experiment increases considerably if the resolution is improved. As a rule the resolution has to be better than for experiments on a triple axis spectrometer, where frequently interferences of neighbouring phonon branches can be avoided by a suitable measuring program.

Much thought has to be put into the problem, how the large amount of data, which can be obtained with a multi-detector, can be condensed into a form, which is suited for communication, without losing important information by the condensation process.

The applications of a direct geometry time-of-flight spectrometer for phonon measurements at the SNQ has to be seen in connection with other competing types of spectrometers. It was shown in a separate report, that for studies of selected phonons the triple axis spectrometer is still the adequate instru-

ment. In survey studies of the phonon dispersion curves inverted geometry time-of-flight spectrometers with multi-analyzers offer certain advantages, which are discussed in Ref. 3. Therefore we do not propose to build a direct geometry time-of-flight spectrometer with a multi-detector specifically for measurements of collective excitations in single crystals. An instrument of this type will certainly be installed at the SNQ for measurements of isotropic scattering laws. This instrument should be designed in such a way, that also single crystal work can be done. This means, that the instrument should be equipped with a computer controlled turning table or Eulerian cradle for the sample. Each detector must be recorded individually and a high flexibility concerning the choice of the primary energy and the resolution is required.

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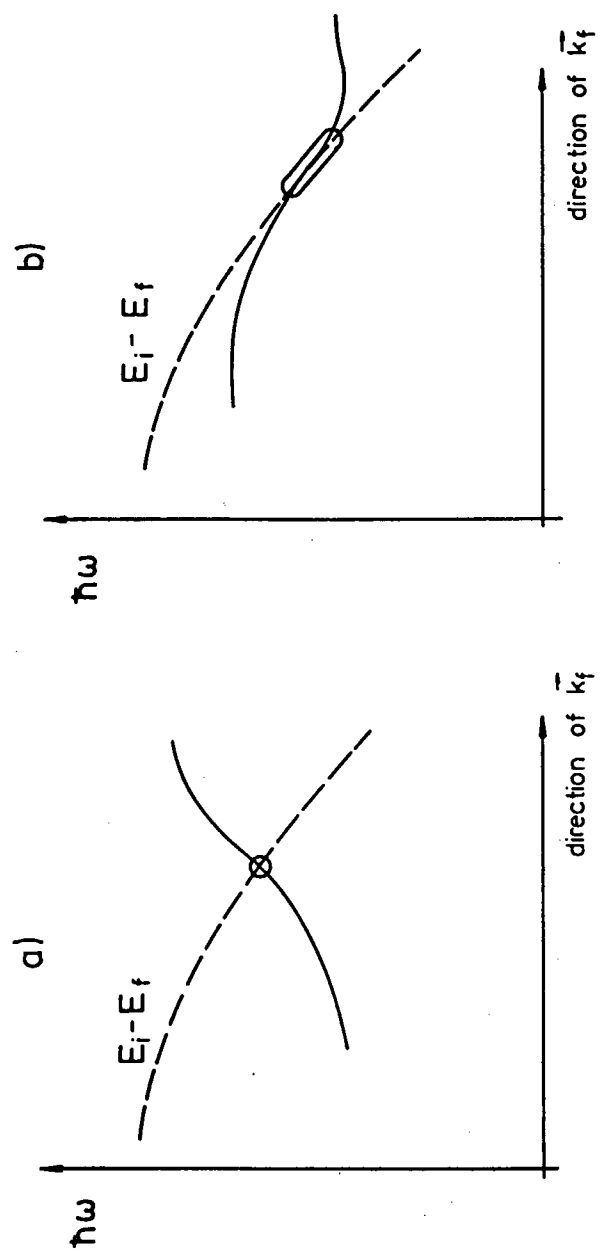


Fig. 2: Two cases for phonon measurements by time of flight  
a) : favorable situation  
b) : unfavorable situation

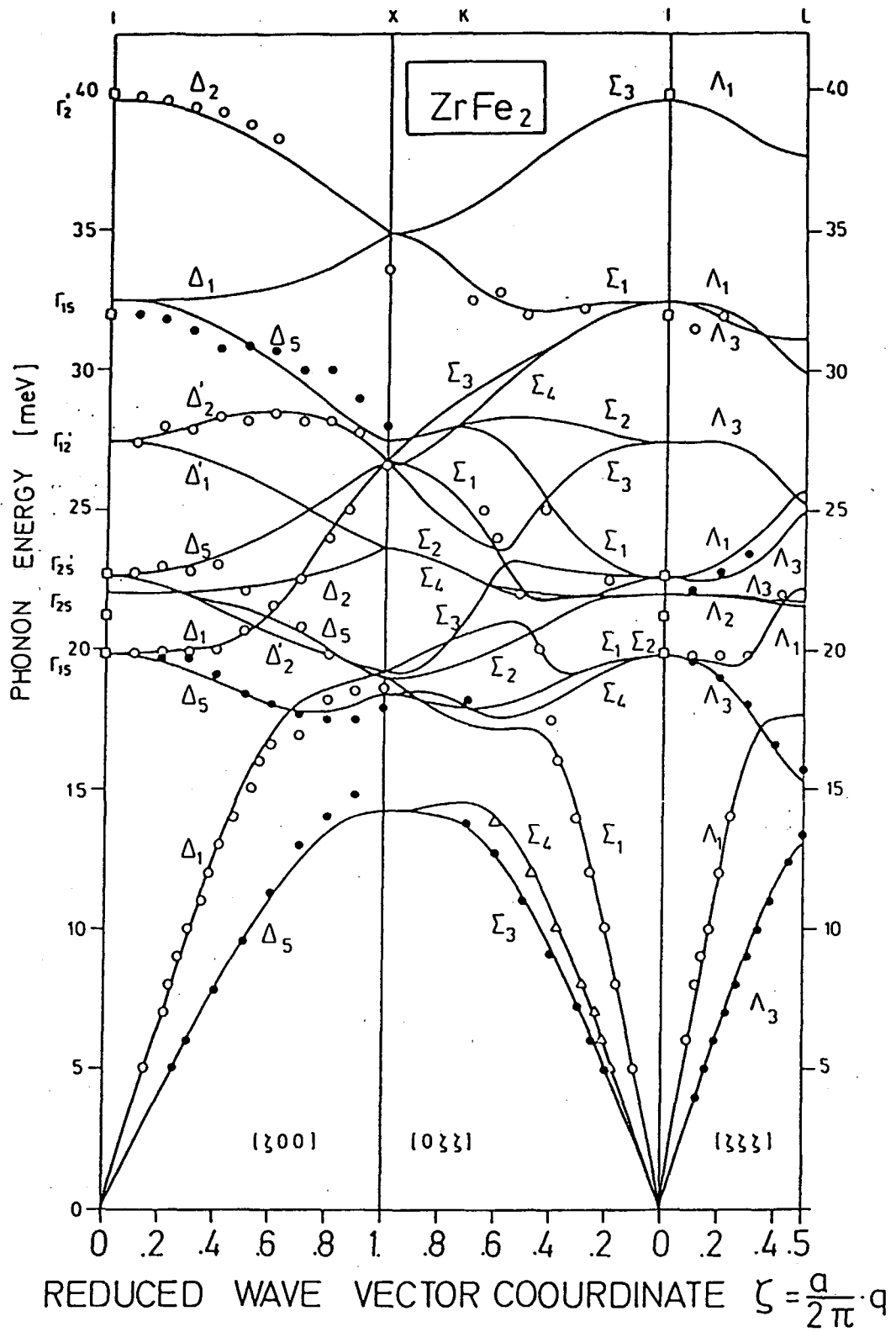


Fig. 3 Dispersion curves of  $\text{ZrFe}_2$  at 296 K

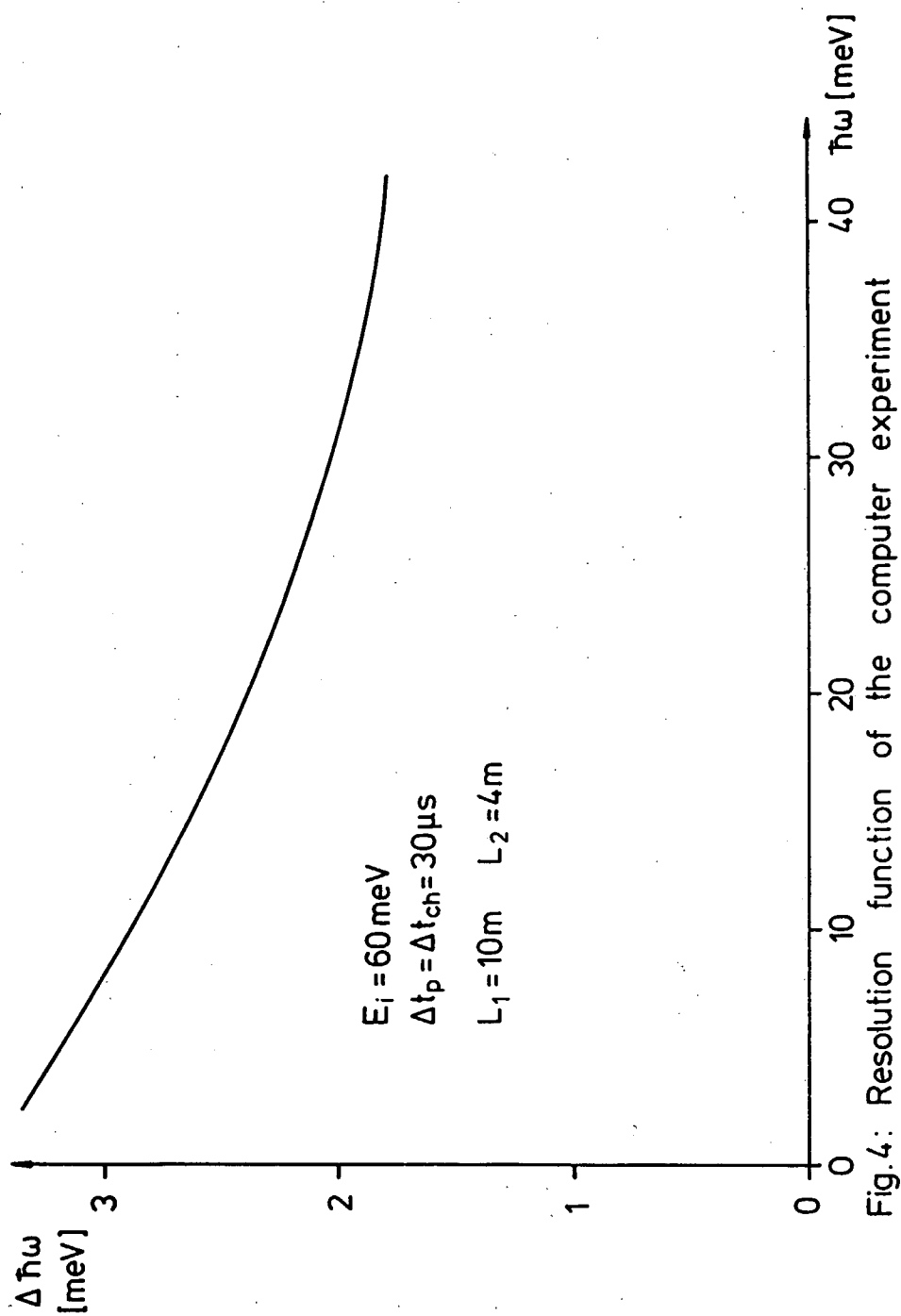


Fig.4: Resolution function of the computer experiment

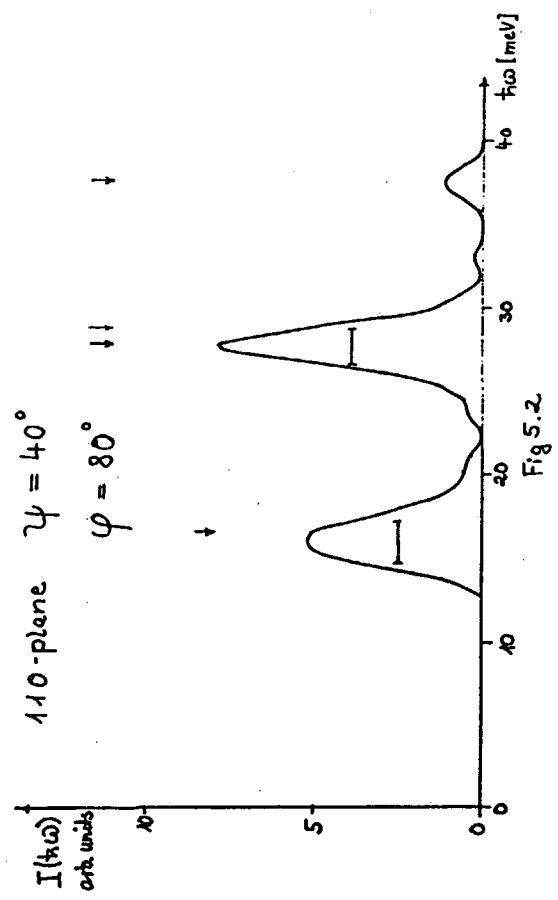
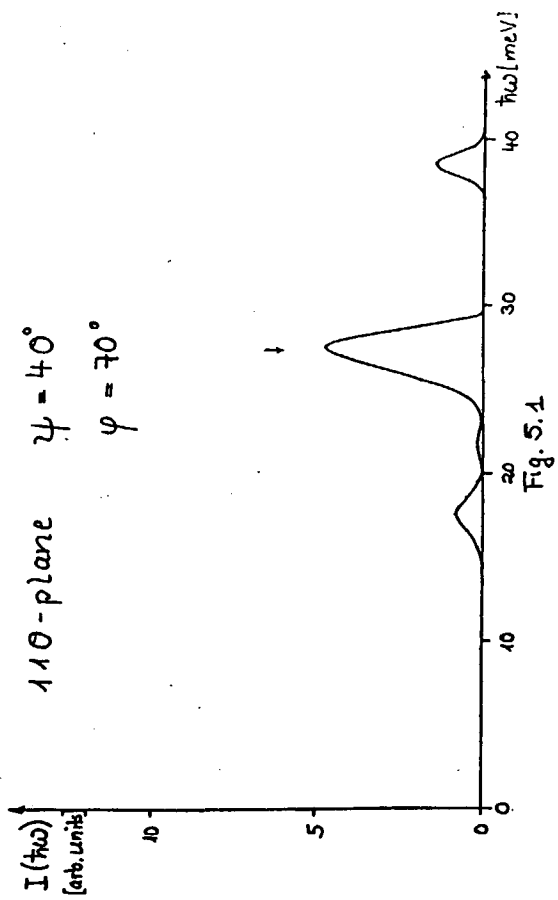
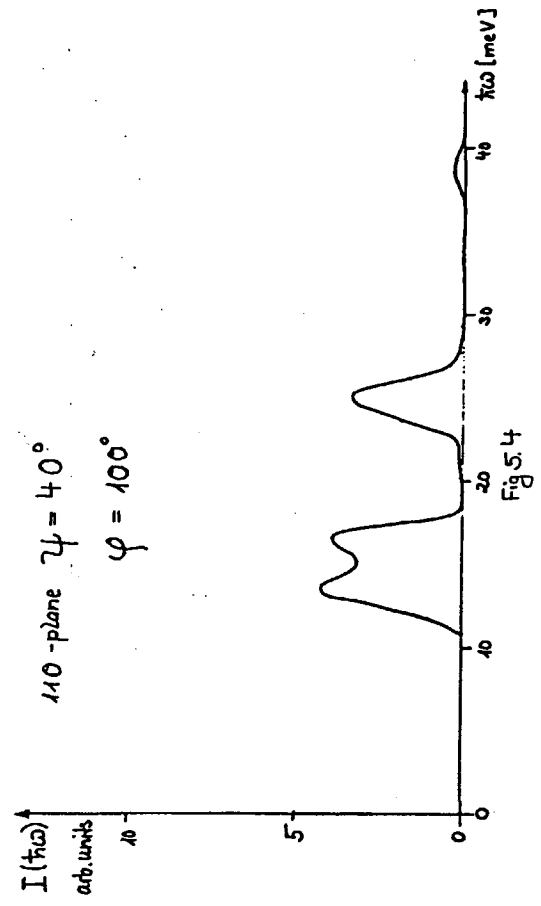
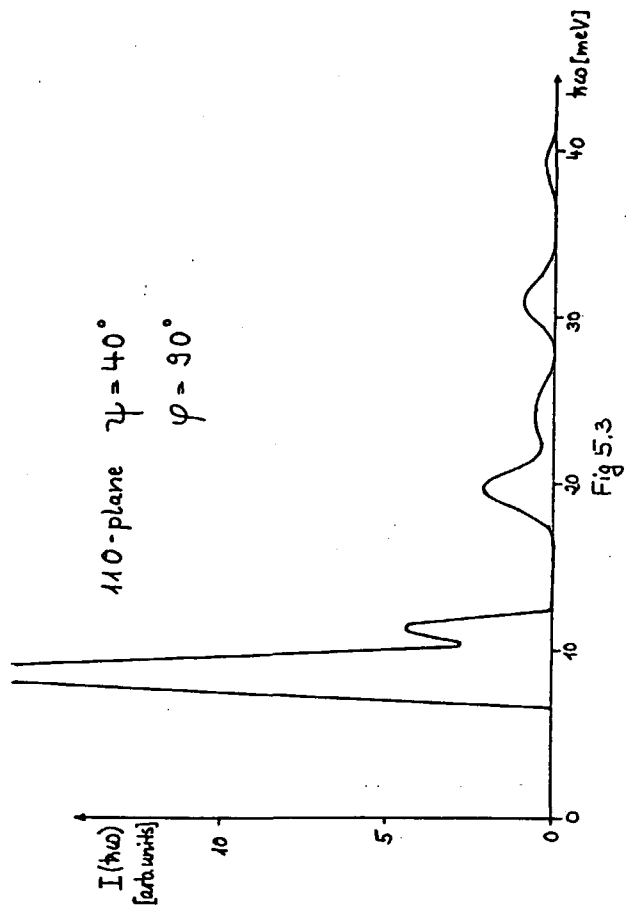


Fig. 5 Energy distributions as a function of the scattering angle ( $110$ -plane,  $\psi=40^\circ$ )



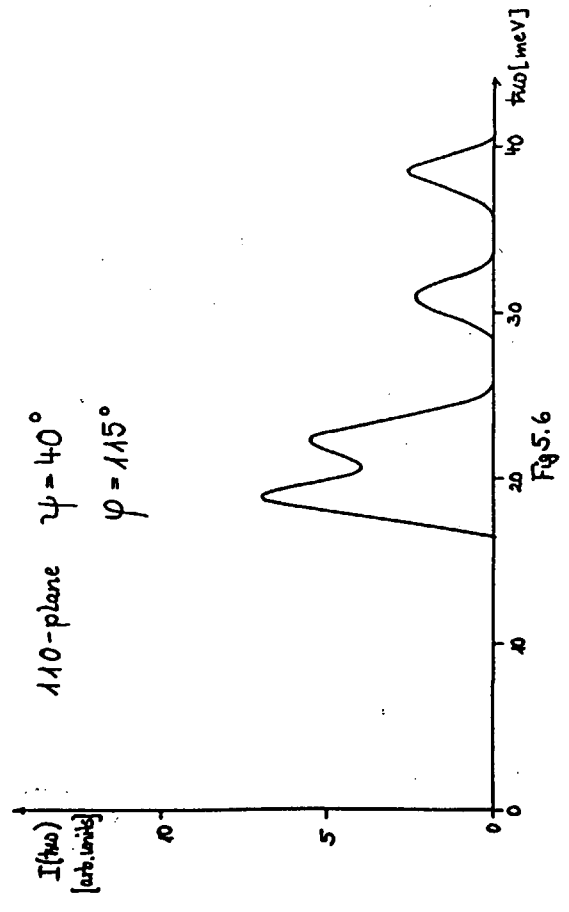
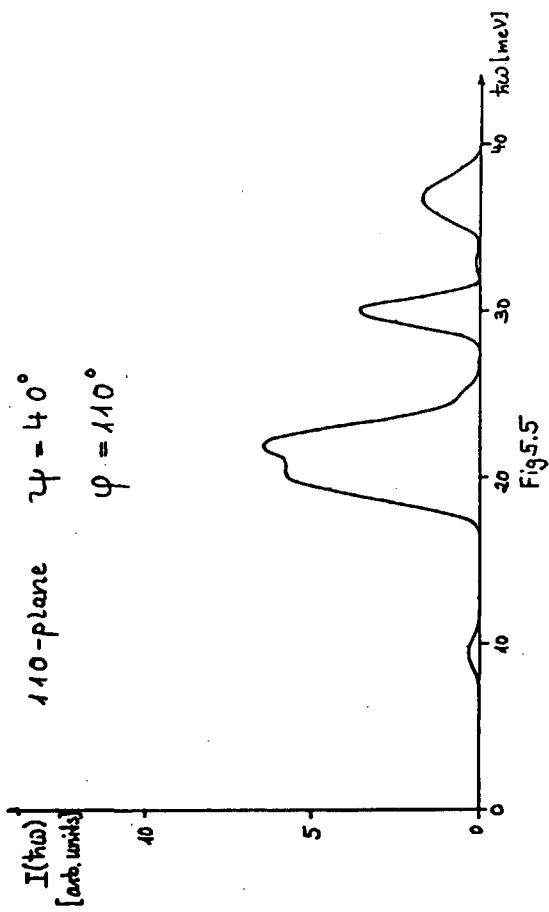
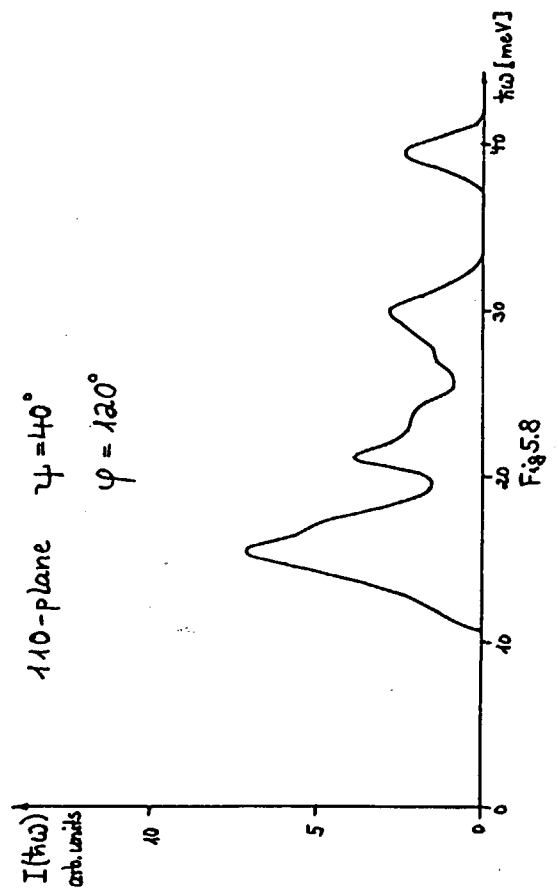
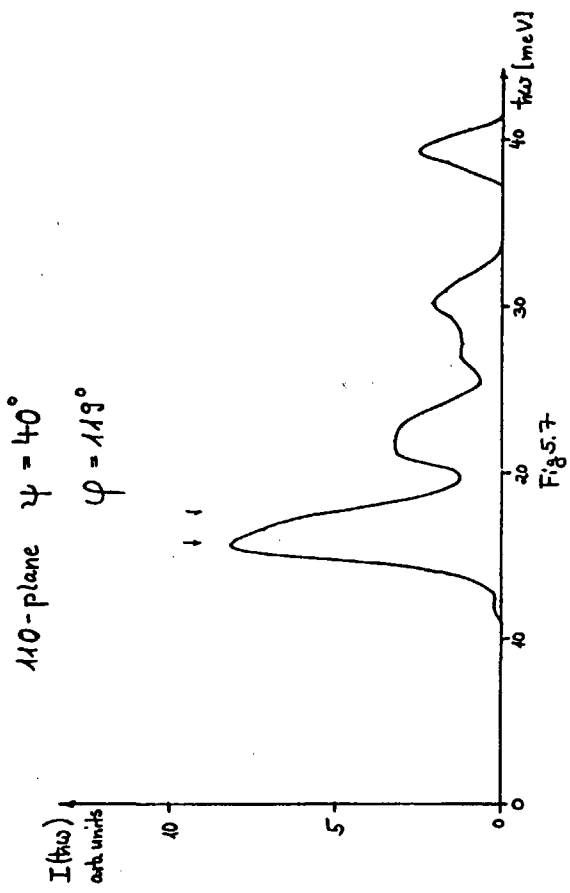


Fig. 5 continued

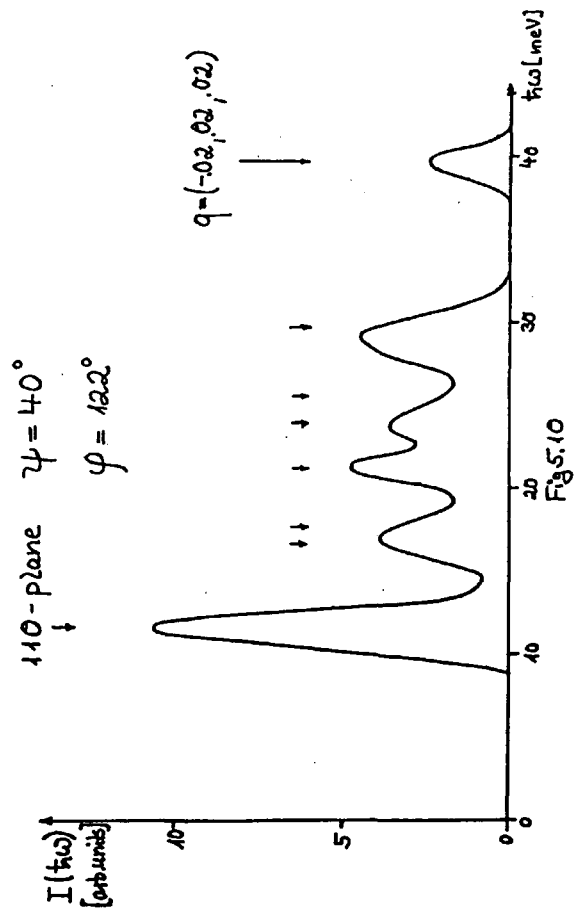
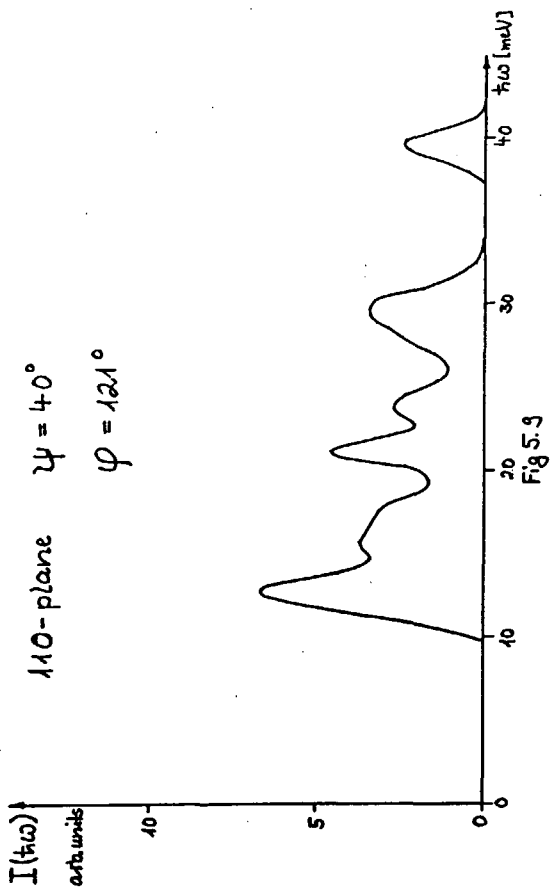
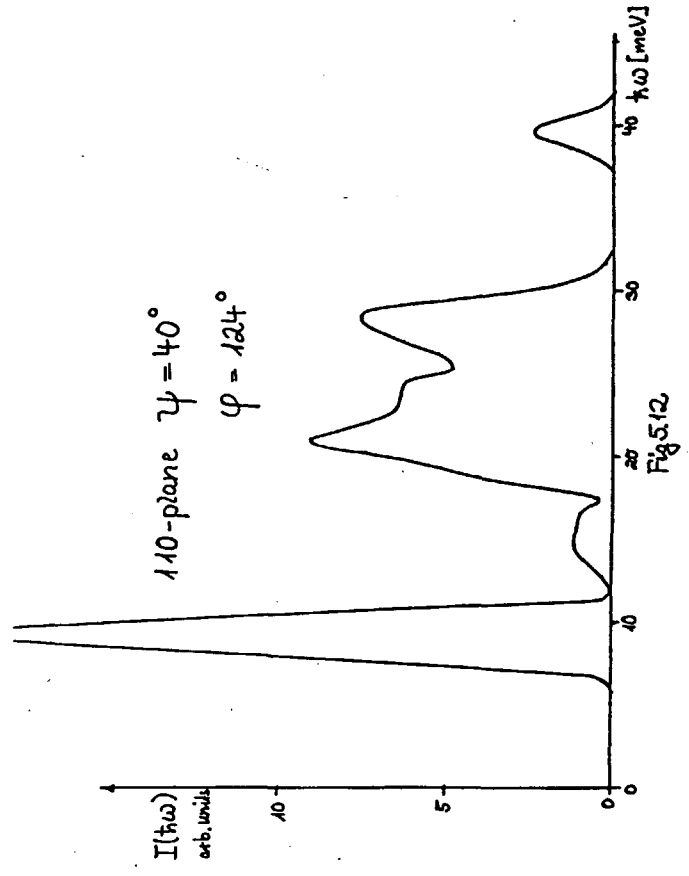
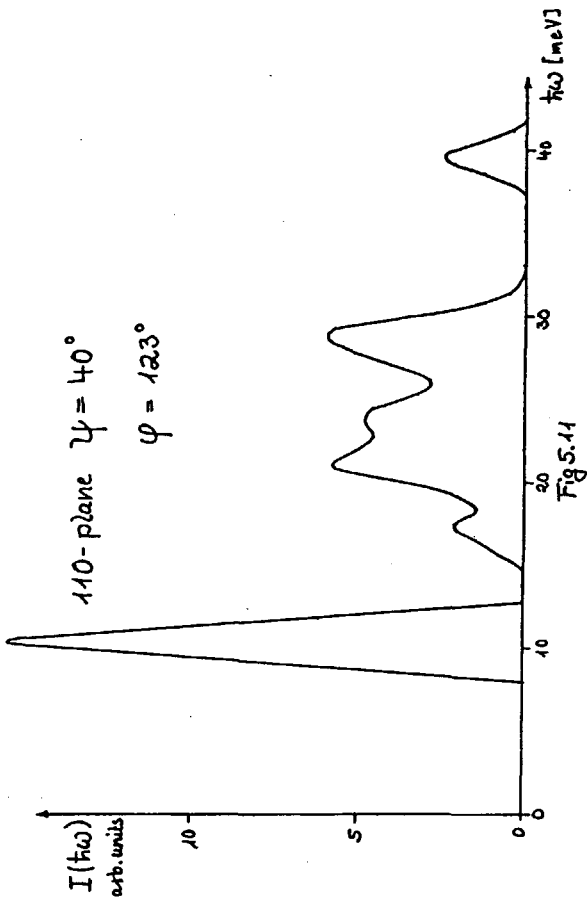


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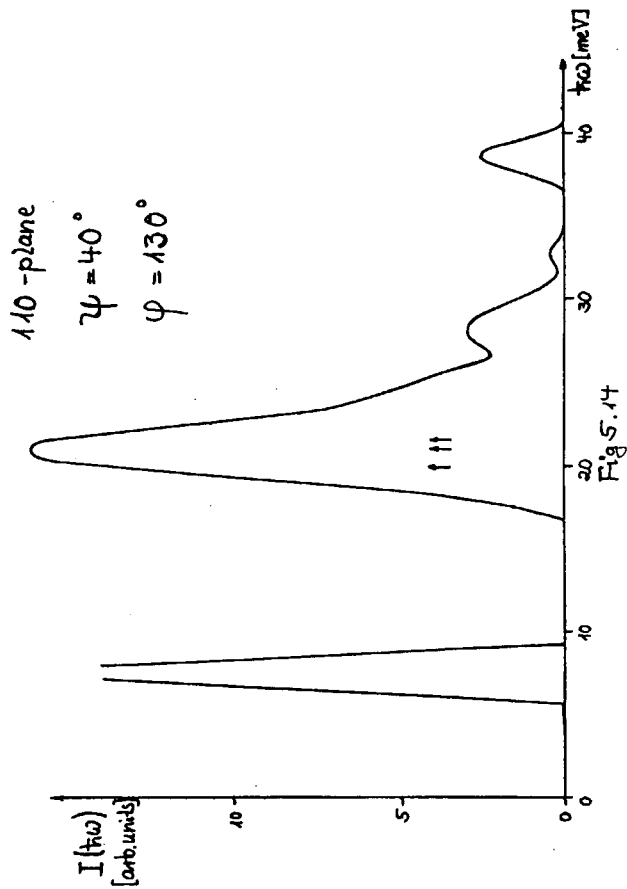
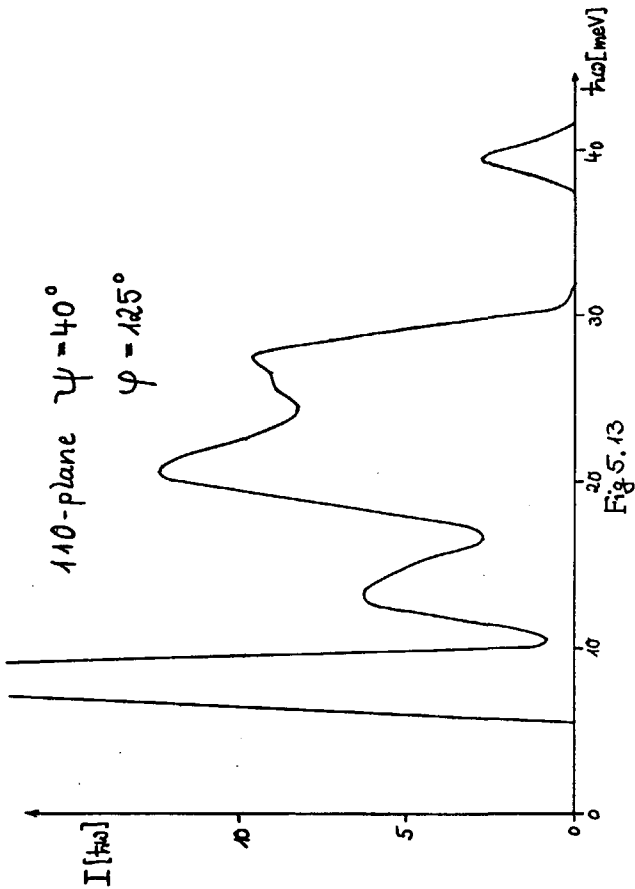


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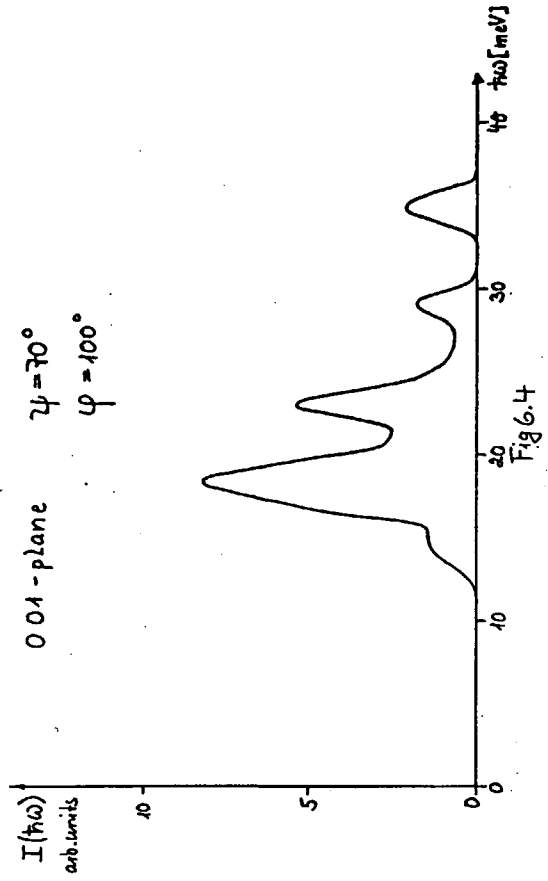
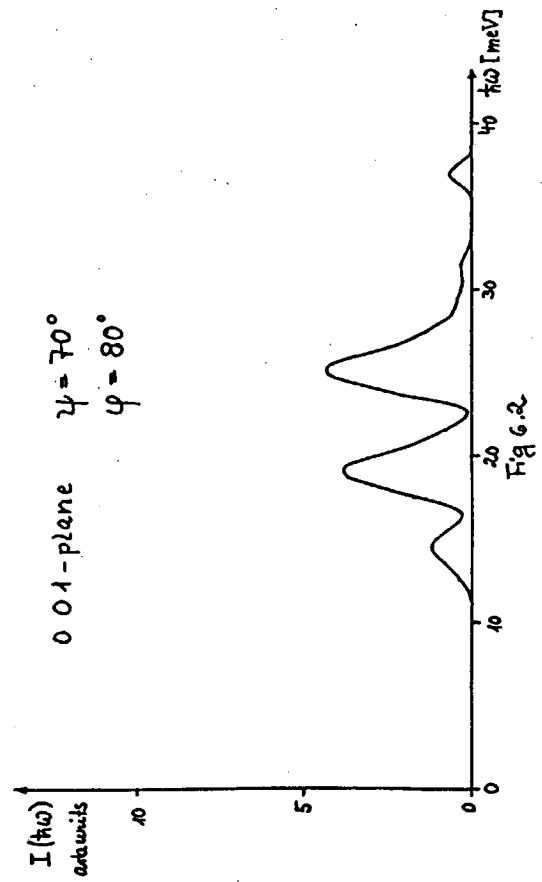
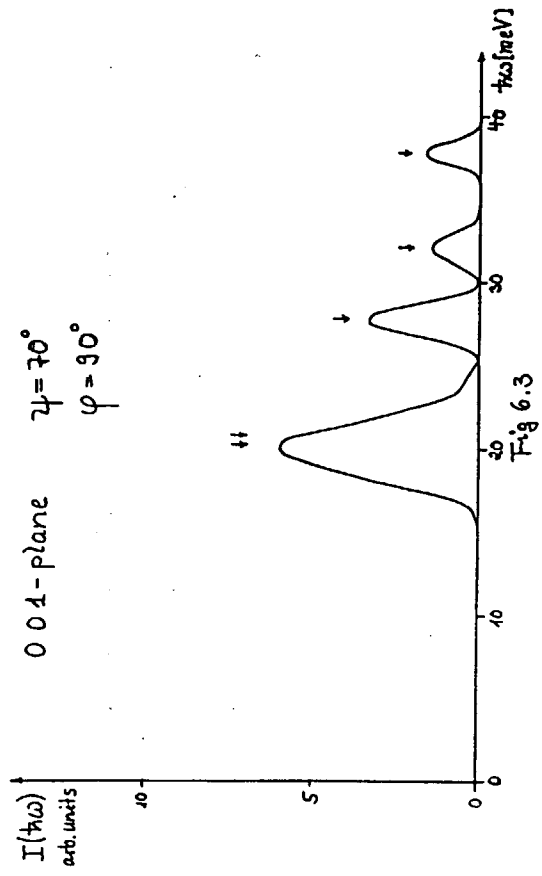
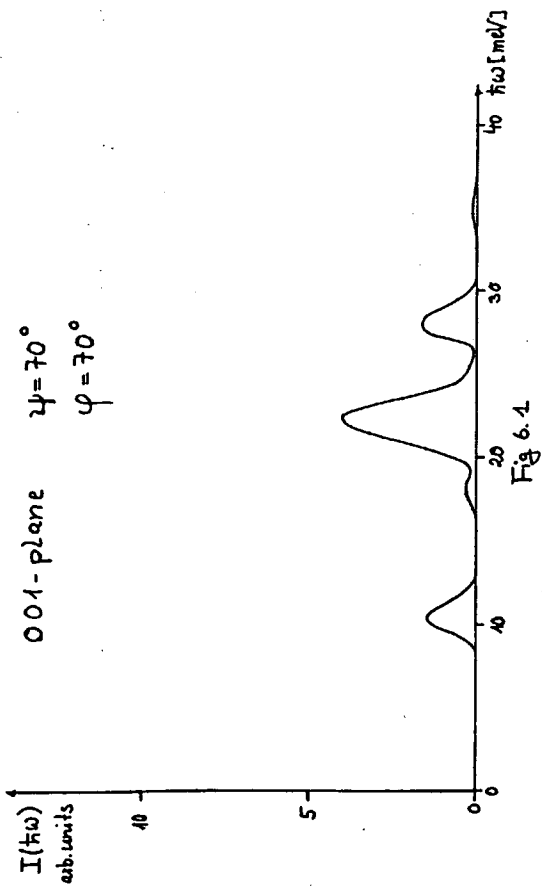


Fig. 6 Energy distributions as a function of scattering angle (001-plane,  $\psi=70^\circ$ )

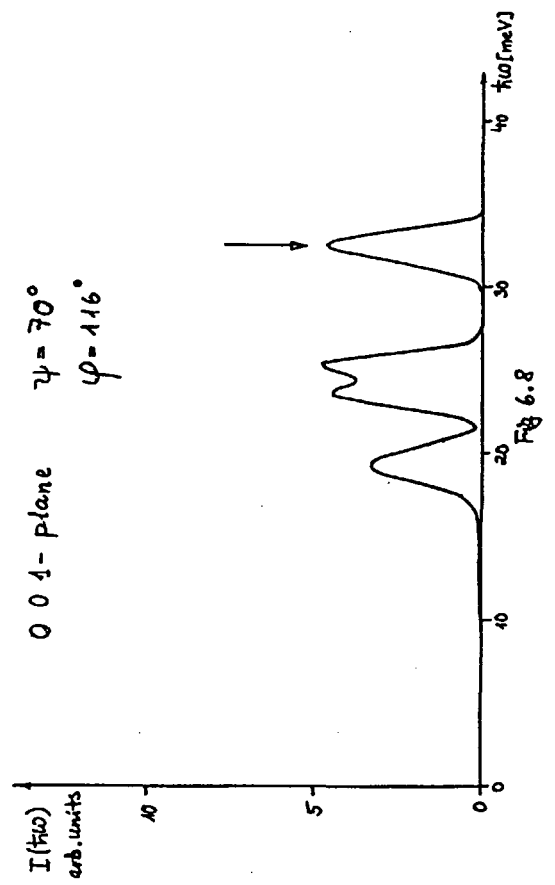
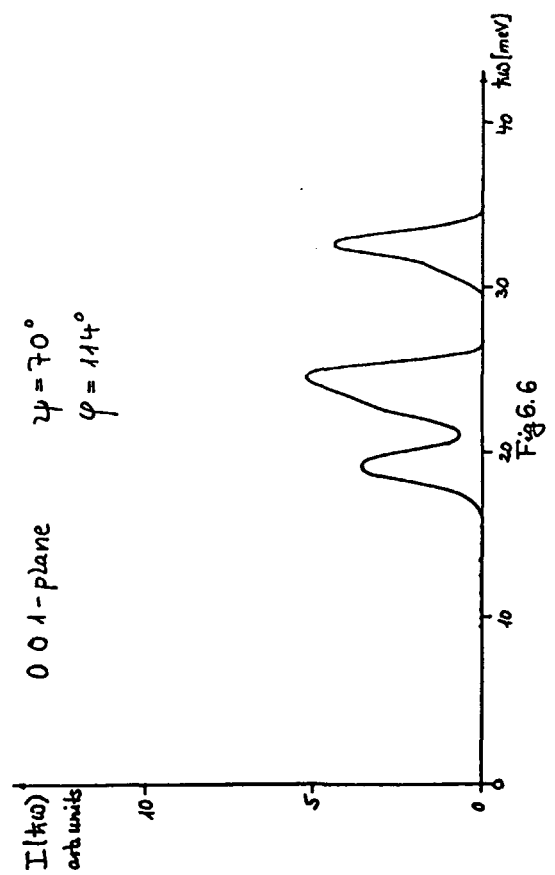
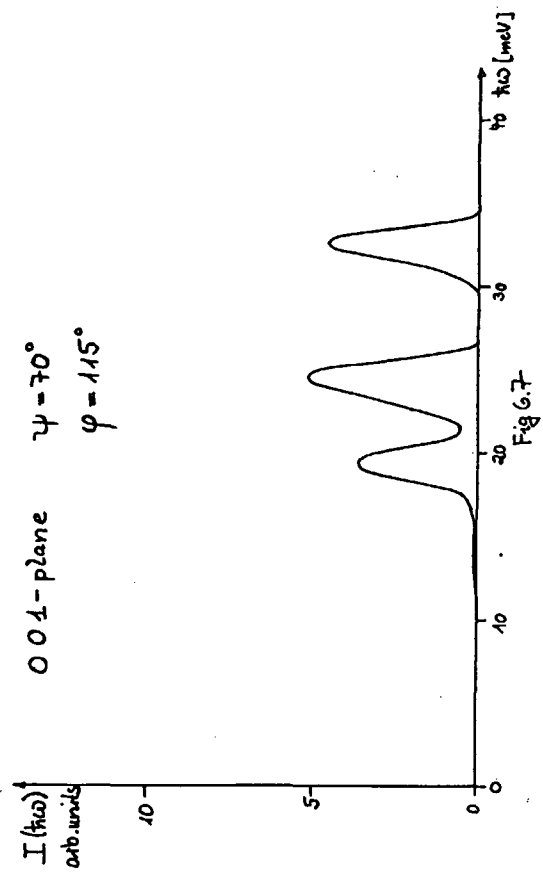
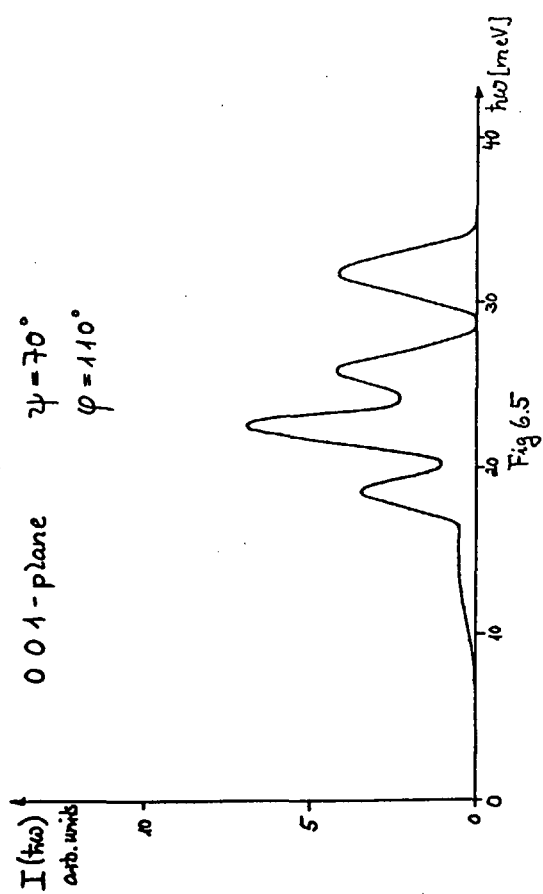


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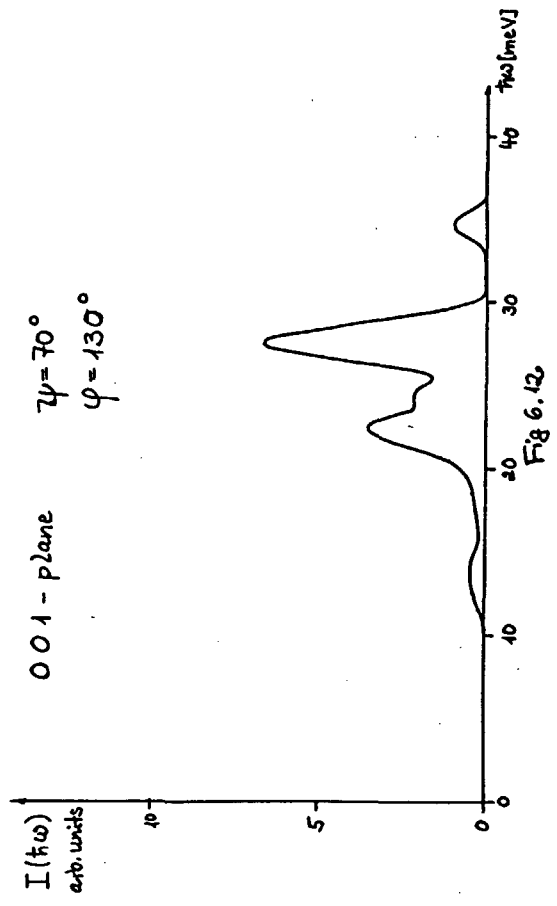
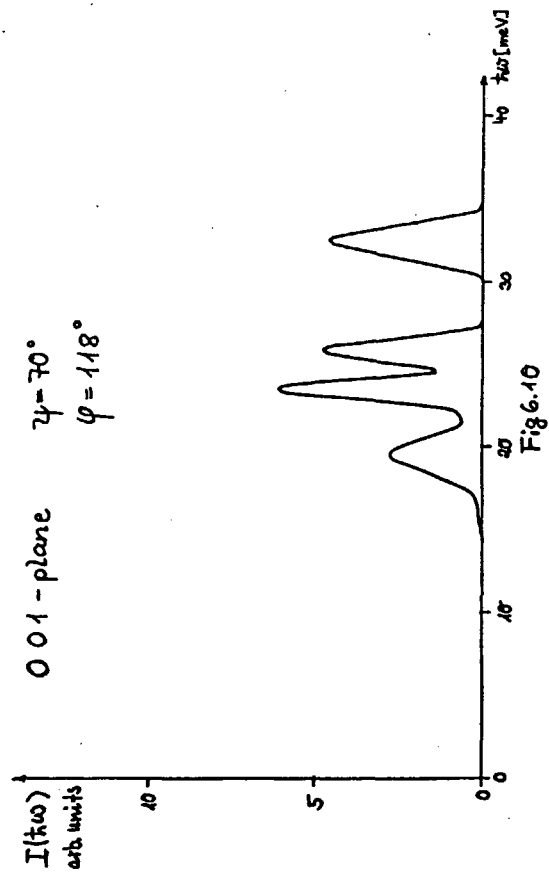
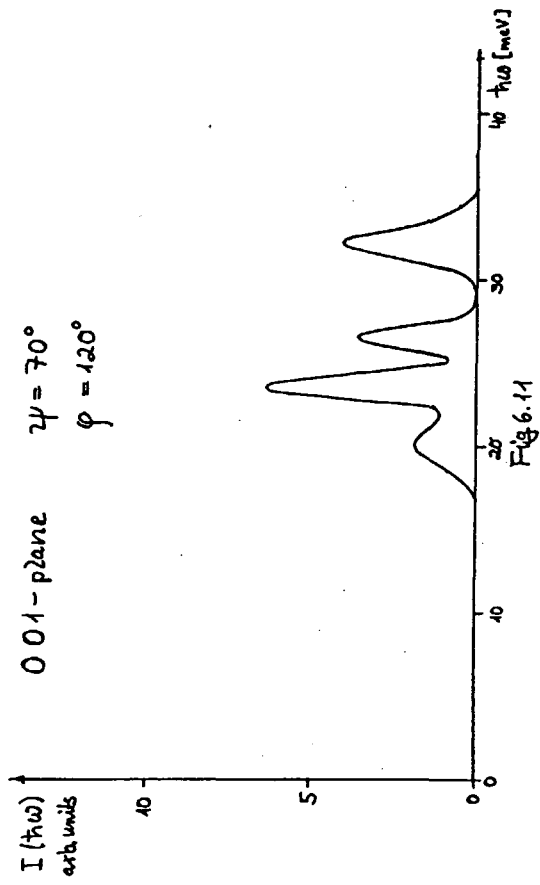
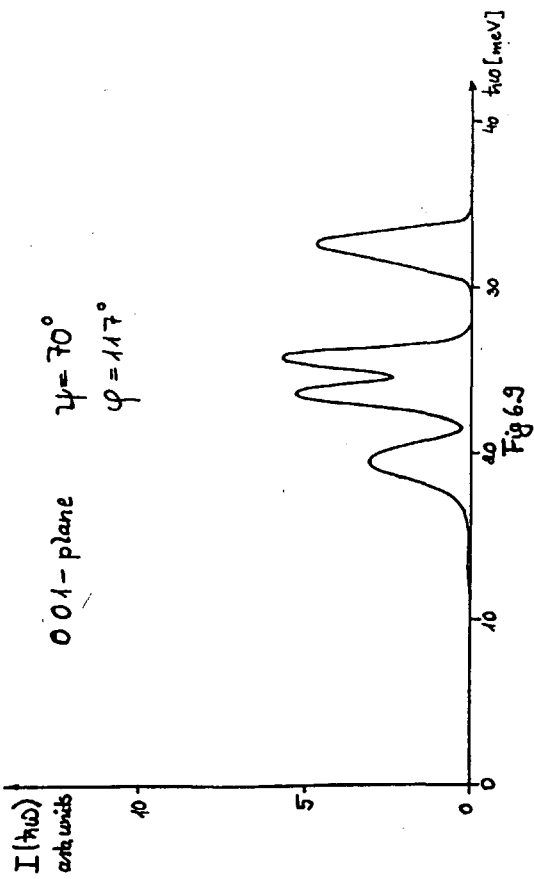


Fig. 6 continued

SOME CONSIDERATIONS CONCERNING THE MEASUREMENTS OF DYNAMIC  
STRUCTURE FACTORS AND FREQUENCY DISTRIBUTIONS OF ISOTROPIC  
SAMPLES AT THE SNQ

J.-B. SUCK, KFK<sup>x</sup>

M. LOEWENHAUPT, KFA

ABSTRACT

Measurements with thermal neutrons of dynamic structure factors  $S(Q, \omega)$  in a large region of  $Q$ - $\omega$  space for isotropic samples at the SNQ are considered. It is concluded that the large range of scientific problems in this field can be covered with three t-o-f spectrometers of different specifications.

The problems of t-o-f spectrometry at the SNQ are pointed out taking a chopper spectrometer as an example. Despite of the unpleasant long pulses and the fixed low repetition rate of the SNQ t-o-f spectrometers at this source still gain from the high peak flux compared with a reactor with the same mean flux.

A compressor ring (plus cooled moderator) improves drastically the situation of t-o-f spectrometers at the SNQ especially for higher incident energies. Further improvements could be gained from higher repetition rates in many cases and possibly from thinner moderators.

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<sup>x</sup>Present address : Institut Laue-Langevin, 156X,  
38042 Grenoble Cedex, France

## 1. INTRODUCTION

Due to lack of time we do not propose any specific instrument here. Instead, we shall mention a few general arguments which could be useful in the construction of instruments at the SNQ appropriate to cover the broad region of science (physics, chemistry and biology) concerned with the atomic and spin dynamics in isotropic samples (polycrystals, amorphous systems, liquids and gases). Our arguments are not based on any detailed knowledge about the SNQ or pulsed sources in general, but just on some experience we gained in investigations of isotropic samples at reactors and the IPNS. Our arguments are based on the science to be done and not on the knowledge of the techniques to be used<sup>x</sup>.

One of the main virtues of thermal neutrons is the fact that an energy transfer  $\hbar\omega$  in the meV-range is accompanied with a momentum transfer  $(\hbar)\underline{Q}$  in the  $\text{\AA}^{-1}$  range and that this momentum transfer vector can be chosen relative to the inner coordinate system of an "anisotropic" sample (single crystal, oriented magnet a.s.o.). This latter possibility is lost if isotropic samples are used. This very severe loss of detailed information about the atomic motions in the sample can partly be compensated by

- a) measurements of the dynamic structure factor in the whole region of relevant energy and momentum transfers

$$\hbar|\underline{Q}| = \hbar Q, \text{ and}$$

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<sup>x</sup>In the following we shall restrict our considerations to thermal neutrons (10 - 250 meV).



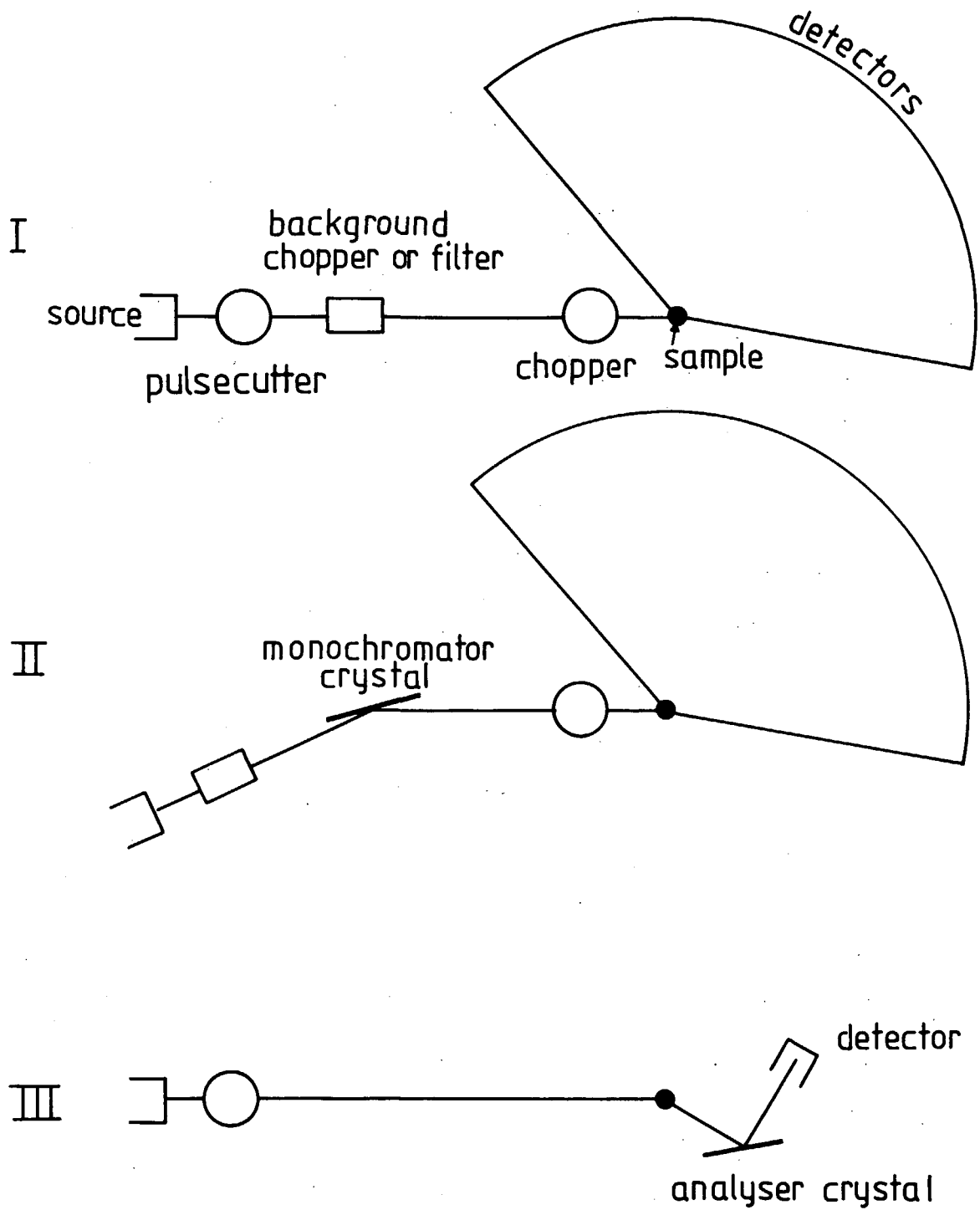
- b) measurements of absolute cross-sections including exact line shapes for comparison with model calculations from which then often a more detailed information about the atomic and spin dynamic can be extracted than from the experimental results alone.

Instruments suited for such experiments therefore have to have the following properties :

- 1) They must be very reliable, i.e. reproducibility has to be guaranteed on absolute scale.
- 2) All corrections to the measured data have to be as small as possible including those for the finite resolution of the spectrometer. A good resolution of the spectrometer is required even in the case of broad scattering distributions.
- 3) A very large region of Q- $\omega$ -space must be accessible with different incident energies.
- 4) They must be efficient but not necessarily "fancy" since they are used for the investigation of the atomic and spin dynamic in a very broad region of science.

A type of instrument which - if properly constructed - fulfils all these requirements are time-of-flight (t-o-f) instruments, because all instrumental parameters (except the time) are kept fixed during the experiment and the corrections can therefore be handled. Three basic types to which all real instruments can be related are shown in Fig. 1.

Types I and II are direct geometry spectrometers which work with a monochromatic incident neutron pulse i.e.  $\underline{k}_0$ ,

Figure 1

the mean wave vector of the incident neutrons is kept fixed during the experiment and the wave vector  $\underline{k}_1$  of the scattered neutrons is determined from their time-of-flight between the sample and the detectors, which are fixed on a large circle around the sample. The two types of t-o-f spectrometers shown in Fig. 1 (I and II) only differ in the way the monochromatic pulse is produced. In the rotor spectrometer (type I) a "white" neutron pulse is produced by some pulsing device (pulsed source or chopper) and one energy is selected by a second chopper from this energy band after it has passed a primary flight path. In the hybrid spectrometer this energy selection is done by Bragg reflection from a stationary (or rotating) single crystal (the monochromator), according to its d-spacing and the chosen Bragg angle.

Type III is a spectrometer with inverted geometry which works with an incident "white" neutron pulse which impinges on the sample. The scattered neutrons are analysed by a fixed single crystal (analyser) or filter so that only neutrons with a fixed wave vector  $\underline{k}_1$  can pass to the detector. The different incident wave vectors  $\underline{k}_0$  are determined by the t-o-f of the incident neutrons along the primary flight path between the pulsing device and the sample.

From these basic types more complicated t-o-f spectrometers can be deduced like correlation-chopper, multi-chopper, rotating-crystal spectrometer a.s.o.

Because of their favourable properties mentioned above t-o-f spectrometers have already been extensively used for measurements of dynamic structure factors and frequency

distributions of isotropic samples at many reactors. This was done despite of the fact that only 5 to 10 o/oo of the reactor neutrons can be used in this case in order to obtain the short neutron pulse necessary in thermal neutron t-o-f spectroscopy. It is therefore obvious that the same type of instrument should be used for similar types of investigations at pulsed sources too, where a much larger part (if not all) of the neutrons offered by the source can be used in t-o-f experiments.

## 2. SCIENTIFIC REQUIREMENTS

To get an impression of the resolution and intensity requirements of the spectrometers needed to cover adequately the scientific region of interest here, we have divided this region roughly into sub-sections with similar experimental conditions :

- 1) Measurements of the dynamic structure factor of polycrystals, amorphous systems, liquids and gases require good energy resolution ( $\Delta E_0/E_0 < 1-2 \%$ ) and reasonable Q-resolution ( $\Delta Q/Q < 3\%$ ).
- 2) The determination of frequency distributions and chemical, biological and magnetic spectroscopy require good energy resolution but can tolerate relaxed Q-resolution.

(We exclude here high resolution spectroscopy because it is dealt with in a separate contribution).

- 3) Intensity is the first requirement in experiments done with dilute samples, samples with short lifetimes (high temperature, high pressure, corrosion), in time

dependent measurements and if polarisation analysis is applied (separation of magnetic from nuclear scattering or of coherent from incoherent scattering). In most of these cases one will have to be content with an energy- and momentum-resolution of 5 to 15%. (The technique to be used for polarization analysis at the SNQ will not be mentioned here, as they are discussed in a separate contribution.

- 4) Brillouin scattering experiments require very high incident energies with very reduced background so that measurements at small scattering angles with very good energy - and reasonable Q-resolution can be done. We shall not follow this interesting subject here, since it was tackled in a separate contribution by Dr. Reichardt.

From the different requirements mentioned above it is clear that they cannot be met optimally by one single instrument. For economic reasons, however, as many scientific problems as possible have to be covered by each instrument without sacrificing optimization.

### 3. TIME-OF-FLIGHT EXPERIMENTS WITH THERMAL NEUTRONS AT THE SNQ

We shall shortly mention the advantages and disadvantages of all three basic types of t-o-f spectrometers shown in Fig. 1 and then discuss one of them with respect to the parameters of the SNQ.

The advantages of spectrometers with direct geometry is the large amount of  $Q$ - $\omega$  space accessible due to the large amount of counters surrounding the secondary flight path

with scattering angles between 0 and 130 or 140 degrees (see Fig. 9.1 in [1]). The rotor spectrometer (type I in Fig. 1) has the further advantage of its easy way to change the incident energy by a simple phase change of the choppers with respect to the source pulse with no further change of other instrumental parameters, irrespective of the detailed geometry of the secondary spectrometer (vertical or horizontal geometry). Its main disadvantage is the fact that the sample position is in the source beam causing serious background problems, which can only partly be overcome by additional background choppers or filters between the source and the sample.

Hybrid spectrometers (type II in Fig. 1) can only be regarded as convenient instruments if they can be used in horizontal geometry. In that case, the sample table and the secondary spectrometer (mounted on air-cushions) can follow the incident monochromatic beam which changes direction relative to the source beam according to the Bragg angle of the monochromator. If however the sample table has to be kept at a fixed position and no double monochromator is used for intensity reasons then the monochromator crystal has to be moved with respect to the sample as it is shown in Fig. 2. This leads to unwanted large distances between monochromator and sample at high incident energies (because a minimum amount of shielding is necessary for a  $90^\circ$  take-off angle). The primary resolution may change appreciably with incident energy and, if in addition the secondary

spectrometer is fixed, the zero scattering angle and with it the angular range of the secondary spectrometer will change with the incident energy, as it is shown in Fig. 2.

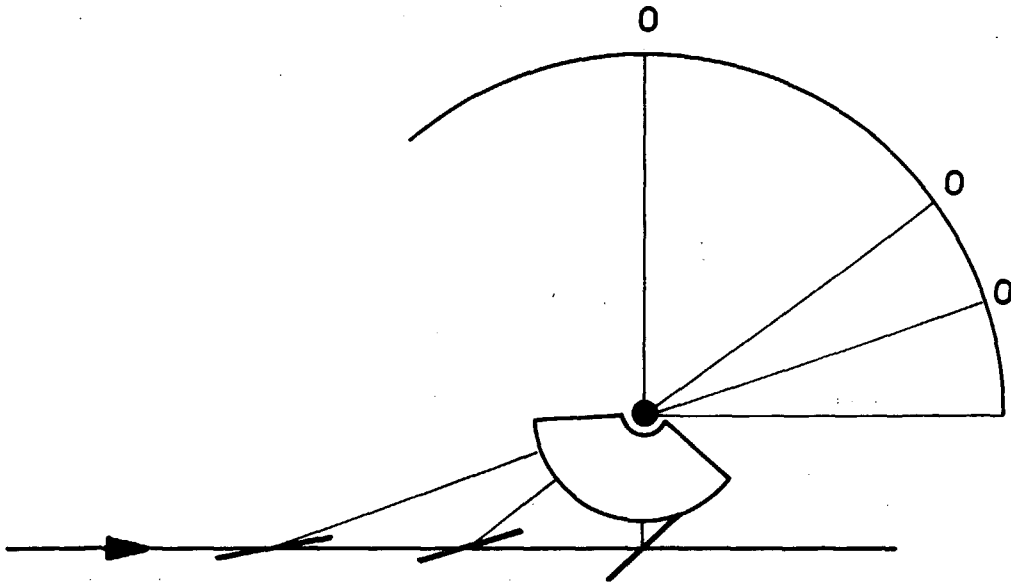


Figure 2

Supposing horizontal flexible geometry and the availability of monochromator crystals with high reflectivity (e.g. Be, PG, a.s.o.) the main advantages of a hybrid spectrometer are : separation of the sample from the source beam, Q- and also the  $\omega$ -resolution can be varied and focussing devices can be used with appreciable gain in intensity. The incident energy is independent of phase errors or aging of moving parts ! The main disadvantages are the small reflectivity of the monochromator crystals especially for neutrons with energies above 100 meV and the additional background from the monochromator due to TDS and the incoherent scattering. Higher order contaminations can be eliminated by background choppers well before the monochromators. In high resolution

configuration additional collimators will have to be used which will cost part of the intensity.

If, however, one accepts the intensity loss due to the second reflection from a double monochromator system (about a factor of 2 to 3), then one could even with a vertical scattering plane combine the advantages of a hybrid spectrometer at incident energies below say 70 meV, where the reflectivity of good monochromator crystals is not far below 0.5 with those of a chopper spectrometer at incident energies above 70 meV. This can be achieved by a secondary spectrometer which can be moved vertically between the two monochromatic beams coming from the choppers and the double monochromator respectively as it is shown in Fig. 3<sup>\*</sup>.

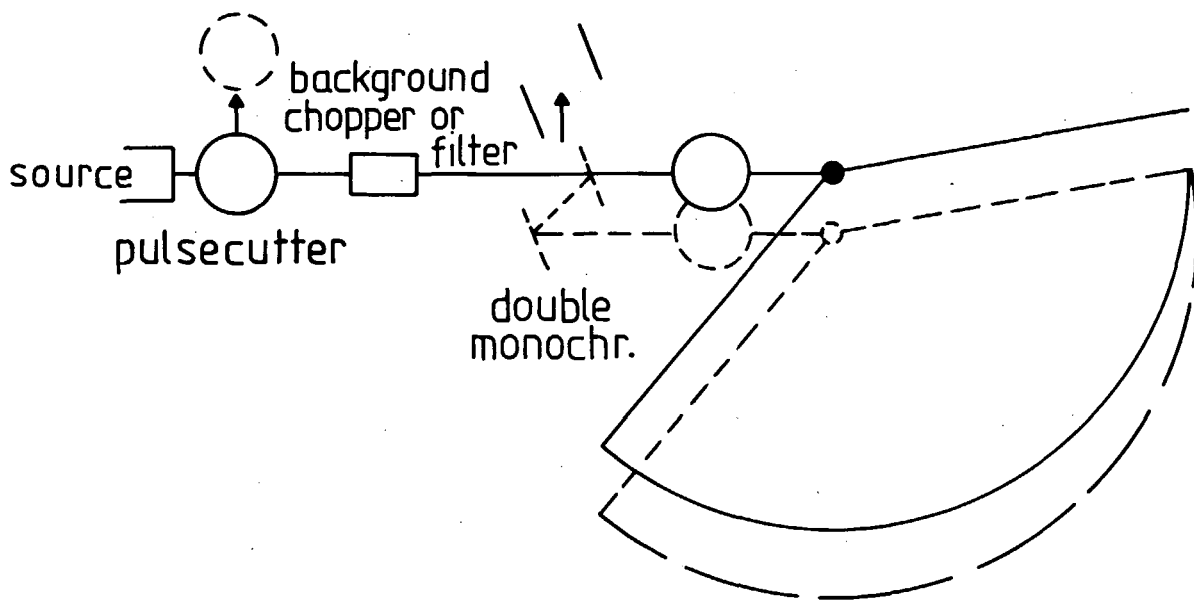


Figure 3

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<sup>\*</sup>This idea was pointed out to one of us (J.-B S) by  
Andrew Taylor



Spectrometers with inverted geometry (type III in Fig. 1) have the advantage of high neutron intensity at the sample, because of the "white" source beam impinging on the sample (broad energy band in reality due to the necessary pulse cutter near the moderator of the SNQ). The analysing methods are well suited : t-o-f analysis for the high incident energies over a long primary flight path, crystal or filter analyser for the slow scattered neutrons (high reflectivity, good resolution of the analyser crystal). The geometry of the spectrometer (long primary flight path, short sample-analyser-detector distances) give this spectrometer a rather convenient geometry. The disadvantages of this type of spectrometer are the high (in  $\omega$ ) but narrow (in  $Q$ ) accessible  $Q$ - $\omega$  range for small  $k_1$  (resolution arguments !) compared with spectrometers with direct geometry and the worse background conditions due to the sample position in the source beam.

Without even having done any quantitative comparison between these three basic types of t-o-f spectrometers, not regarding all possible combinations and additional devices we shall nearly arbitrarily choose the rotor spectrometer to discuss the relevant parameters of t-o-f experiments at the SNQ. This choice allows our discussion to be based on a minimum amount of assumptions, e.g. concerning the space available next to the sample position and for the secondary spectrometer.

A reliable and flexible t-o-f spectrometer has to meet the following requirements :

- 1) The secondary flight path has to be covered with efficient (neutrons with high energy have to be absorbed) fixed detectors in an angular range of  $-10$  to  $+130$  degrees, possibly in more than one row and with a circular detector arrangement at low scattering angles with a larger flight path than that of the detectors at larger scattering angles (see Fig. 4).

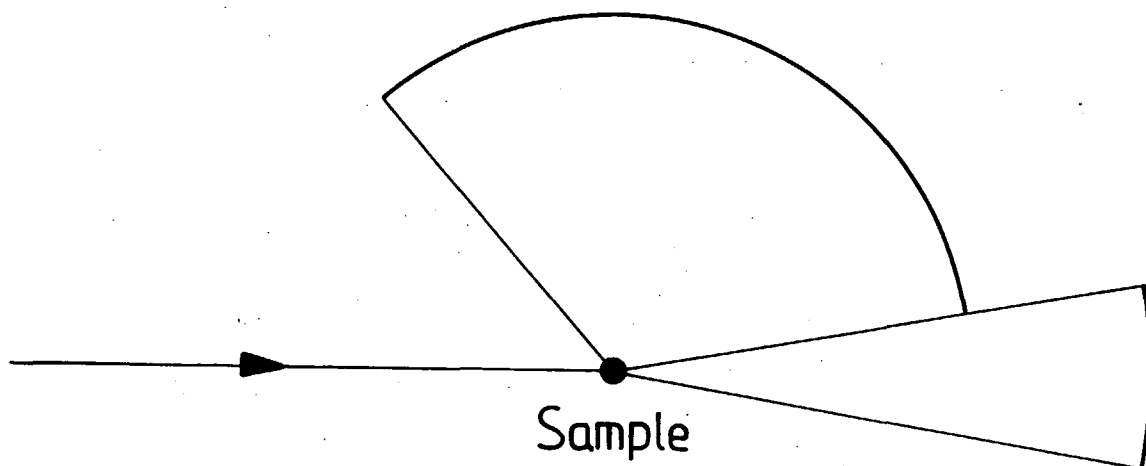


Figure 4

- 2) A sufficient amount of storage space has to be available at the instrument.
- 3) Heavy choppers have to be reliably spun at speeds at least up to 600 Hz (36000 rpm) with phase changes less than  $1 \mu\text{sec}$  with respect to each other and to the source pulse. This requires magnetic bearings throughout.
- 4) The detector electronics has to be very reliable.

In addition to these four basic requirements we shall assume :

- a) The sample is put as near to the moderator as possible for intensity reasons. Allowing for some additional

shielding for the detectors at 130 degrees this leads to a moderator - sample distance of 10 m.

- b) The distance between the two choppers should be at least 7.5 m (moderator-pulse cutter 1.1 m, second chopper-sample 1.4 m with about 1 m shielding between chopper and sample).
- c) We assume a secondary flight path of 4 m with sample and detector dimensions in the scattering plane  $< 2$  cm so that  $\Delta L/L_1 < 1\%$ . The time channel width can be chosen in a way that it is small compared to all other time uncertainties. For a spectrometer with worse resolution the flight path can be shorter.

At a t-o-f spectrometer with these parameters spectra can be recorded for incident energies above 20 meV, if neutron energy loss spectra are measured down to  $E_1 = 0.3 E_0$ , as it is shown in Table 1. For lower incident energies part of the spectrum would be lost in the gating time of the next source pulse. Consequently one will have to consider a second spectrometer with different geometrical parameters for measurements with incident energies between 5 and 20 meV.

Since we want the overall energy resolution of this spectrometer to be between 1 and 2% the 300  $\mu$ sec source pulse must be cut down to some reasonable value using a chopper next to the moderator (pulse cutter). As we require time uncertainties at the sample in a distance of approximately 9 m from the chopper of 5  $\%$  we arrive at the pulse lengths  $\Delta t_0$  given in Table 1.

The values given in Table 1 can be realised with Fermi choppers with a collimation of 1.3 degrees and larger, if the choppers are spun at 600 Hz. Optimisation for different regions of incident energies can be achieved by a set of appropriate choppers mounted on an automatic chopper changer and by different chopper speeds  $V = n.f_s$  with  $n$  integer and  $f_s$  the source frequency.

The angle of divergence of the choppers has to be large compared to the acceptance angle of the sample in order to avoid spoiling of the pulse width by the "light house effect" (see Fig. 4.19 in [1]).

Because the pulse cutter has a certain distance from the moderator its effect is that of a band selector. As the mean energy of this band can be chosen by an appropriate phase of the pulse cutter relative to the source pulse, this is a limiting factor only for spectrometers with inverted geometry where the spectrum of incident energies should be as broad as possible.

In addition to these two choppers, the pulse cutter near the moderator and the energy selector near the sample, further massive chopper will be needed between them to reduce the background of the sample. This is especially necessary for measurements at small scattering angles, which are most important in many of the investigations mentioned in section 2. Arguments for experiments at low scattering angles include : magnetic form factors, less multiphonons, pronounced coherent effects, diffusion a.s.o.

A special problem arises due to the large space necessary for the secondary spectrometer. If a horizontal scattering plane can be used (which would strengthen the arguments for a hybrid spectrometer) this would be highly preferable, because of the simpler sample environment and the easier access to the detectors. If however a vertical scattering plane has to be used, preferably below the source beam, then extreme sample conditions like very high and very low temperature can hardly be realized. The flight path length is anyhow limited by the present high cost of the detectors, which in turn limits the sample and the detector dimensions in the scattering plane.

The type of spectrometer described so far is the easy one at the SNQ. It has an energy resolution ( $\Delta E_0/E_0$ ) of 1 to 2 % and a Q-resolution of 5 % and better. For experiments, which can be done with relaxed Q-resolution (point 2 in section 2), this can easily be realized in the secondary spectrometer by adding up detector spectra. However, in that case the primary and secondary resolution are mismatched and intensity is sacrificed. In a hybrid spectrometer one could possibly overcome this problem by adding further monochromators in the scattering plane to the central one with appreciable increase in intensity. In that case one could cover the scientific problems mentioned in points 1 and 2 in section 2 with only one spectrometer for incident energies between 20 and 250 meV and with a second t-o-f spectrometer (of similar type) for incident energies below 20 meV.

A more difficult problem poses the third scientific point in section 2. Here a spectrometer is required with much intensity and relaxed momentum- and energy resolution. This problem becomes even more difficult if we include the interesting option of polarisation analysis !

#### 4. THERMAL NEUTRON TIME-OF-FLIGHT SPECTROMETER AT THE SNQ WITH COMPRESSOR RING

As can be seen from Table 1 for t-o-f spectrometers with reasonable resolution the source pulse of about 300  $\mu$ sec length has to be shortened, so that only a small part of the available neutrons are used (2 % at  $E_0 = 250$  meV). This situation is drastically improved if the source delivers a shorter and therefore intenser pulse as it is the case for the SNQ with a compressor ring and cooled moderator. For incident energies below 60 or 80 meV a chopper will still be needed, however the shorter pulse already allows to use 40 to 50 % of the entire pulse intensity, if the values given for pulse lengths for the SNS apply to the SNQ as well [2]. For higher incident energies the natural pulse length of the SNQ with compressor ring are short enough so that no pulse cutter is needed and all the neutrons delivered by the source can be used. For neutrons with 250 meV this means that one can gain a factor of 50 in intensity due to the compressor ring !

## 5. COMPARISON WITH TIME-OF-FLIGHT SPECTROMETERS AT REACTORS OF THE SAME MEAN FLUX

T-o-f spectrometers use the peak flux at pulsed sources and the integral intensity of these pulses if these are short enough. The disadvantage of pulsed sources in comparison with reactors is the fixed repetition rate and the thicker shielding. T-o-f experiments with thermal neutrons - and also with cold neutrons as long as the spectrum is concentrated near the quasielastic region and no appreciable frame overlap occurs - allow repetition rates which are higher by a factor of 2 to 3 than the  $f_S = 100$  Hz offered by the SNQ. At reactors the repetition rate is chosen in accordance with experimental requirements. This is not possible at a pulsed source and has therefore to be taken into account in the comparison with t-o-f spectrometers at reactors. Neglecting the differences in sample-source distances we conclude that we gain in intensity with t-o-f experiments at the SNQ in comparison with reactors of the same mean flux and a spectrometer with the same primary and secondary resolution by a factor of

$$G = \frac{\text{Peak flux}}{\text{Mean flux}} \cdot \frac{f_S}{f_R} = 40.0.4 = 16$$

without compressor ring ( $f_R$  = repetition rate of the chopper or rotating crystal at a reactor). This factor has to be multiplied by the gain factors given in Table 1 if a compressor ring is available.

This comparison does not include any considerations concerning the difference in background conditions at a pulsed source and a reactor which are difficult to assess.

Reference

- [1] C.G. Windsor, "Pulsed Neutron Scattering", Taylor & Francis, London 1981
- [2] "Blue Book", SNS



TABLE 1

| $E_0$ | $v_0$ | $t_0^*$ | $E_1$ | $v_1$ | $t_1^{**}$ | $t_0+t_1$ | $\Delta t_0^+$ | $I_0/I_s^{\S}$ | $I_0/I_s^{\S\S}$ | $GF^{++}$ |
|-------|-------|---------|-------|-------|------------|-----------|----------------|----------------|------------------|-----------|
| meV   | m/sec | msec    | meV   | m/sec | msec       | msec      | $\mu$ sec      | %              | %                |           |
| 5     | 980   | 9.2     | 1.5   | 535   | 7.5        | 16.7      | 46             | 15             | 40               | 2.7       |
| 10    | 1380  | 6.5     | 3     | 760   | 5.3        | 11.8      | 33             | 11             | 40               | 3.6       |
| 20    | 1955  | 4.6     | 6     | 1070  | 3.7        | 8.3       | 23             | 7.7            | 50               | 6.5       |
| 50    | 3090  | 2.9     | 15    | 1690  | 2.4        | 5.8       | 15             | 5              | 50               | 10        |
| 100   | 4370  | 2.1     | 30    | 2390  | 1.7        | 3.8       | 10             | 3.3            | 100              | 30        |
| 150   | 5350  | 1.7     | 45    | 2930  | 1.4        | 3.1       | 8              | 2.7            | 100              | 37        |
| 250   | 6910  | 1.3     | 75    | 3785  | 1.1        | 2.4       | 6              | 2              | 100              | 50        |

\* Flight time of neutrons with  $E_0$  over 9m

\*\* " " " " "  $E_1$  " 4m

+ puls length for primary time resolution of 0.5%

$\S$  ratio of used puls intensity  $I_0$  to total puls intensity  $I_s$  without compressor ring

$\S\S$  same as  $\S$ , but with compressor ring

++ gain factor with compressor ring with cooled moderator, based on SNS information

$E_0, E_1$  energy of neutron before and after scattering

$v_0, v_1$  velocity of neutron before and after scattering

DESIGN PROPOSAL FOR A MATERIAL SCIENCE SPECTROMETER  
AT THE SNQ-JÜLICH, F.R.G.

by

Walter Kley

Department of Physical and Natural Sciences  
Commission of the European Communities  
Joint Research Centre - Ispra Establishment  
21020 Ispra (Va) - Italy

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## ABSTRACT

After three decades of intensive research and development the advent of fusion power is imminent. However, the technical and economic success of fusion power reactors will depend on the endurance and availability of materials suitable for the radiation environment of a fusion reactor. In this context, the construction of a Material Science Spectrometer is suggested that is capable to measure simultaneously the density of point defects, the density and size distribution of precipitates, damage cascades and voids, the texture, the mobility of hydrogen as well as the microchemical evolution of candidate materials irradiated to end of life conditions. Therefore, it is suggested to construct a universal spectrometer that measures simultaneously Small Angle Scattering (SAS), Elastic Diffuse Scattering (EDS), Quasi Elastic Scattering (QES) and Bragg Scattering (BS).

## 1. INTRODUCTION

After three decades of intensive research and development, the plasma physicists may realize Lawson's break-even condition in their large Tokamak fusion test facilities, such as JET, towards the second half of the 80ies. This situation indicates that the design and construction of future fusion power reactors may be considered now more realistically. However, the technical and economic success of fusion power reactors will depend on the endurance and availability of materials suitable for the radiation environment of a fusion reactor. For the evaluation of the technical and economic feasibility, predictions are needed concerning the evolution of the mechanical properties of materials being exposed to complex radiation fields of a future fusion reactor.

It is now the general understanding of the fusion materials research community that the end-of-life of the first wall or blanket materials is determined by a considerable number of complex and competing phenomena. Due to non-linear effects in the evolution towards end-of-life conditions and due to the absence of a synergetic theory how damage energy is being stored in a material, one cannot extrapolate from low dose irradiations to end-of-life conditions. Therefore, we need neutron irradiations up to the end-of-life that may occur between 20 and 100 dpa (displacement per atom) or at the respective fluences between  $10^{22}$  and  $10^{23}$  n/cm<sup>2</sup>. Fluence = neutron flux . time:  $\varphi = \phi \cdot t$  (n/cm<sup>2</sup>). Since one year has about  $\pi \cdot 10^7$  sec, we need at least a fast neutron flux of  $10^{15}$  n/cm<sup>2</sup>.sec in order to reach in one year irradiation time end-of-life conditions for a candidate material.

It can be shown that only accelerator based neutron sources can provide the necessary strength with the required neutron energy distribution. There are essentially only two competing nuclear reactions that might be used for the production of an intense neutron source: they are based either on the D<sup>+</sup>-Li-stripping, or on the spallation reaction. Recently, it has been demonstrated that not only the D<sup>+</sup>-Li stripping reaction but also the spallation neutrons can produce a helium to dpa ratio of

13 appm He/dpa while for fusion reactor first wall materials this value ranges from 10 to 20 depending on blanket design conditions. Due to this excellent agreement the use of an intense spallation neutron source as a materials test and development facility has to be considered definitely.

Materials irradiated in an intense neutron source up to end-of-life conditions are very precious items and pulsed neutron sources such as the SNS at the Rutherford Appleton Laboratory, the WNR-Facility at Los Alamos and the SNQ at Jülich, offer a unique possibility for rather complete, non-destructive investigations of highly radioactive materials and fuel pins concerning their density of point defects, the density and size distribution of precipitates, damage cascades and voids, the texture, the mobility of hydrogen (induced by (n,p)-reactions) and possibly of point defects as well as the microchemical evolution in general. These parameters determine to a great extent the creep, swelling, embrittlement, fatigue, annealing, and in general the mechanical properties of highly irradiated materials. Simultaneous measurements of these parameters as a function of temperature will provide valuable information on the annealing properties and consequently on the endurance of a material. It can provide guidelines if annealing cycles can be introduced in the reactor operation sequences in order to prolong the reactor life time. Due to the pulsed nature of the SNQ, the samples can be studied under periodically applied stress loads, magnetic and electric fields in order to study fatigue phenomena of pre- or non-irradiated materials as a function of temperature and time.

In view of the above it is obvious that our main interest is concerned with simultaneous multiparametric measurements since the precious highly radioactive samples are changing their characteristic properties with temperature, cycling conditions and time. Therefore, we ought to measure simultaneously:

- Small Angle Scattering (SAS);
- Elastic Diffuse Scattering (EDS);
- Quasi-Elastic Scattering (QES);
- Bragg-Scattering (BS).

For that reason we propose the construction of a Material Science Spectrometer (MSS) that incorporates the four classical instruments in one. Depending on the sample condition, the intensity may differ very largely in the four spectrometers leading to very different measuring times for each of them. However, the total measuring time in the MSS will be considerably shorter than for four independent measurements with the possible disadvantage of loosing correlated phenomena. Estimations indicate that the construction cost of a MSS equals about the neutron production cost per year for one of the beam lines. Therefore the use of a  $2\pi$ -detector system favours strongly the overall economy of the experiments.

In the following we give a short description of the conceptional design of the spectrometer of which an artistic view is given in Fig. 15.

## 2. BASIC DESIGN CRITERIA FOR THE MATERIAL SCIENCE SPECTROMETER

### 2.1 Design criteria for the SAS-spectrometer

In Figs.1 and 2 a neutron scattering experiment is represented in real and momentum space. With the notations of Figs.1 and 2 the intensity at the detector is proportional to

$$I \approx d_T^2 \left( \frac{d_E}{l} \right)^2 \left( \frac{d_D}{L} \right)^2 \Delta E_i \cdot \Delta E_f = d_T^2 \cdot \Delta \Omega_i^* \cdot \Delta \Omega_j^* \cdot \Delta E_i \cdot \Delta E_f \quad (1)$$

or

$$I \approx d_T^2 \cdot \Delta v_i \cdot \Delta v_f$$

and for an elastic small angle scattering experiment the square of the width of the  $k$ -resolution function is given by

$$\begin{aligned} \langle \Delta k^2 \rangle &= \frac{1}{12} [(\Delta k_{\perp}^i)^2 + (\Delta k_{\perp}^f)^2 + (\Delta k_{\parallel} \vartheta)^2] \\ &= \frac{k^2}{12} \left[ \left( \frac{\Delta k_{\perp}^i}{k} \right)^2 + \left( \frac{\Delta k_{\perp}^f}{k} \right)^2 + \left( \frac{\Delta k_{\parallel}}{k} \cdot \vartheta \right)^2 \right] \\ &= \frac{k^2}{12} \left[ \Delta \Omega_i + \Delta \Omega_f + \left( \frac{\Delta k_{\parallel}}{k} \cdot \vartheta \right)^2 \right] \\ &= \frac{k^2}{12} \left[ \left( \frac{d_E + d_T}{l} \right)^2 + \left( \frac{d_T + d_D}{L} \right)^2 + \left( \frac{\Delta k_{\parallel}}{k} \cdot \vartheta \right)^2 \right] \end{aligned} \quad (2)$$

where  $\Delta k_{\perp} = \Delta k_x^{i,f} = \Delta k_y^{i,f}$  and  $\Delta k_{\parallel} = \Delta k_z^{i,f}$ .



Combining equations (1) and (2) we obtain a minimum for  $\langle \Delta \kappa^2 \rangle$  if

$$\Delta \Omega_i = \Delta \Omega_f = \left( \frac{\Delta k_{||}}{k} \vartheta \right)^2 \quad (3)$$

or

$$\frac{d_E + d_T}{\ell} = \frac{d_T + d_D}{L} = \frac{\Delta k_{||}}{k} \cdot \vartheta. \quad (4)$$

If we choose a symmetric solution in real space with  $\ell = L$  then

$$d_E = d_D \equiv d. \quad (5)$$

From equations (2), (4) and (5) we obtain

$$\langle \Delta \kappa^2 \rangle^{1/2} = \frac{k}{2} \frac{d + d_T}{\ell} \quad (6)$$

or

$$\frac{\langle \Delta \kappa^2 \rangle^{1/2}}{\kappa} = \frac{d + d_T}{2\ell \cdot \vartheta}. \quad (7)$$

From the above follows that we do not have to choose a symmetric condition in real space, we only have to guarantee that the divergency of the initial and that of the final beam are identical. Typical examples have been described by R.J. Stewart /13/, W. Kley /25/, Ph.A. Seeger /26/ and B. Alefeld and J. Schelten /27/.

Since we want to measure simultaneously elastic, thermal diffuse, quasi-elastic and small angle scattering, we consider a symmetric solution:  $\ell \equiv L$ .

## 2.2 Design criteria for the Elastic Diffuse Scattering Spectrometer

In order to understand mechanical properties and annealing phenomena in materials subjected to fast neutron irradiation and thermal cycling at elevated temperatures, such as in fusion reactors, we must measure simultaneously as a function of temperature the mechanical properties, the density of point defects and point defect clusters or voids, their size distribution as well as the microchemical evolution (precipitates) and texture. Hence, we must combine the SAS-spectrometer with an "Elastic Diffuse Scattering"-Spectrometer (EDS).

We assume now that the initial and final beam have the same divergency  $\Delta\vartheta$  and the same  $\Delta k_{||}/k$ , then we obtain from Fig.3 the momentum transfer resolution function

$$\frac{\Delta\kappa}{\kappa} = \frac{\Delta\vartheta}{\sqrt{3}} \cotg \frac{\vartheta}{2} + \frac{1}{\sqrt{12}} \cdot \frac{\Delta k_{||}}{k} (1 + \Delta\vartheta \cdot \cotg \frac{\vartheta}{2}) \quad (8)$$

which is shown in Fig.4 for a given set of values. For "back-scattered" neutrons the  $\kappa$ -resolution matches the time of flight resolution for a flight path of 78 m and a source pulse width of 100  $\mu$ sec. The use of cold neutrons is mandatory to avoid any double Bragg-reflexes and since the samples are either at ambient or elevated temperatures ( $T_s \leq 1000^\circ\text{C}$ ) the inelastically upscattered neutrons can be discriminated from the elastic scattered neutrons by detector gating. Therefore, the SAS-spectrometer can be combined with the elastic diffuse spectrometer without any disadvantage for either one.

## 2.3 Design criteria for the Quasi-Elastic Neutron Spectrometer

The neutron spectra in the first wall and blanket of fusion reactors produce hydrogen and helium besides other transmutation products. The hydrogen causes embrittlement and the helium is partly responsible for swelling.

The diffusion properties and activation energies of hydrogen, interstitials and vacancies in materials approaching end-of-life conditions will certainly be different from unirradiated materials. By means of measurements of the  $\kappa$ -dependence of the width of the quasi-elastic line, we can determine the diffusion properties and activation energies of hydrogen as a function of dose and temperature. Using pyrolytic graphite crystals in "back-scattering" geometry, we can easily match the time of flight resolution of 1  $^{\circ}$ /oo for  $\lambda = 6.69 \text{ \AA}$  and a flight path of 80 m. Again, this spectrometer can be incorporated in the SAS- and EDS-spectrometer without any disadvantage for either one.

#### 2.4 Design criteria for the Bragg-Scattering Spectrometer

For the understanding of the mechanical properties of dynamically stressed and plastically deformed construction materials, it is important to measure among other parameters the variations of the textures. For highly radioactive materials this is done best by mapping the intensity variation in a number of Debey-Scherrer-Rings by using Position Sensitive He<sup>3</sup>-detectors in the "Semi-Ring-Detectors" also used for EDS-measurements and by rocking the sample.

### 3. CONCEPTUAL DESIGN OF THE MATERIAL SCIENCE SPECTROMETER

#### 3.1 Introduction

The first small angle scattering of X-rays has been observed by Krishnamurti<sup>1</sup> in 1930 and in the subsequent decades particular attention has been devoted to this new field<sup>2</sup>.

Small angle neutron scattering has been discussed first by Halpern<sup>3</sup> in 1948 and small angle scattering of neutrons was first found by Hughes, Burgy, Heller and Wallace<sup>4,5</sup> in 1949.

The advent of medium and high flux reactors have stimulated also the development of new instruments and methods<sup>6-13</sup>. The design and construction of pulsed neutron sources is still furthering developments in these areas<sup>14-19</sup>. The advent of the new generation of small angle neutron scattering facilities<sup>11,12,13</sup> has stimulated the investigations of structural problems in solid state physics, physico-chemistry, metallurgy and biology.

For a SAS-spectrometer there are two competing solutions for the formation of the initial beam, which are concerned with the part of the spectrometer between the neutron source and the target. One is based on the use of neutron guide tubes as practiced at Jülich and Grenoble, the other is based on the use of filters, iris diaphragma or rotating collimators as described by R.J. Stewart<sup>13</sup>, W. Kley<sup>25</sup>, Ph. Seeger<sup>26</sup> and B. Alefeld and J. Schelten<sup>27</sup>, which permit to decouple the collimation of the primary beam from the sample size. However, the beam divergence of the scattered beam is determined in both cases by the target and detector size. Both solutions have their advantages and drawbacks and the use of either one depends partially on the background situation, on the "program" and the available sample size. For a Material Science Program the sample size must not be a limiting factor in most of the cases. If the sample size ( $d_T^2$ ) can be matched to a large detector area element ( $d_D^2$ ) the intensity is proportional to the square of the length of the SAS-spectrometer if measurements with equal momentum transfer resolution are compared.

Furthermore, we have to deal primarily with polycrystalline materials

which demand the use of cold neutrons instead of a white neutron beam in order to avoid double Bragg reflections in the sample.

In view of the above and considering our program that requests the simultaneous measurement of elastic small angle, elastic diffuse, quasi-elastic and Bragg-scattering from highly radioactive samples, we consider a symmetric solution but keep in mind that for certain cases the intensity and resolution requirements for the EDS-, QES- and the BS-spectrometers might favour an asymmetric solution.

### 3.2 The design proposal

The sketch of the layout of the Materials Science Spectrometer is given in Figs. 5 and 6. A straight nickel plated, "mobile" glass guide with an inner cross section of  $2.5 \cdot 5 \text{ cm}^2$  guides the neutrons from the cold neutron source to the sample. The 1000  $\mu\text{sec}$  pulse from the cold neutron source may be shaped, according to a suitable compromise between required resolution and intensity, by two He-cooled neutron pulse shaping choppers, constructed of 10 cm wide aluminium cylinders, sprayed on the inside either with boron-carbide or cadmium depending on the heat transfer problems. The magnetically suspended choppers produce neutron pulses in the range from 50 to 1000  $\mu\text{sec}$ . The Space-Time-Diagram of the two Neutron-Pulse-Shaping-Choppers is given in Fig. 7. For each 1000  $\mu\text{sec}$  Neutron-Source-Pulse, the first chopper produces three neutron pulses with various band widths ranging from

11 000 to 909 m/sec  
1 000 to 476 m/sec and  
500 to 333 m/sec.

The pulse duration is zero for the highest and lowest neutron energy in each band and reaches a pulse width of 80  $\mu\text{sec}$  for a certain region within each neutron band. The pulse forming slit "1" is 2.5 cm wide, matched to the width of the neutron guide. The second chopper is phased in such a manner as to select one of the three pulses.

Outside the biological shield two overlap rotors are positioned in such a manner as to produce at the two-dimensional detector in its maximum

distance from the target, a continuous quasi-white neutron beam without overlap<sup>28</sup>.

We suppose that the first overlap rotor may be installed  $\ell_1$  metres from the moderator. From Fig. 8 follows that the position of the second rotor ought to be fixed by the following condition:

$$2T - t_2 = t_1 - \left( \frac{\ell_2}{v_0} + T \right) \quad (9)$$

and consequently

$$\ell_2 = \frac{3}{2} \ell_1. \quad (10)$$

If there are still neutrons below  $v_3$  then a third rotor has to be installed at  $\ell_3$  which is determined by the following condition:

$$4T - t_4 = t_3 - \left( 3T + \frac{\ell_3}{v_0} \right) \quad (11)$$

from which follows:

$$\ell_3 = \frac{7}{4} \ell_1.$$

Note that these positions are independent of the neutron source pulse frequency  $f$  and the total length  $\ell_t$  of the SAS-spectrometer. However, these parameters determine the overlap condition:

$$\frac{v_{\max} - v_{\min}}{v_{\max} \cdot v_{\min}} \cdot \ell_t \approx \frac{\Delta v}{v} \cdot \frac{\ell_t}{v} = T = \frac{1}{f}. \quad (12)$$

For a total length of the flight path,  $\ell_t = 150$  m,  $v = 900$  m/sec and  $f = 100$  Herz, we obtain a band width of  $\Delta v/v = 0.06 \approx 6\%$ . The useful band width or in general the neutron source economy is inversely proportional to the neutron source pulse frequency as long as the average neutron flux stays constant.

The overlap rotors consist actually each of two disks which turn both at the same angular speed and direction but their relative position to each other can be changed in order to vary the width of the "window"

and to guarantee a continuous "quasi-white" neutron beam at each position of the SAS-detector.

The function of the third overlap rotor at  $\ell_3$  can be taken over by the "low frequency" neutron source pulse shaping chopper if it can be installed at a position  $\ell_0 \leq \ell_x$  in the main shield. From Fig. 8 follows that

$$\ell_0 \leq \ell_x = \frac{\ell_1}{2 + \frac{\ell_1}{V_0} \cdot f} \quad (13)$$

where  $\ell_1$  is the position of the first overlap rotor,  $V_0$  the minimum neutron velocity and  $f$  the neutron source pulse frequency. For  $\ell_1 = 6$  m,  $V_0 = 900$  m/sec, and  $f = 100$  Herz we obtain  $\ell_x = 2.25$  metres.

Two background rotors, each 75 cm long, shield target and detectors from direct fast neutron background. The target station must be accessible to man even during operation without closing the main shutter.

At the entrance to the target station, the beam cross section is limited to  $2.5 \times 2.5$  cm<sup>2</sup> leaving half the neutron guide cross section available for another facility.

The evacuated scattering chamber has a diameter of 7 m and it incorporates semi-ring detectors partially or totally equipped with position sensitive He<sup>3</sup>-detectors, 7 analysing crystal spectrometers, shown in Fig. 6, in "quasi-back scattering" position, and electron guns to heat the samples on temperatures up to 1000°C without using heat shields.

The scattering chamber can be shut off from the neutron guide and the 1.5 m wide, 75 m long neutron drift tube by vacuum valves.

For neutrons of  $\lambda = 5.1243$  Å or  $k = 1.226$  Å<sup>-1</sup> the neutron guide delivers a beam with a divergence of  $\Delta\vartheta = 1^\circ$ . In order to adapt the beam divergence of the scattered beam to the initial beam, we assume a target and detector diameter of 2.5 cm which leads to a secondary flight path,  $\ell_f \approx 300$  cm for the EDS-spectrometer. In the case of polycrystalline or homogeneous samples, the neutrons with the same  $|\vec{k}|$  are scattered into cones around the axis of the impinging neutrons. We want to detect as many of them as possible in so-called ring detectors built up of straight

detectors, as shown in Fig. 9. From Fig. 9 follows

$$\frac{\Delta\rho}{\rho} = -1 + \sqrt{1 + \left(\frac{x}{\rho}\right)^2} \leq \frac{\Delta\kappa}{\kappa} \quad (14)$$

or

$$x = \rho \sqrt{2 \frac{\Delta\kappa}{\kappa}} = \ell_f \cdot \sin\vartheta \sqrt{2 \frac{\Delta\kappa}{\kappa}} \quad (15)$$

From equations (8) and (15) follows

$$x = \sqrt{2} \ell_f \cdot \sin\vartheta \left[ \frac{\Delta\vartheta}{\sqrt{3}} \cotg \frac{\vartheta}{2} + \frac{1}{\sqrt{12}} \cdot \frac{\Delta k_{||}}{\kappa} (1 + \Delta\vartheta \cdot \cotg \frac{\vartheta}{2}) \right]^{1/2} \quad (16)$$

For  $\Delta\vartheta = 1^\circ$  and  $\ell_f = 300$  cm, the maximum admissible detector length,  $\ell_D = 2x$ , is plotted as a function of  $\vartheta$  in Fig. 10.

In Fig. 6 the position of 7 pyrolytic graphite crystals is shown in "back-scattering" geometry. For the QES-spectrometer we assume a  $\Delta\vartheta \approx 1^\circ$  and  $\Delta\lambda/\lambda = 10^{-3}$  or a scattering angle of  $\vartheta = 86.71^\circ$  for a  $\lambda = 6.686$  Å or a neutron temperature of 21 K, the liquid hydrogen moderator temperature. The crystals are positioned 1 m from the target and the distance target-crystal-detector is 5 m. Special collimators prevent that neutrons can arrive directly from the target at the detectors of interest for the QES-spectrometer. For crystals of 20 cm height, the detector may be as long as 1 m.

The SAS-detector consists of 50 position-sensitive  $\text{He}^3$ -detectors, each with a diameter of 2.5 cm and an active length of 50 cm. The spatial resolution along the detector is smaller than 1 cm. The detector covers an active area of  $125 \times 50 \text{ cm}^2$ . It is positioned in a drift tube with a diameter of 1.5 m and a length of 75 m. The detector can be positioned in concordance with the mobile neutron guide tube at a distance from the target equal to 3, 10, 20, 30, 40, 60 and 75 m. As can be seen from Fig. 6, the detector covers only one half of a plane perpendicular to the beam axis in order to measure the largest possible range of  $\kappa$  simultaneously.



For all technological experiments we cannot miss any essential information. However, in some special cases, important Bragg reflexes could be missed, etc. They can be found by turning the sample by  $180^\circ$  around an axis perpendicular to the beam axis in order to measure the complete symmetry. For technological experiments we have most of the time complete symmetry around the beam axis and therefore we could use "ring-detectors" instead of a two-dimensional SAS-detector.

From equations (7) and (15) follows

$$\begin{aligned} \ell_D &= 2x = 2\ell_f \cdot \sin\vartheta \cdot \sqrt{2 \frac{\Delta\kappa}{\kappa}} \\ &= 2\ell_f \cdot \sin\vartheta \sqrt{2 \frac{d+d_T}{2\ell_f \cdot \vartheta}} = 2\sqrt{(d+d_T) \cdot \ell_f \cdot \vartheta} \\ \ell_D &= 2\sqrt{(d+d_T) \cdot \rho} \end{aligned}$$

For  $d = d_E = d_D = d_T = 2.5$  cm,  $\rho = 5$  and  $55$  cm leads to  $\ell_D^5 = 10$  cm and  $\ell_D^{55} = 33$  cm.

Therefore, we could easily build such a ring detector with conventional straight  $\text{He}^3$ -detectors. Since we want to cover a broader range of interest we use the two-dimensional  $\text{He}^3$  position-sensitive detector.

### 3.3 The General Overlap-Criteria

Since our samples are predominantly at ambient or high temperature (low temperatures are not excluded), we are interested in separating the elastically and inelastically scattered neutrons by time gating. From Fig. 11 follows:

$$t_1 = \frac{\ell_s}{v_{\text{el min}}} + \frac{(\ell_D - \ell_s)}{v_{\text{inel min}}} = \frac{\ell_D}{v_{\text{el max}}} \quad (17)$$

and

$$t_o = t_2 - T = \frac{\ell_s}{v_{\text{el max}}} + \frac{(\ell_D - \ell_s)}{v_{\text{inel max}}} = \frac{\ell_D}{v_{\text{el min}}} - T. \quad (18)$$

From equation (17) and (18) we obtain:

$$v_{\max}^{\text{el}} = \frac{(\ell_D^2 - \ell_s^2)}{\left[ T \cdot \ell_s + \frac{\ell_s (\ell_D - \ell_s)}{v_{\max}^{\text{inel}}} + \frac{\ell_D (\ell_D - \ell_s)}{v_{\min}^{\text{inel}}} \right]} \quad (19)$$

$$v_{\min}^{\text{el}} = \frac{(\ell_D^2 - \ell_s^2)}{\left[ T \cdot \ell_D + \frac{\ell_D (\ell_D - \ell_s)}{v_{\max}^{\text{inel}}} + \frac{\ell_s (\ell_D - \ell_s)}{v_{\min}^{\text{inel}}} \right]} \quad (20)$$

and from (19) and (20) follows for the admissible neutron band width

$$\frac{1}{2} \cdot \frac{\Delta v}{\bar{v}} = \frac{T \cdot v_{\max}^{\text{inel}} \cdot v_{\min}^{\text{inel}} + c(v_{\min}^{\text{inel}} - v_{\max}^{\text{inel}})}{T \left( \frac{2\ell_s}{c} + 1 \right) \cdot v_{\max}^{\text{inel}} \cdot v_{\min}^{\text{inel}} + (2\ell_s + c)(v_{\min}^{\text{inel}} + v_{\max}^{\text{inel}})} \quad (21)$$

where

$$\ell_D - \ell_s = c \quad \text{and} \quad \bar{v} = \frac{1}{2} (v_{\max}^{\text{el}} + v_{\min}^{\text{el}}).$$

$v_{\max}^{\text{el}}$ ,  $v_{\min}^{\text{el}}$  and  $\frac{\Delta v}{\bar{v}}$  depend on the design parameters of the MSS-spectro-

meter and the sample temperature  $T_s$  that determine  $v_{\max}^{\text{inel}}$  and  $v_{\min}^{\text{inel}}$ . In our case

we assume  $T_s \geq T_n$ , the temperature of the impinging neutrons. Considering a fixed distance between sample and detector:  $\ell_D - \ell_s = \text{constant} = c$ , the useful band width or the intensity is increasing and the time resolution is decreasing with  $\ell_s^{-1}$  as in the case of pure elastic scattering. However, the intensity does not increase linearly with  $T$ , the neutron source pulse period, as it is the case for pure elastic scattering. This is not surprising since our "general overlap condition" is formulated in such a way that the "time-utilisation factor" of the detector system is equal to one. The "time utilisation factor" is the fraction of  $T$  during which neutrons arrive at the detector, characterizing also to what extent optimum conditions are achieved at a time of flight spectrometer. Since the inelastically scattered neutrons have to be separated from the elas-

tically scattered, the useful band width  $\frac{\Delta V}{V}$  or the intensity of the impinging neutrons cannot be increased proportionally to  $T$ .

For a typical set of parameters the intensity is plotted as a function of  $l_s$  and  $T$  in Fig. 12 and in Fig. 13.

If  $T_s \gg T_n$  detector gating becomes very inefficient leading to a small "time utilisation factor". For this case it is necessary to use a "chopper-assisted" detector gating procedure in order to eliminate inelastically scattered neutrons, as shown in Fig. 14, and to maintain a high time utilisation factor for purely elastically scattered neutrons, that are only of interest in the EDS-spectrometer.

From Fig. 14 follows:

$$t_o = \frac{l_s}{v_{el}^{max}}, \quad t_1 = \frac{l_D}{v_{el}^{max}} \quad (22)$$

and

$$t_{2n+1} = \frac{l_D}{l_s} \cdot t_{2n} \quad \text{for } n = 1, 2, 3, 4, \dots, \quad (23)$$

$$t_{2n+2} = t_{2n+1} - \frac{(l_D - l_s)}{v_{inel}^{min}} \quad \text{for } n = 0, 2, 4, 6, \dots, \quad (24)$$

$$t_{2n+2} = t_{2n+1} - \frac{(l_D - l_s)}{v_{inel}^{max}} \quad \text{for } n = 1, 3, 5, \dots \quad (25)$$

For a certain value  $n \equiv X$  :

$$t_{2X+1} - t_1 \leq T \quad (26)$$

These time values define the time gates and the "chopper" design parameters. For  $v_{el}^{max} = 1000$  m/sec,  $v_{el}^{min} = 875$  m/sec,  $v_{inel}^{max} = 3000$  m/sec and  $v_{inel}^{min} = 1500$  m/sec one obtains a time utilisation factor of about 35%, as it can be seen from Fig. 14.

#### 4. CONCLUSIONS

It is shown that four classical instruments for elastic small angle, elastic diffuse, quasi-elastic (including inelastic) and Bragg-scattering can be combined in one instrument at a pulsed neutron source such as the SNS at the Rutherford Appleton Laboratory (UK) or the SNQ at Jülich (FRG). Therefore, multiparametric measurements can be carried out simultaneously and a high neutron source economy is achieved for studies concerning fission and fusion reactor construction materials, fuel elements, reactor safety and economy, radiation damage and the testing and development of new alloys under cycling stress-conditions. In spite of the large number of detectors the construction cost of this facility equals about the neutron production cost per year for one beam line. Therefore, the use of a large number of detectors favours the overall economy of the experiments. An artistic view of the Materials Science Spectrometer is given in Fig.15.

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## 6. FIGURE CAPTIONS

- Fig.1 Collimation conditions for a SANS-experiment.
- Fig.2 Representation of a neutron scattering experiment in momentum space.
- Fig.3 The momentum space representation for the case of elastic diffuse scattering (EDS).
- Fig.4 The momentum transfer resolution function for the EDS-spectrometer.
- Fig.5 Conceptual design of the material science spectrometer.
- Fig.6 Conceptual design of target- and detector system.
- Fig.7 The space-time diagram of the two neutron-pulse-shaping-choppers.
- Fig.8 The space-time diagram of a "quasi-white" neutron beam.
- Fig.9 Representation of neutron scattering in real and momentum space for the case of polycrystalline or homogeneous samples.
- Fig.10 The maximum admissible length of a straight detector for the EDS-spectrometer.
- Fig.11 The general overlap-criteria.
- Fig.12 Admissible intensity as a function of  $l_s$ , the distance between neutron-source and sample.
- Fig.13 Admissible intensity as a function of  $T$ , the neutron source pulse period.
- Fig.14 Chopper assisted detector-gating for pure elastic scattering in the EDS-spectrometer.
- Fig.15 Artistic view of the material science spectrometer.

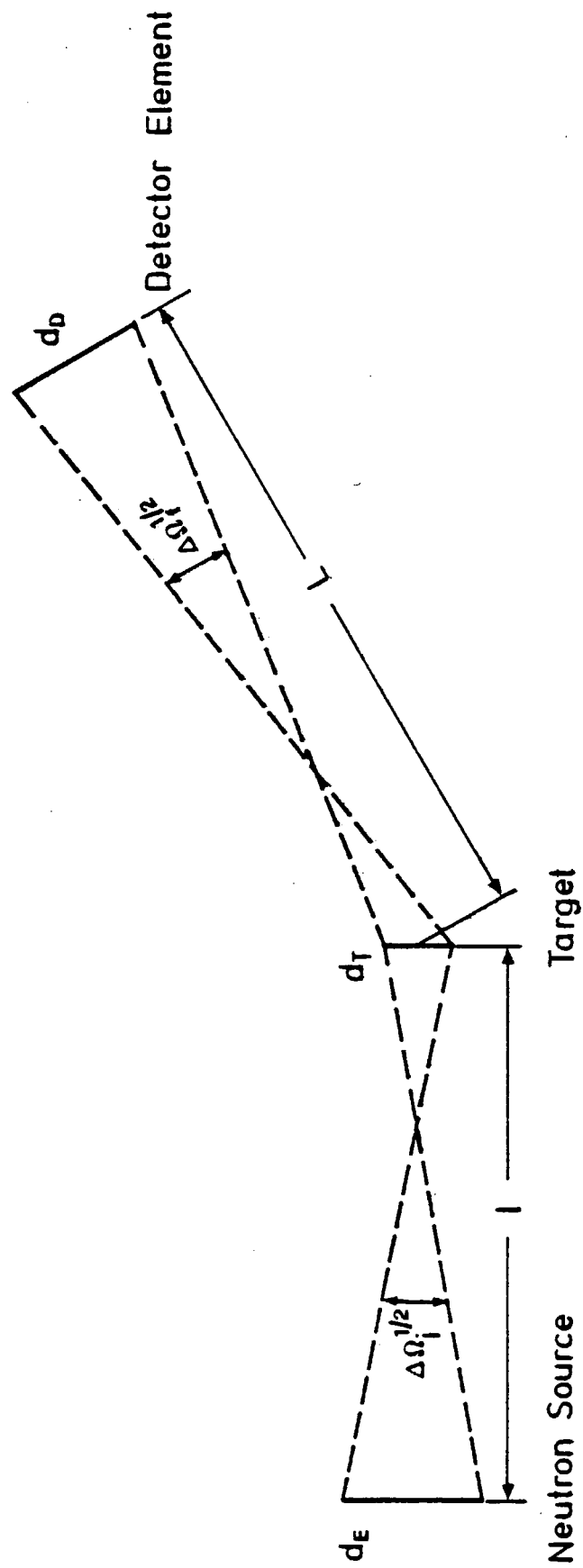


Fig.1. Collimation Conditions for a SANS - Experiment



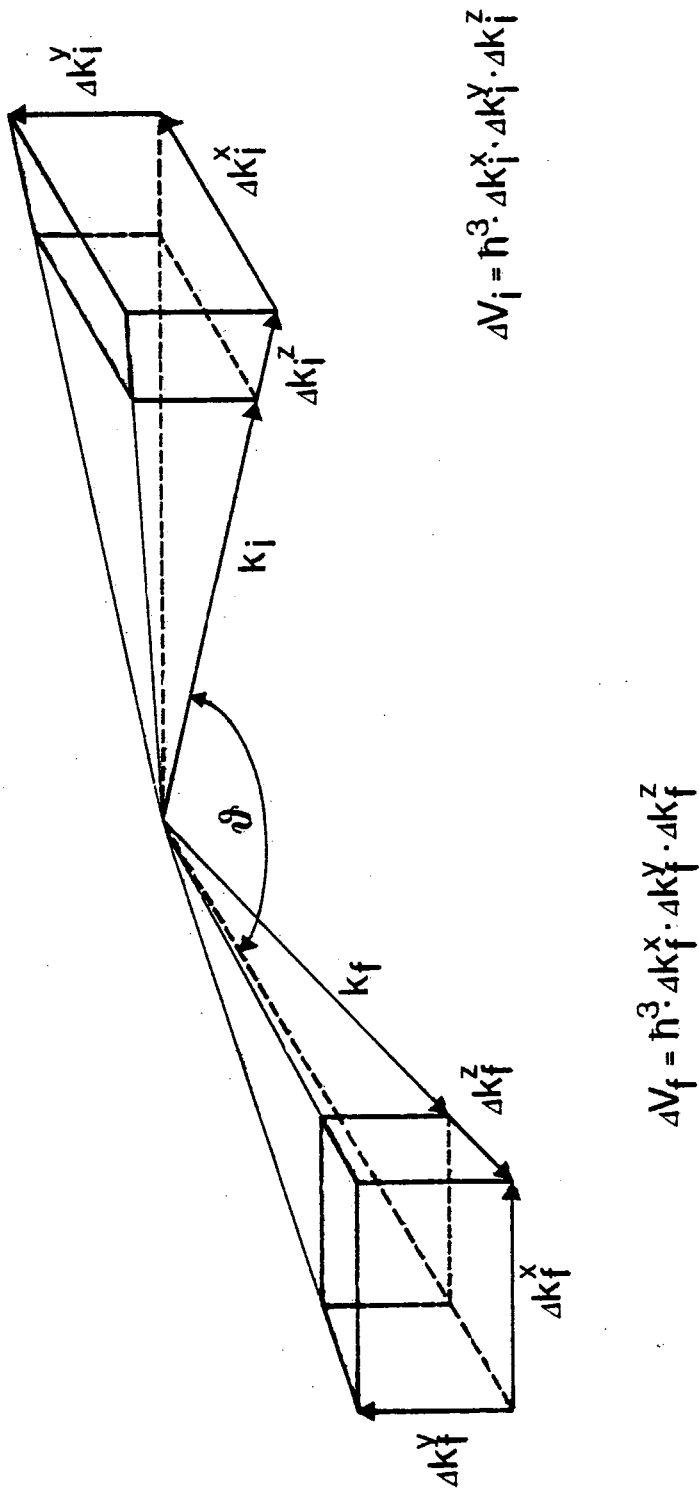


Fig.2 : Representation of a Neutron Scattering Experiment  
in Momentum Space

$$\frac{\Delta K}{K} = \frac{1}{\sqrt{12}} \left( \frac{K_{max} - K_{min}}{K} \right)$$

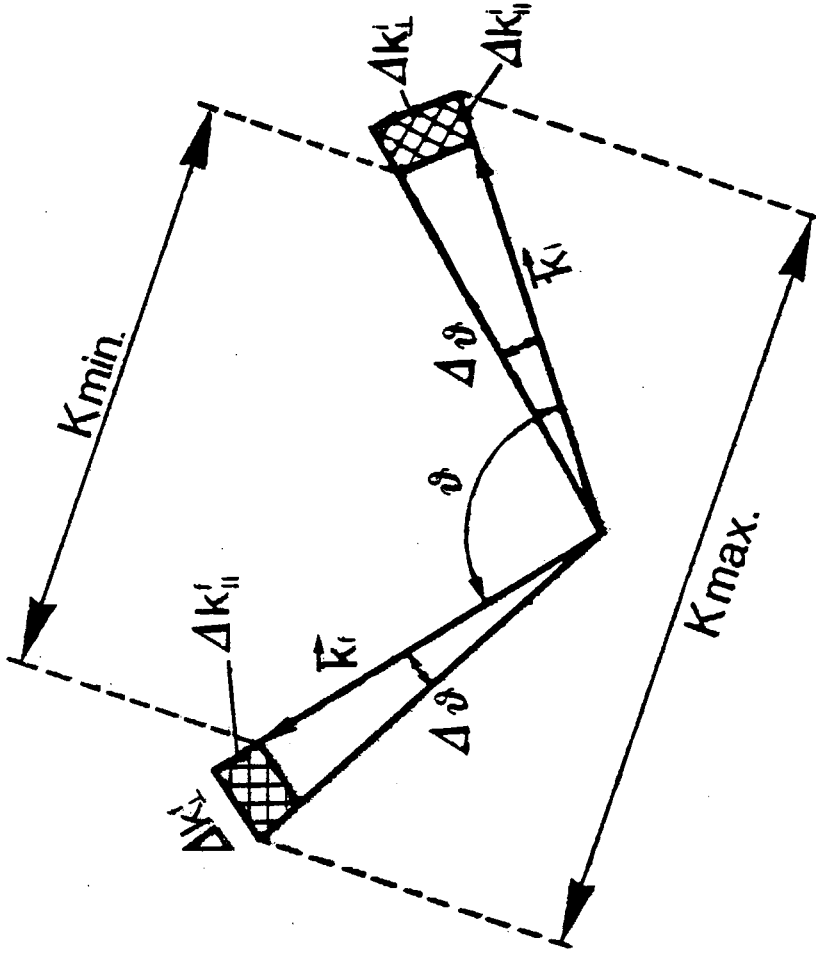


Fig 3 The momentum space representation for the case of elastic diffuse scattering (EDS)

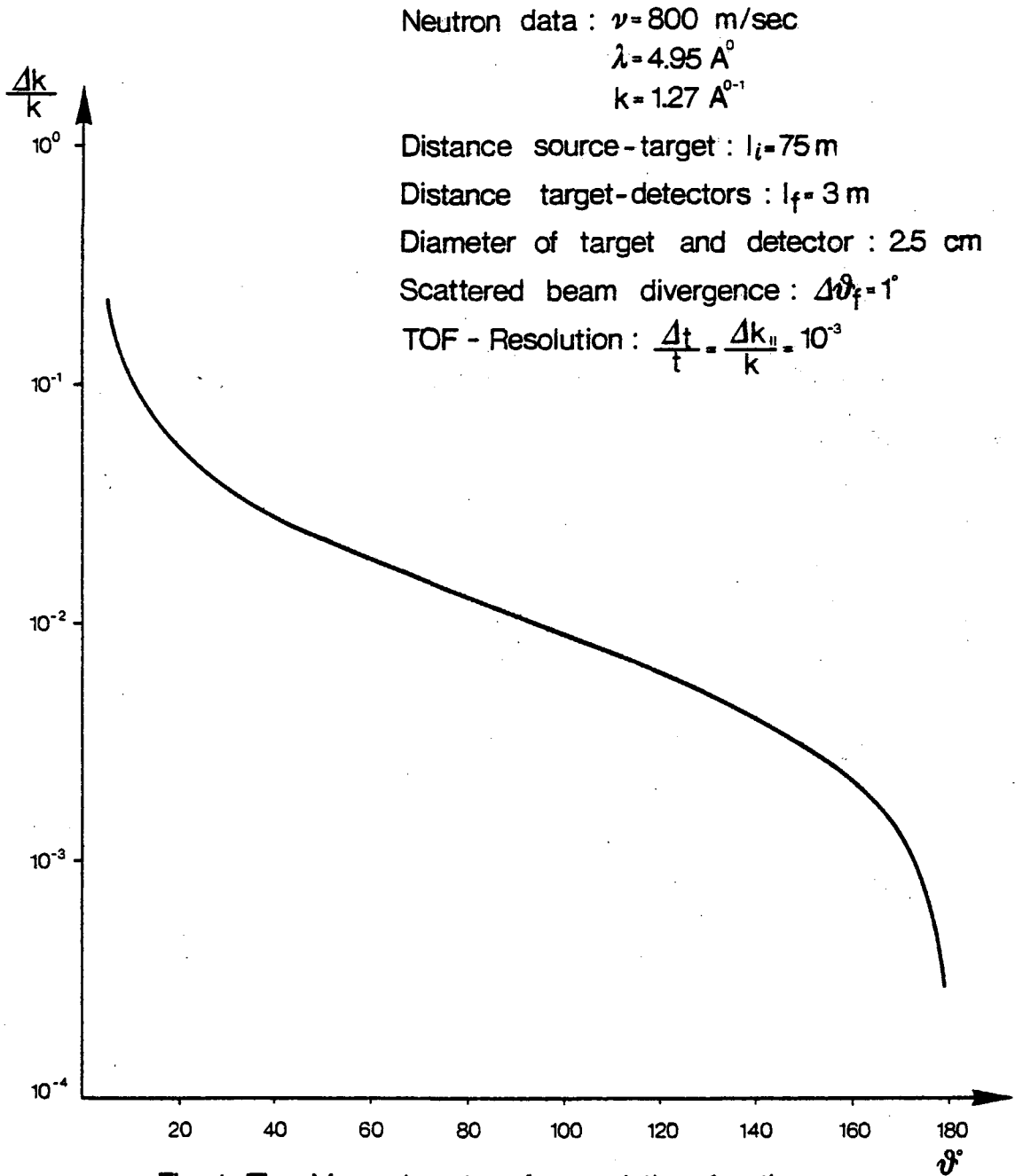
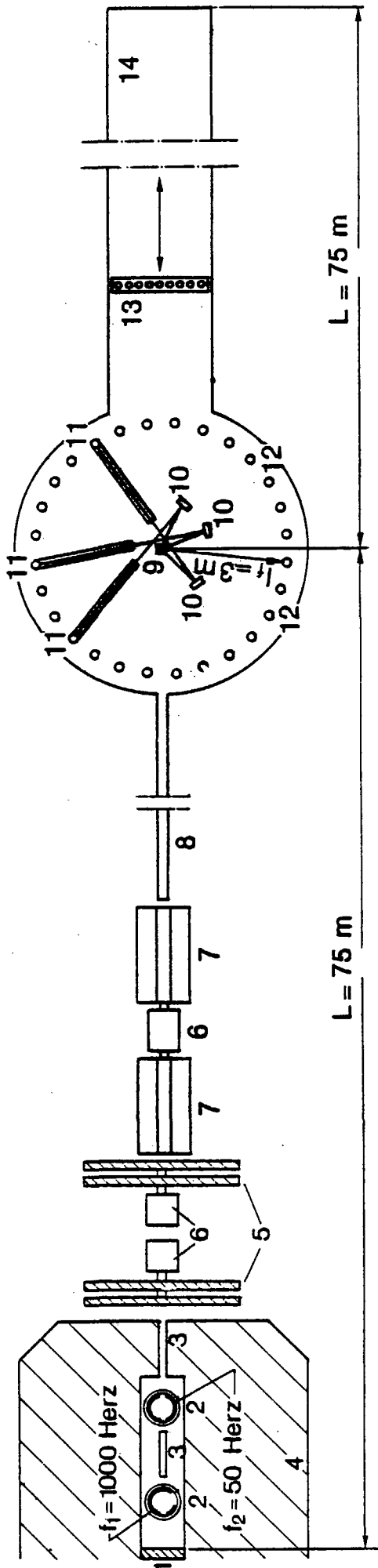


Fig. 4 The Momentum transfer resolution function for the EDS-spectrometer



- |                                                                   |  |
|-------------------------------------------------------------------|--|
| 1 Cold Neutron Source of SNQ                                      |  |
| 2 Neutron-Pulse-Shaping-Choppers                                  |  |
| 3 Fixed Neutron Guide: 2.5 x 5 cm <sup>2</sup>                    |  |
| 4 Biological Shield                                               |  |
| 5 Overlap-Rotors                                                  |  |
| 6 Driving Motors                                                  |  |
| 7 Background-Rotors                                               |  |
| 8 Mobile Straight Neutron Guides 2.5 x 5 cm <sup>2</sup>          |  |
| 9 Target                                                          |  |
| 10 Analysing Crystals                                             |  |
| 11 Straight He <sup>3</sup> -Detectors: 2.5 x 100 cm <sup>2</sup> |  |
| 12 Semi-Ring-He <sup>3</sup> Detectors                            |  |
| 13 Mobile Two-Dimensional Detector                                |  |
| 14 Drift Tube $\phi = 135$ cm                                     |  |

$$f = 100 \text{ Herz}$$

## Fig. 5a Conceptual Design of The Material - Science Spectrometer

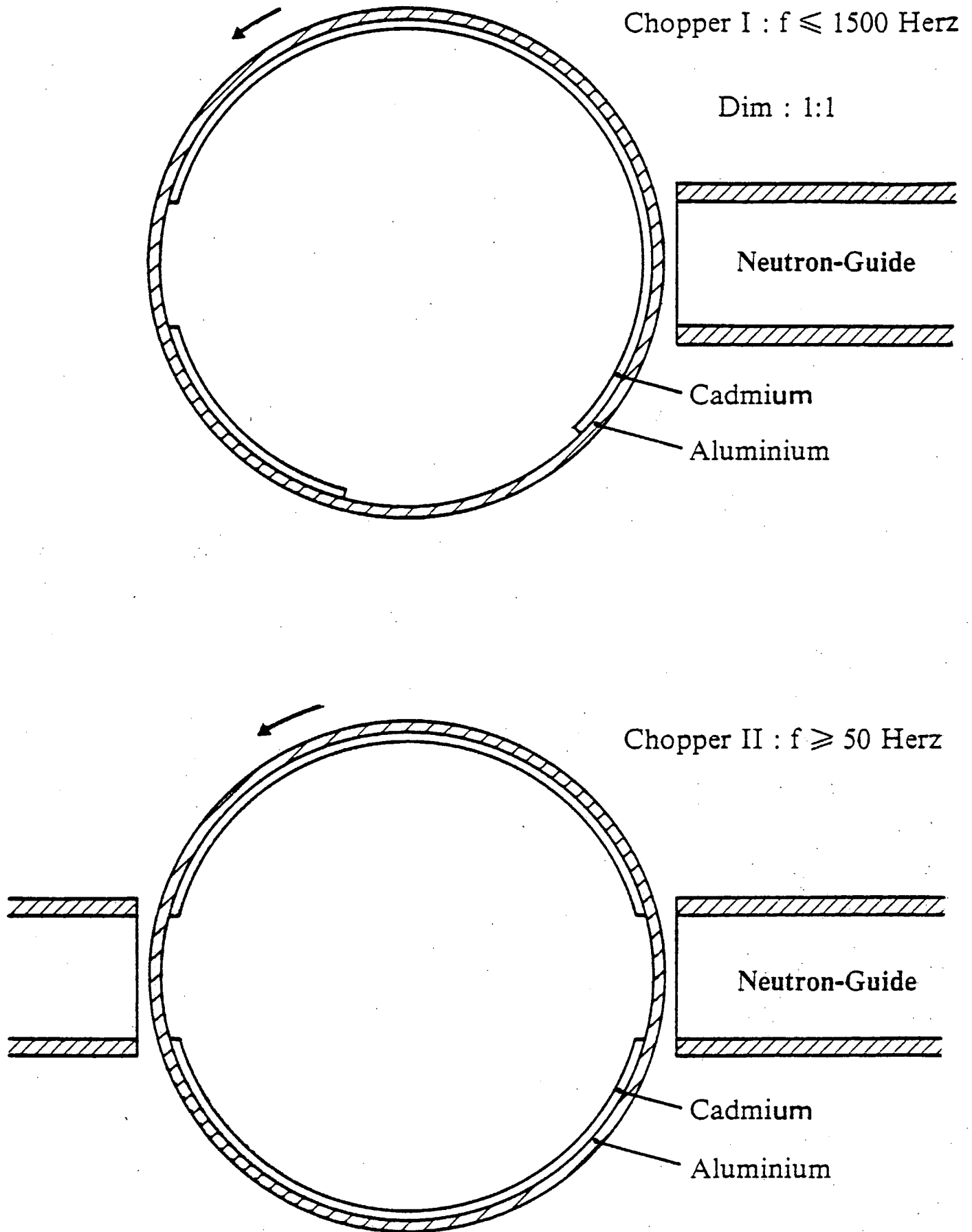


Fig. 5.b. The Neutron Pulse Shaping Choppers

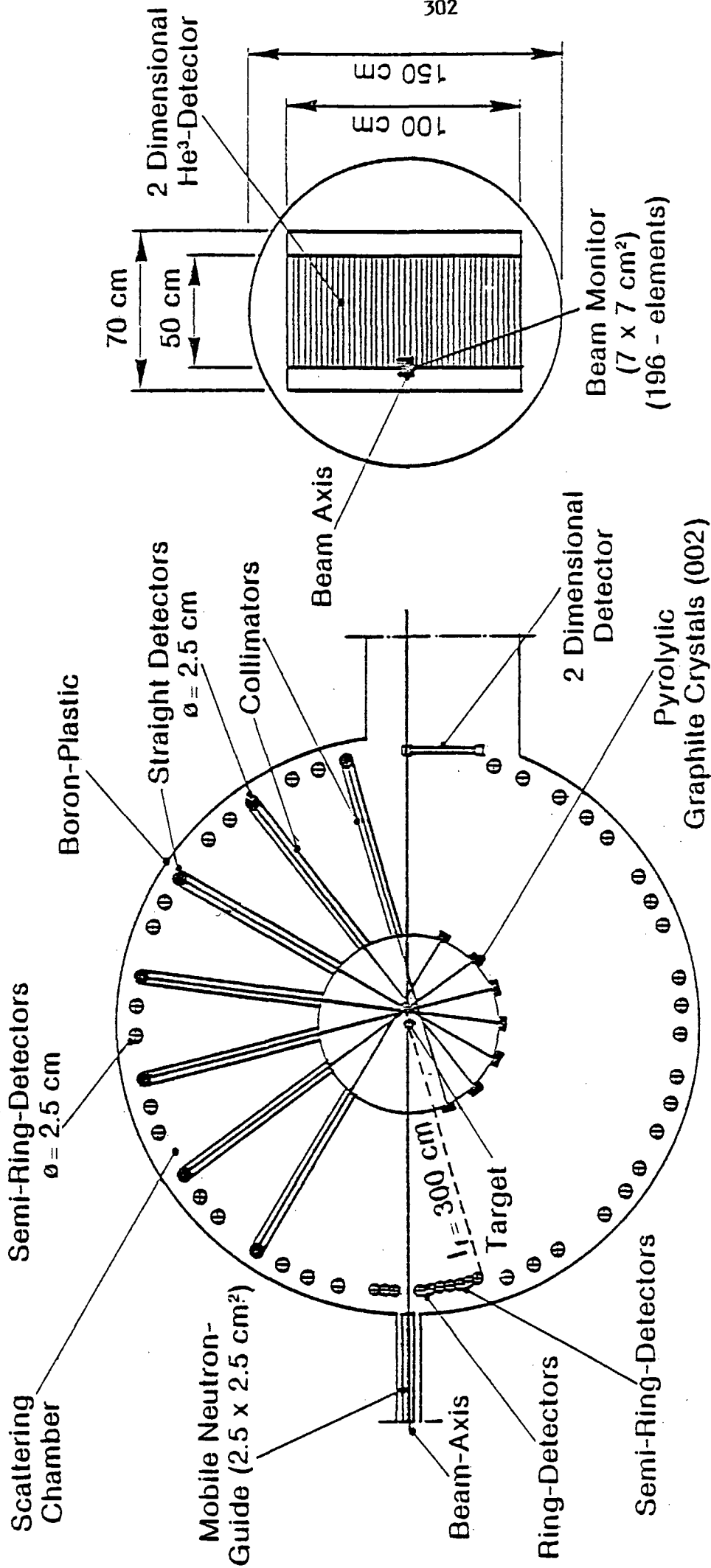


Fig. 6.a.

Fig. 6.b.

Cross Section of EDS - and QES - Spectrometer

Cross Section of Drift Tube  
and SAS - Detectors

# Fig. 6 Conceptional Design of Target and Detector System

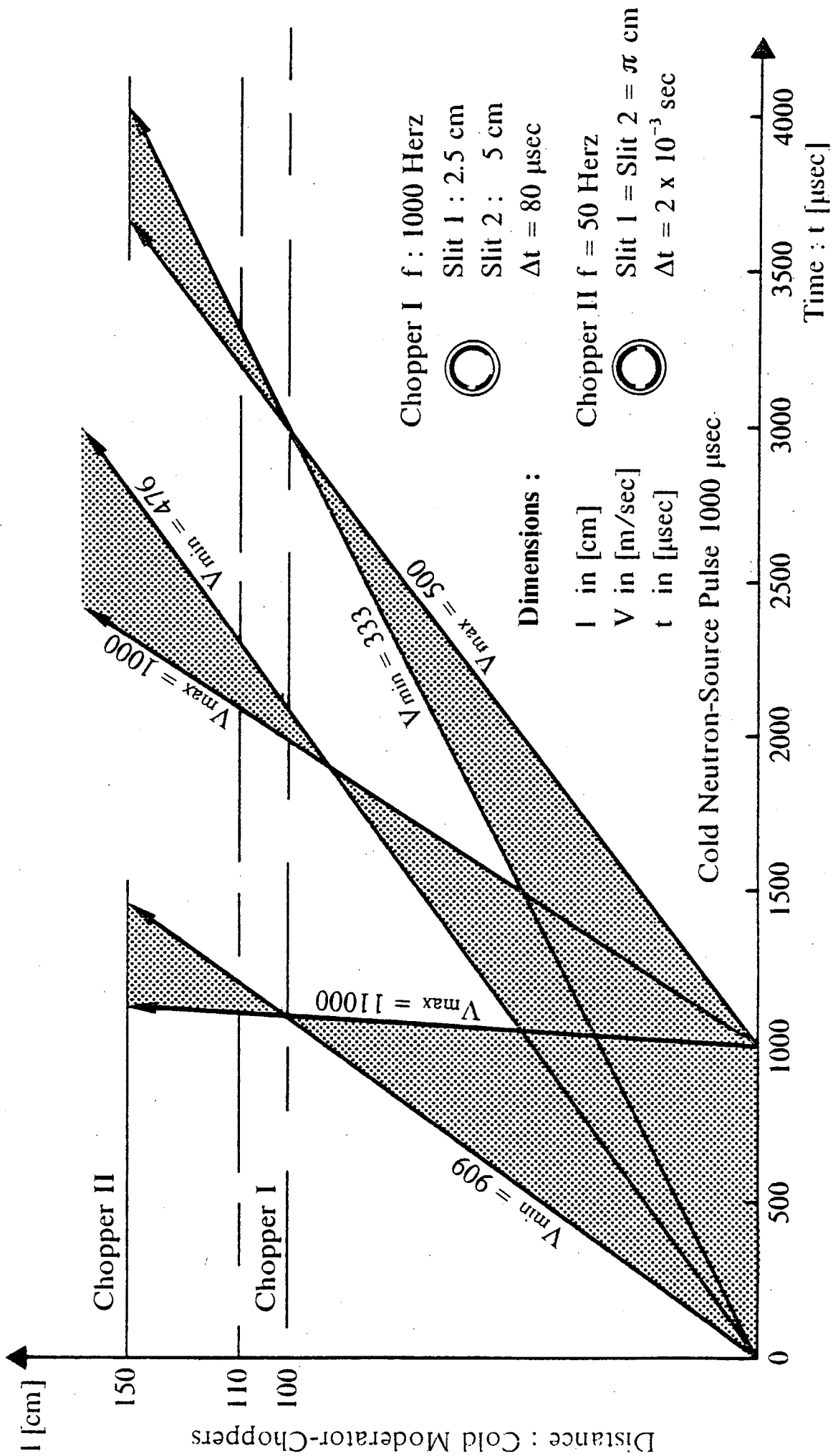


Fig. 7 The Space-Time-Diagram of the two Neutron-Pulse-Shaping-Choppers

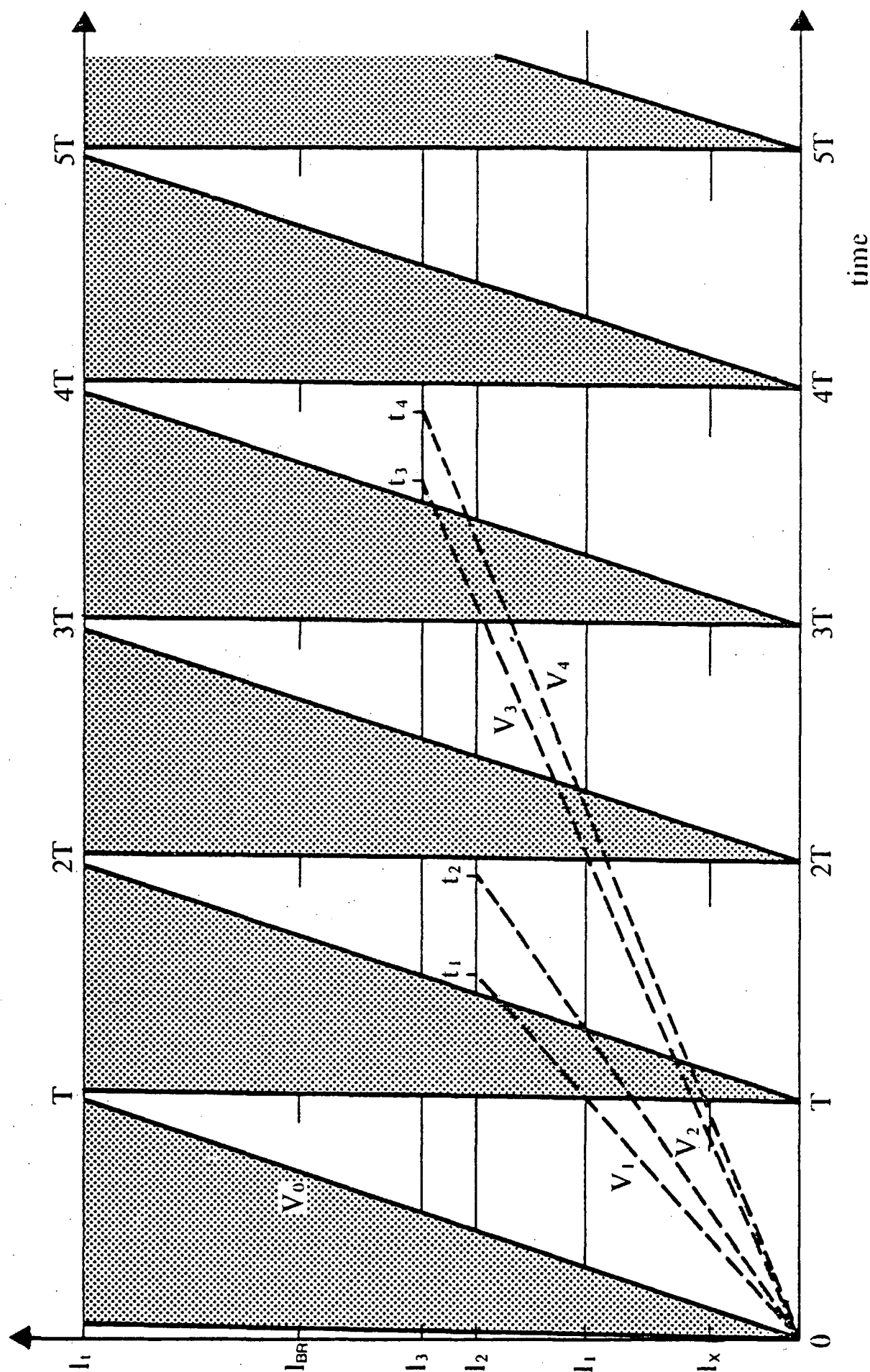


Fig. 8 The time space diagram of a "Quasi-White" neutron beam



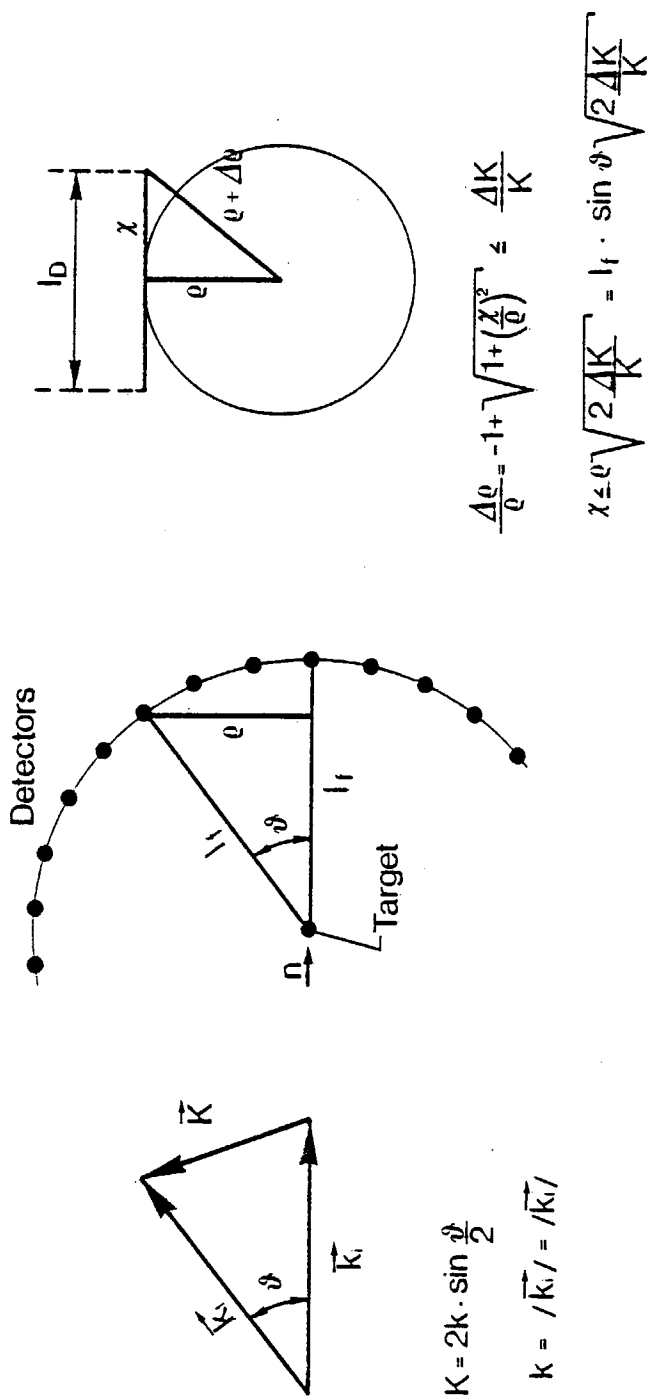


Fig.9 representation of neutron scattering in real and momentum space for the case of polycrystalline or homogeneous samples

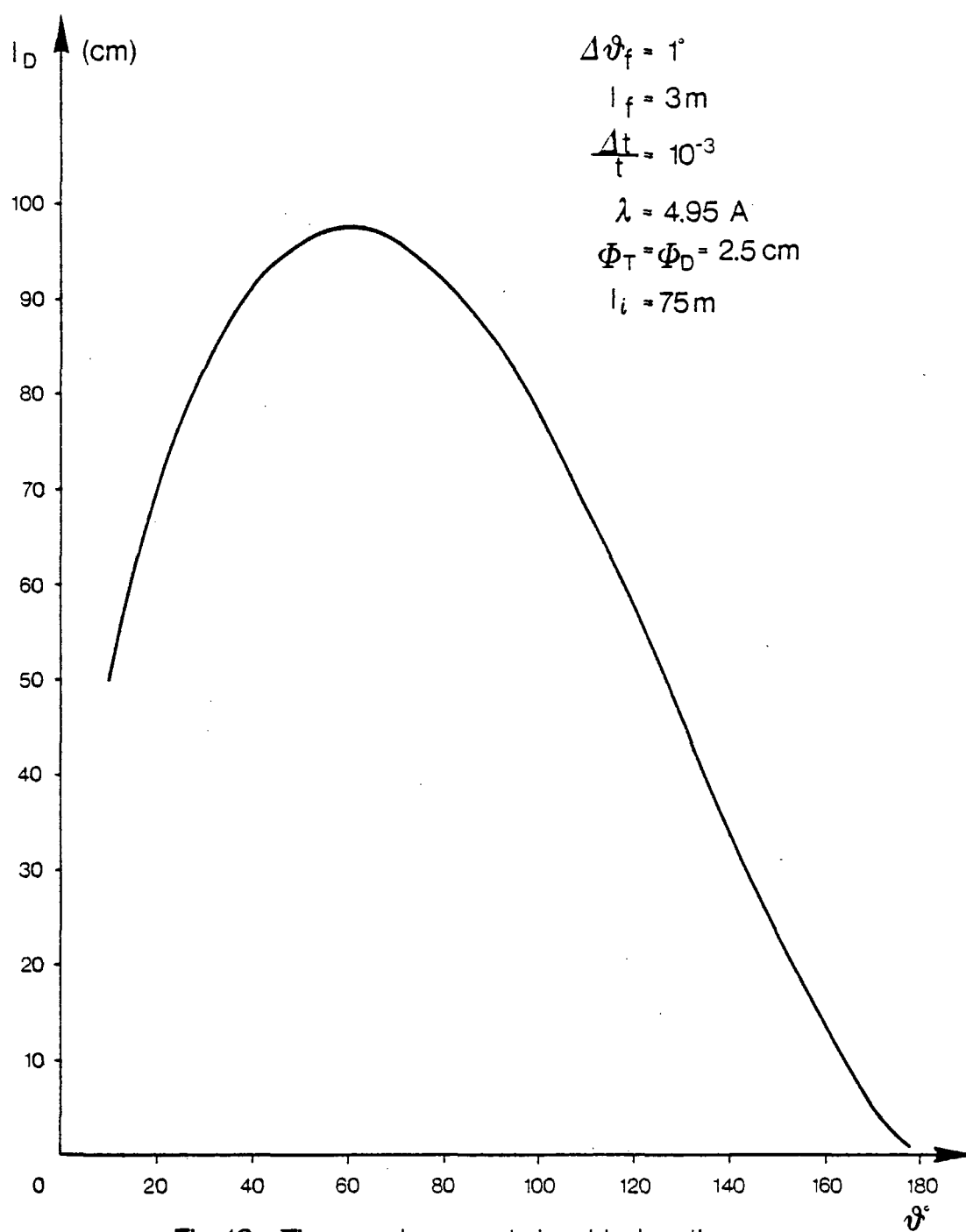


Fig. 10 The maximum admissible length of a straight detector for the EDS - spectrometer

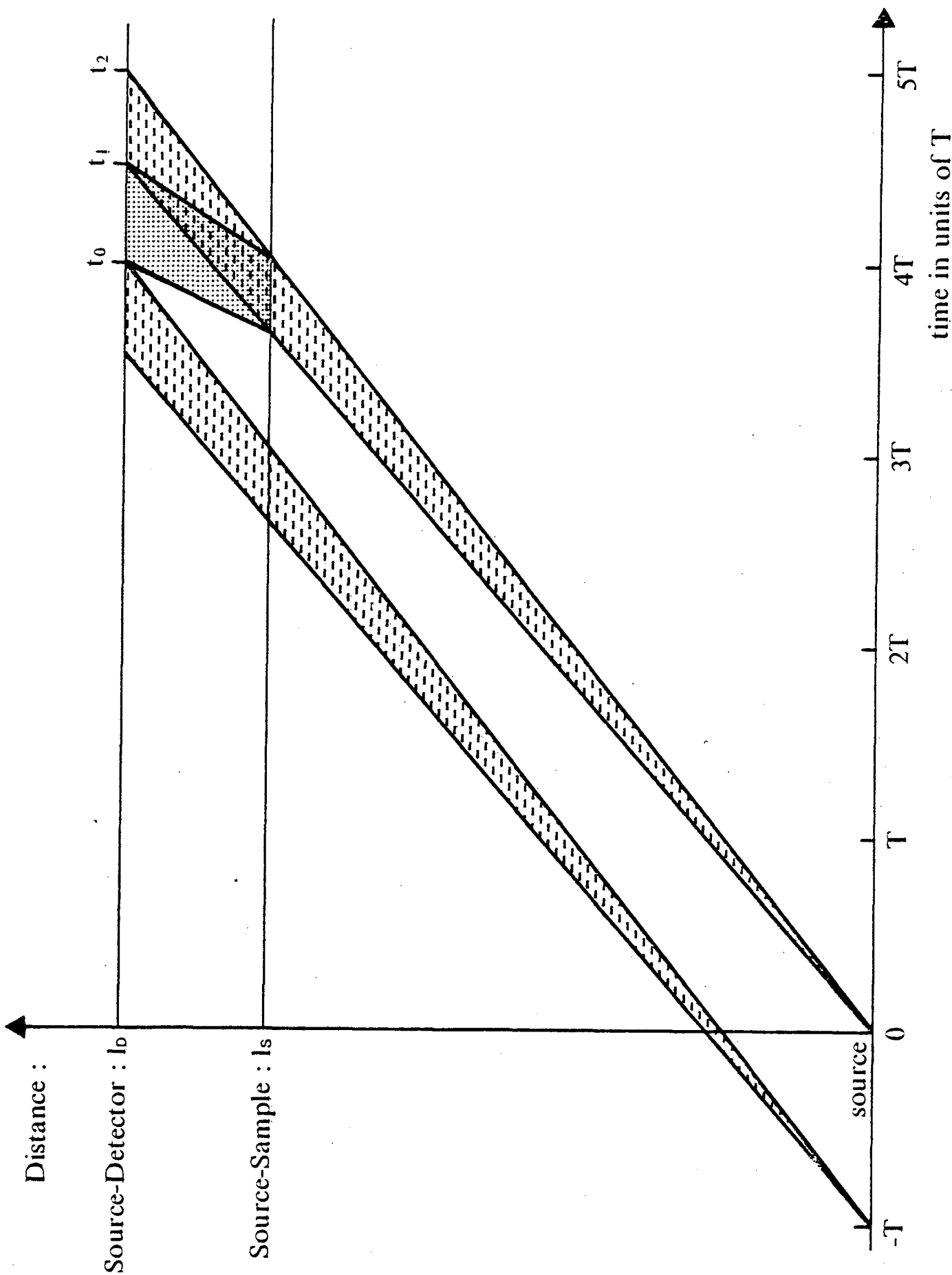


Fig. 11 The General Overlap - Criteria

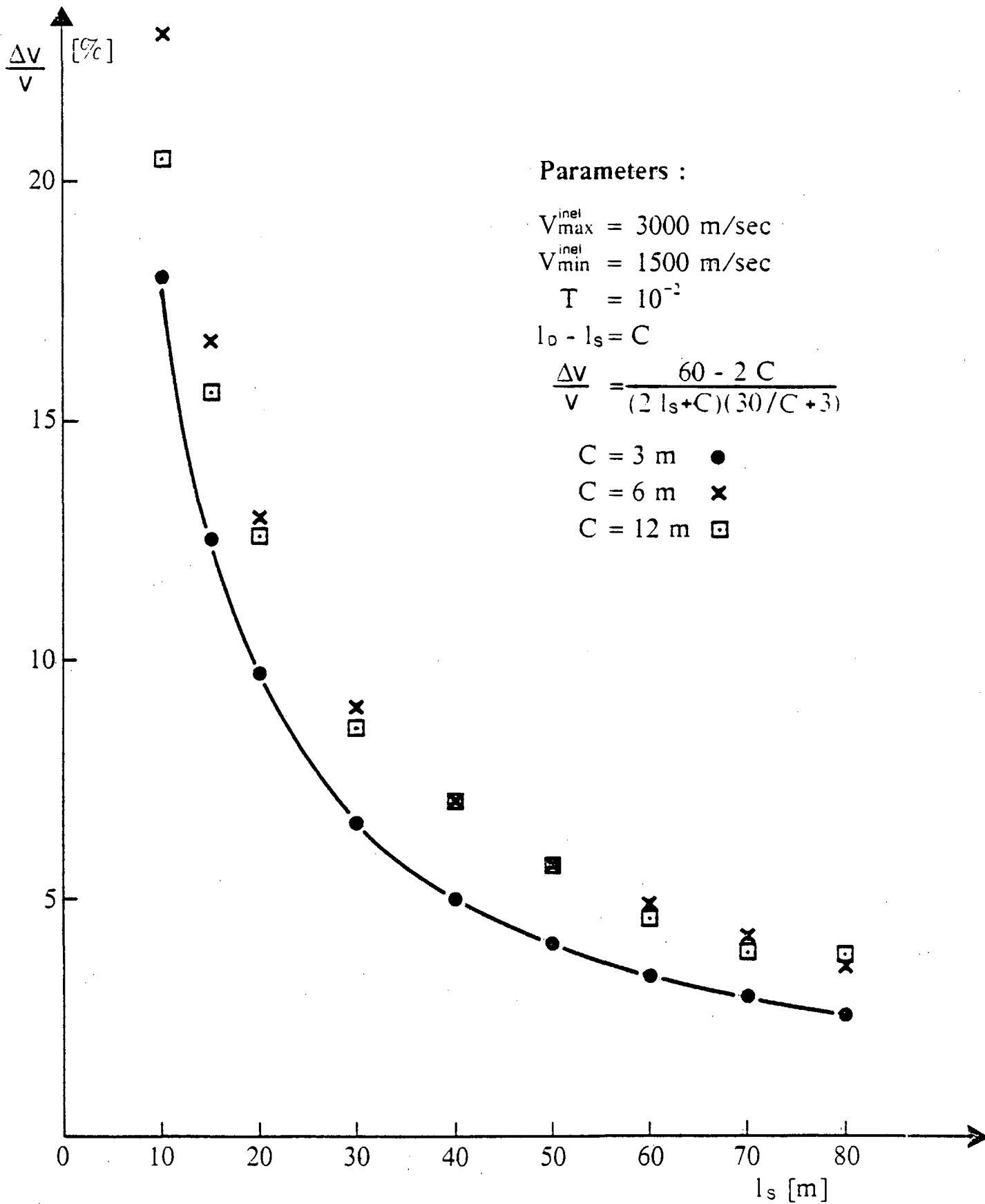


Fig. 12 Admissible Intensity as a function of  $l_s$ , the Distance between Neutron-Source and Sample

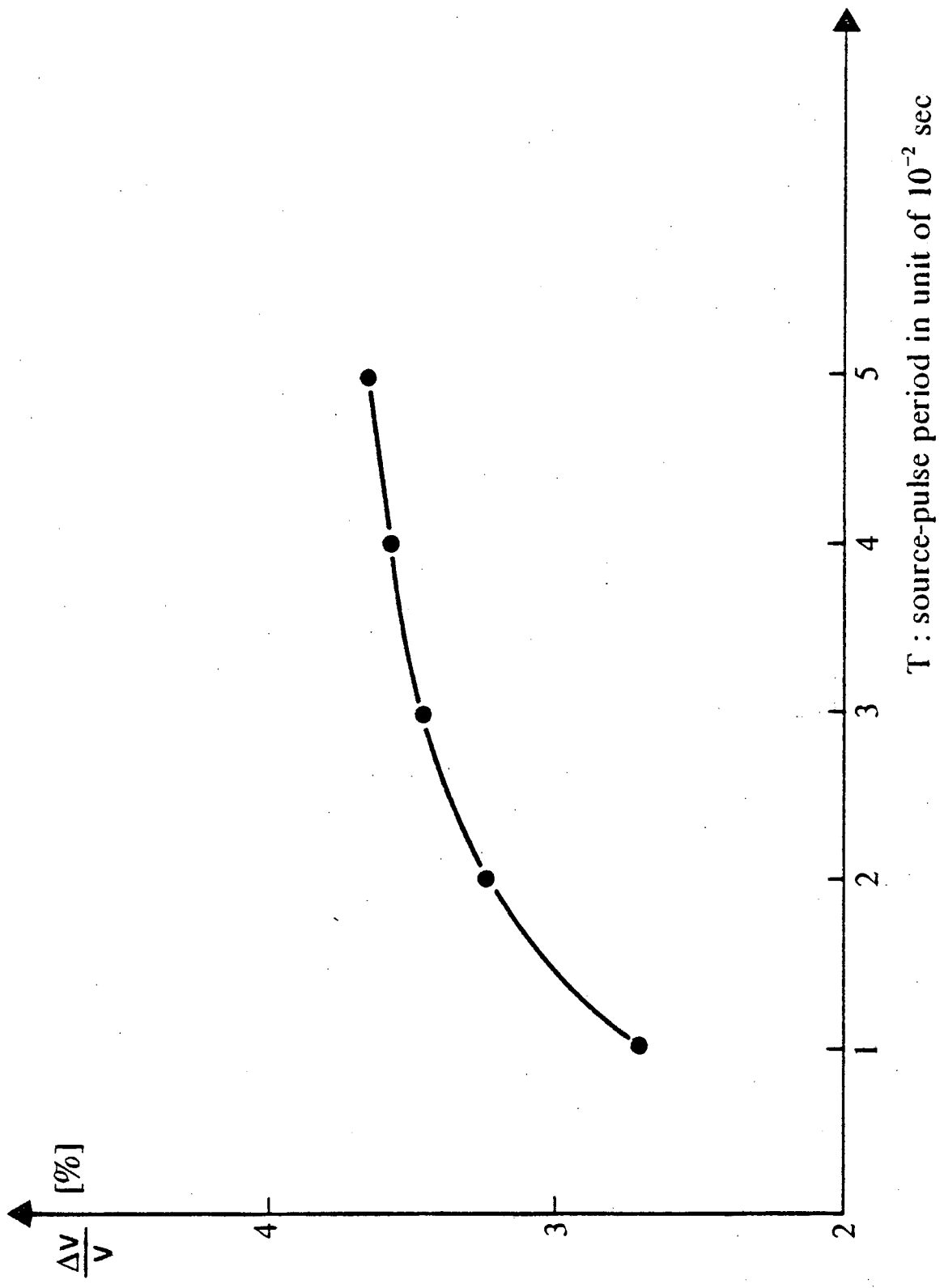


Fig. 13 Admissible intensity as a function of  $T$ , the neutron source pulse period

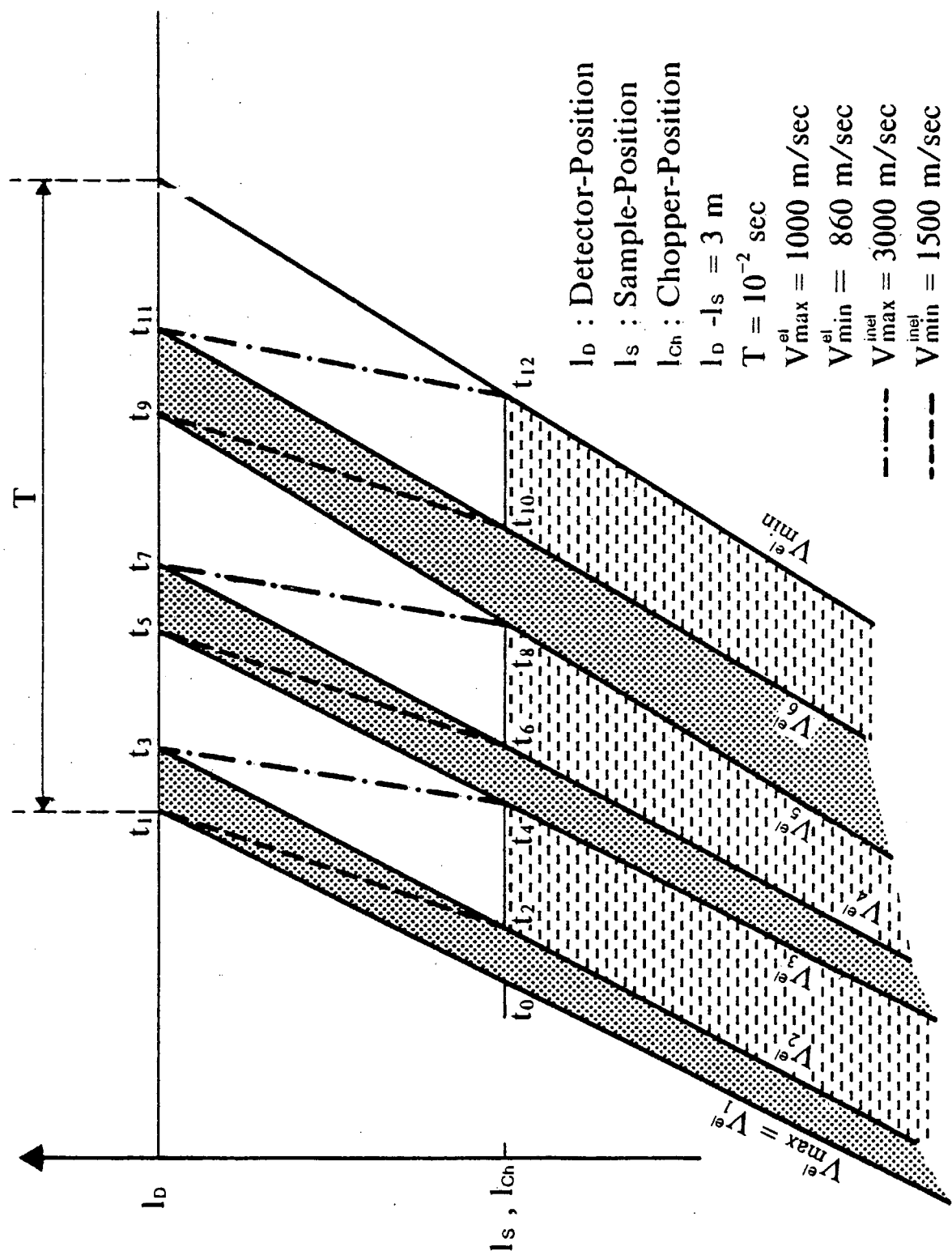
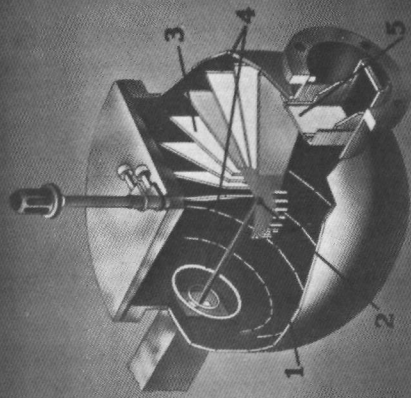
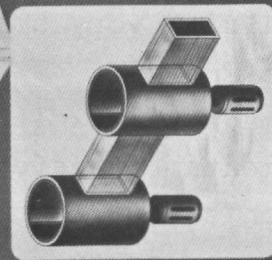


Fig. 14 Chopper Assisted Detector-Gating for pure elastic scattering in the EDS-Spectrometer

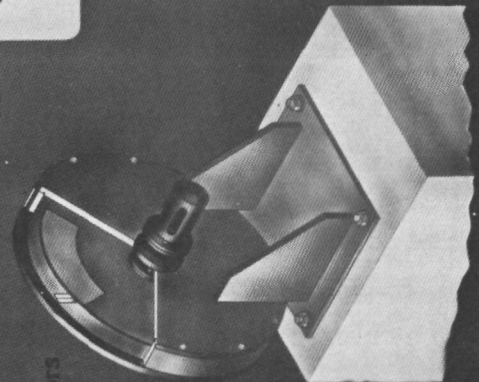
# THE MATERIAL SCIENCE SPECTROMETER



Target Station



Neutron  
Pulse  
Shaping  
Choppers



Overlap Rotors

Background  
Rotors

Target Station

- 1) Analysing Crystals
- 2) Target
- 3) Collimators
- 4) Detectors
- 5) Mobile - Detectors

## A Small Angle Spectrometer for Brillouin Scattering

W. Reichardt

Kernforschungszentrum Karlsruhe

Institut für Nukleare Festkörperphysik

Postfach 3640, D-7500 Karlsruhe, FRG

### ABSTRACT

In this report a small angle spectrometer is described which is intended for measurements of the scattering law at small values of the momentum transfer. It is designed for measurements of energy transfers which are small compared to the incident energy. Excitation energies can be determined with resolutions, which are about 1% of the primary energies. For primary energies around 250 meV the range of momentum transfers is  $0.3 \text{ \AA}^{-1} \lesssim Q \lesssim 1.5 \text{ \AA}^{-1}$ . The main areas of application for this spectrometer are:

1. Determination of the scattering law of liquids and amorphous materials at very small momentum transfers (Brillouin scattering).
2. Measurements of magnetic excitations of moderate energies at very small momentum transfers in order to avoid contamination by phonon scattering.
3. Measurements of high energy magnetic excitations at intermediate momentum transfer.

For problems of the first and second kind primary energies between 200 meV and 400 meV will be used, for problems of the third kind primary energies up to 1 eV are necessary.

It is obvious that this spectrometer requires the compressor ring of stage 2 of the SNQ, but even then the desired resolutions in energy cannot be reached below 300 meV with the present concept of the moderator. More favorable conditions will be obtained by using a cooled moderator which extends the epithermal region to lower energies.



## INTRODUCTION

The future German Spallation Neutron Source SNQ is a strong source of epithermal neutrons. After the installation of the compressor ring the neutron intensity will be concentrated in narrow pulses of a few microseconds. The pulse width is given by  $\Delta t = 2/\sqrt{E}$ , with  $\Delta t$  in  $\mu s$  and the energy  $E$  in eV. Thus, with time-of-flight experiments rather good time resolutions can be achieved. Therefore measurements of the scattering law in the region of very small momentum transfers  $Q$  may be feasible. These experiments are rather demanding, because they require a very good resolution in energy at high neutron energies as well as rather small scattering angles. Furthermore very often the cross-sections for inelastic scattering are small (the one phonon cross-section is proportional to the square of the momentum transfer). In this paper a time-of-flight spectrometer is described, which is specialized mainly on measurements of the scattering laws in the region  $0.3 < Q < 1.5 \text{ \AA}^{-1}$  for moderate energy transfers  $\hbar\omega \leq 30 \text{ meV}$ .

## 2. SOME USEFUL RELATIONS

For small values of  $\hbar\omega/E_i$  energy and momentum conservation lead to the relation

$$\frac{Q^2}{k_i^2} = \frac{1}{4} \left( \frac{\hbar\omega}{E_i} \right)^2 \left( 1 + \frac{1}{2} \frac{\hbar\omega}{E_i} + \frac{5}{16} \left( \frac{\hbar\omega}{E_i} \right)^2 \right) + 4 \sqrt{1 - \hbar\omega/E_i} \sin^2 \phi / 2 \quad (1)$$

$Q, \hbar\omega$ : momentum transfer, energy transfer

$E_i, k_i$ : energy and wave number of the incident neutron

$\phi$  : scattering angle

For small scattering angles and  $\hbar\omega/E_i \ll 1$  this expression reduces to

$$\left(\frac{Q}{k_i}\right)^2 = \frac{1}{4}\left(\frac{\hbar\omega}{E_i}\right)^2 + \phi^2 \quad (2)$$

The smallest primary energy  $E_{i0}$ , which is necessary to arrive at an energy transfer  $\hbar\omega$  for a given  $Q$ -value, is obtained for  $\phi = 0$ , which leads to

$$\frac{E_{i0}}{k_{i0}} = \frac{1}{2} \frac{\hbar\omega}{Q} \quad (3)$$

Using  $E = 2.07 k^2$  ( $E$  in meV,  $k$  in  $\text{\AA}^{-1}$ ) we obtain

$$k_{i0} = 0.240 \frac{\hbar\omega}{Q} \quad \text{and} \quad E_{i0} = 0.120 \left(\frac{\hbar\omega}{Q}\right)^2 \quad (4)$$

The maximum scattering angle  $\phi_M$  results for

$$E_{iM} = 2E_{i0} \quad (5)$$

with

$$\phi_M = \frac{1}{\sqrt{2}} \frac{Q}{k_{iM}} = \frac{1}{2} \frac{Q}{k_{i0}} = 2.07 \frac{Q^2}{\hbar\omega} [Q \text{ in } \text{\AA}^{-1}, \hbar\omega \text{ in meV}] \quad (6)$$

For energies  $E_i = x E_{i0}$  ( $x \geq 1$ )  $\phi(x)$  is given by

$$\phi(x) = 2 \frac{\sqrt{x-1}}{x} \phi_M = \alpha \phi_M \quad (7)$$

The function  $\alpha = 2\sqrt{x-1}/x$  is shown in Fig. 1. It is seen, that already for  $E_i = 1.4 E_{i0}$  the scattering angle has reached about 90 % of its maximum value. Therefore the choice

$$E_i^{\text{opt}} = 1.4 E_{i0} = 1.68 \left(\frac{\hbar\omega}{Q}\right)^2$$

will be a reasonable compromise between the two attempts, to keep  $E_i$  small for a tolerable energy resolution, and to obtain the largest possible scattering angles, in order to get out of the direct beam.

An example will demonstrate the severe condition one meets with in Brillouin scattering experiments. We want to measure an energy transfer of  $\hbar\omega = 12$  meV at a momentum transfer of  $Q = 0.3 \text{ \AA}^{-1}$ . These values correspond to a longitudinal phonon in Aluminium at the reduced wavevector 0.2 of the 100-direction.

For the smallest possible primary energy, the energy of the maximum scattering angle and the optimum energy we obtain:

$$\begin{aligned} E_{i0} &= 192 \text{ meV} \\ E_{iM} &= 384 \text{ meV} \\ E_i^{\text{opt}} &= 290 \text{ meV} \end{aligned}$$

The maximum scattering angle and the angle for  $E_i^{\text{opt}}$  are

$$\begin{aligned} \phi_M &= .0155 = .89^\circ \\ \phi_M^{\text{opt}} &= .0140 = .81^\circ \end{aligned}$$

### 3. SOME CONSIDERATIONS CONCERNING THE LAY-OUT OF THE SPECTROMETER

The example of the proceeding section may serve as a guideline for the construction of the spectrometer. A primary energy of 290 meV is necessary to measure an excitation energy of  $\hbar\omega = 12$  meV at  $Q = 0.3 \text{ \AA}^{-1}$ . The resulting scattering angle was 0.8 degrees. The postulated resolution  $\Delta\hbar\omega/\hbar\omega = 20 \%$  requires  $\Delta E_i/E_i = 0.6 \%$ , if the uncertainties  $\Delta E_i$  and  $\Delta E_f$  are made equal.

For a chopper spectrometer with identical pulse width of the source and the chopper we obtain in the epithermal region (with compressor ring)

$$\frac{\Delta E_i}{E_i} = 2 \frac{\Delta t_i}{t_i} = \frac{\sqrt{2} \cdot 2/\sqrt{E_i}}{72.5 L_1/\sqrt{E_i}} = \frac{0.0774}{L_1} \quad (9)$$

Here  $L_1$  is the distance moderator-chopper in meter. For  $L_1 = 12$  m results

$$\frac{\Delta E_i}{E_i} = 0.65 \%$$

Starting from these considerations we demand the following characteristics of the spectrometer

1. range of primary energies  $E_i > 100$  meV
2. resolution in energy  $\Delta E_i/E_i < 1 \%$  and  $\Delta E_i \approx \Delta E_f$
3. smallest scattering angle  $\phi_{\min} = 0.5^\circ$
4. angular spread in the primary beam  $\alpha_0 = 0.2^\circ$ .

#### 4. DESCRIPTION OF THE SPECTROMETER

A scetch of the instrument, a chopper time-of-flight-spectrometer, is shown in Fig. 2. The distance between moderator and chopper is 12 m. This yields a resolution  $\Delta E_i/E_i = 0.65 \%$  in the epithermal region. Between Fermi-chopper and sample (distance 1.5 m) a coarse collimator can be inserted. This collimator prevents, that neutrons scattered by the chopper may reach the detector. Along its total length the beam channel is confined to a diameter of 5 cm. This results in an angular divergence in the primary beam of  $\alpha_0 = 0.21^\circ$ .

The secondary flight path  $L_2$  was chosen as 12 m. This is considerably longer than the length of  $L_1/\sqrt{2} = 8.5$  m required for  $\Delta E_i = \Delta E_f$ , in order to allow the analysis of energy loss and energy gain scattering and to obtain a larger distance from the direct beam for detectors at small scattering angles. It should be mentioned, that the extension of  $L_2$  does not change the intensity per solid angle. The price one has to pay for the improved secondary energy resolution is the costs for the detectors, which increase as  $L_2^2$ , if the range of scattering angles is kept constant.

The scattered neutrons are record in an angular area detector with an outer diameter of 3.4 m. The range of scattering angles is  $0.5^\circ < \phi < 8^\circ$ . The upper limit of the scattering angle was determined by the requirement that a momentum transfer of  $1.5 \text{ \AA}^{-1}$  should be reached for primary energies around 250 meV.  $1.5 \text{ \AA}^{-1}$  corresponds to the border of the Brillouin zone for many substances.

It is not reasonable to extend the range of scattering angles of the spectrometer in order to measure the scattering law at larger Q-values at the same time. In this region it is no longer necessary that  $\hbar\omega \ll E_i$ . Therefore those measurements can be performed under more favorable conditions concerning resolution and intensity using conventional time-of-flight spectrometers with multidetectors. Furthermore this spectrometer requires special conditions for the geometry of the sample: In order to exclude differences in flight distances, the sample has to be a flat disc, which is placed perpendicular to the primary neutron beam. The strong background at small angles due to multiple scattering must be reduced by subdividing the sample in two directions by neutron absorbing lamellas. This excludes measurements at large scattering angles.

The area detector should have a radial resolution of about 2.5 cm. In order to obtain a time resolution of a few permille, uncertainties in pathlength in the detector should be of the order of 1 cm. Therefore, probably scintillation counters with Li glass have to be used. The inner part of the detector ( $\approx 50$  cm  $\varnothing$ ) should be easily removable. This will allow modifications of shielding and detectors close to the primary beam according to the requirement of the specific experiment.

The most serious problems of the spectrometer is certainly the background at small scattering angles. As can be seen from Fig. 3 the lower limit of the scattering angle lies outside of the directbeam, that is, the inner segments of the detector have no view on the moderator. In case of too high background the cross-section of the beam may be reduced by inserting a tube into the beam channel. An alternative solution is the use of a girder with a cross-like section, which reduces the angular divergence (see Fig. 3).

Several Fermi-choppers have to be used which cover pulse widths between 2  $\mu$ s and 6  $\mu$ s. For suppressing parasitic energies (the Fermi-chopper opens several times between two moderator pulses) and for background reduction several background choppers are necessary.

An approximate estimation of the neutron intensity at the sample position may be obtained by starting from data for the British pulsed source SNS:

$$(I\Delta E)_{\text{SNS}} = \frac{8.8 \cdot 10^{12}}{L_1^2} \frac{\Delta E}{E} [\text{ncm}^{-2} \text{sec}^{-1}] \quad (10)$$

[distance L in cm]

This expression applies for a moderator surface of 100 cm<sup>2</sup>. If we take into account, that the proton beam of SNQ is 25 times more intense and that the beam cross-section of the spectrometer under consideration is smaller by a factor of 5, we arrive at

$$(I\Delta E)_{KWSP} = \frac{25}{5} \cdot \frac{8.8 \cdot 10^{12}}{1350^2} \cdot 0.0065 \cdot \frac{1}{2} = 8 \cdot 10^4 \text{ [ncm}^{-2}\text{sec}^{-1}\text{]}$$

The factor 1/2 stands for the transmission of the chopper.

For the example discussed in section 2 ( $\hbar\omega = 12 \text{ meV}$ ,  $E_i = 290 \text{ meV}$ ) we obtain the following performance of the spectrometer

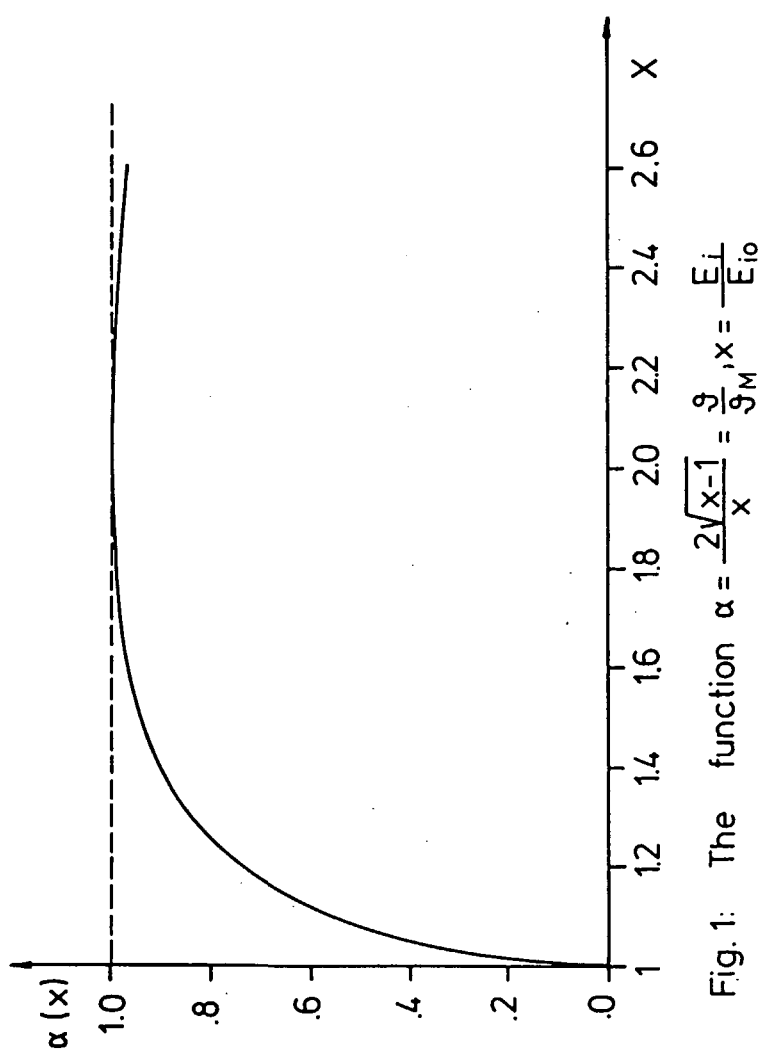
$$\Delta t_{\text{chopper}} = 3.7 \text{ } \mu\text{s}$$

$$\frac{\Delta E_i}{E_i} = 0.0065 \quad \Delta E_i = 1.88 \text{ meV}$$

energy gain:  $\Delta E_f = 1.42 \text{ meV}$  ,  $\Delta \hbar\omega = 2.36 \text{ meV}$  ,  $\Delta \hbar\omega / \hbar\omega = 20 \%$

energy loss:  $\Delta E_f = 1.25 \text{ meV}$  ,  $\Delta \hbar\omega = 2.26 \text{ meV}$  ,  $\Delta \hbar\omega / \hbar\omega = 19 \%$

The energy resolution of the instrument may be improved by enlarging the flight path. Then the intensity at the detector per scattering angle drops with the square of the distance between the moderator and the sample. Increasing  $L_2$  leaves the intensity unchanged. Therefore  $\Delta \hbar\omega$  is approximately proportional to the square of the intensity at the detector. This has to be compared with time-of-flight spectrometers at stationary sources, where  $\Delta \hbar\omega$  is proportional to the third power of the intensity. This difference is caused by the fact, that at high neutron energies the flight paths can be increased without reducing the repetition rate, as there are no overlap problems. Typical flight times are 2 ms for 12 m, whereas the separation of the pulses is 10 ms.





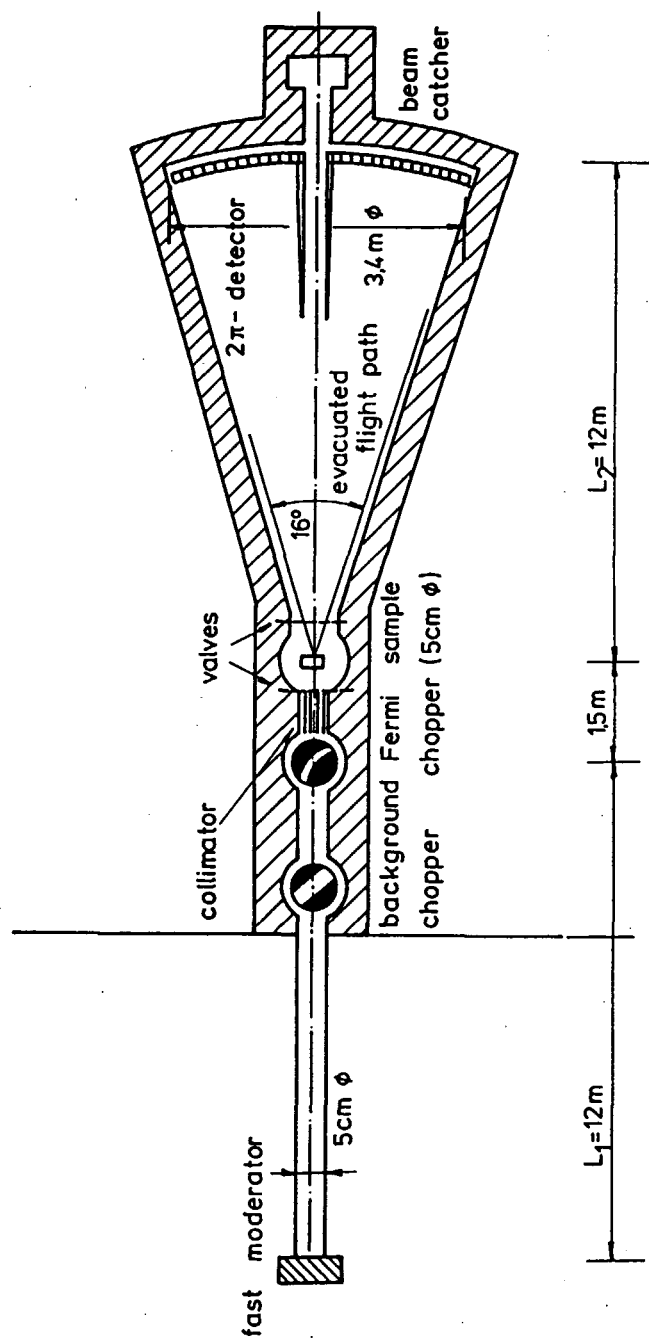
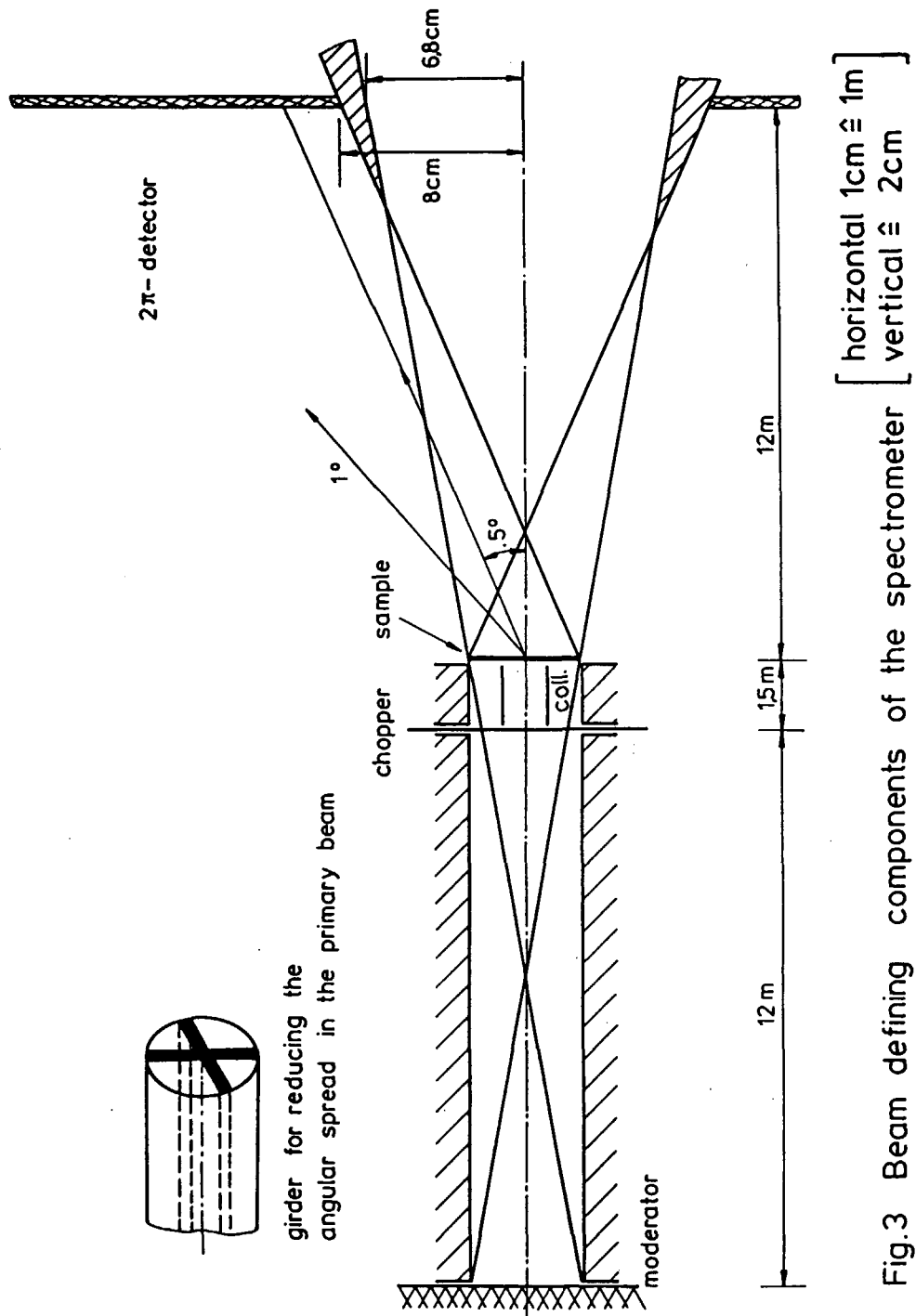


Fig.2. Lay-out of the small angle spectrometer (not to scale)



## CORRELATION AND FOURIER TIME-OF-FLIGHT SPECTROSCOPY AT A PULSED NEUTRON SOURCE

Gerald Badurek

Institut für Kernphysik, Technische Universität Wien, Austria

### 1. Introduction

Some 15 years ago two techniques - the pseudostatistical correlation method /1-7/ and the so-called Fourier method /8-11/ - have been proposed to increase the efficiency of neutron time-of-flight (TOF) scattering experiments. Whereas standard TOF spectroscopy at continuous neutron sources suffers from the low duty cycle of typically 1% or less of the necessary chopper devices both the correlation and the Fourier approach offer the possibility to reach duty cycles of up to 50% without any loss in time resolution. Although both techniques are very similar in many respects it turns out that the pseudostatistical correlation method is most favourably applied for electronic spinflip chopping of polarized neutron beams /12-14/ whereas with mechanical neutron choppers the Fourier technique in general has the advantage of much more flexibility and, most important if adequately sized samples are available, offers an inherent decoupling of the effective beam cross-section and the achievable time resolution /11-15/.

But in spite of this neither of both methods has found large scale application in experimental slow neutron spectroscopy until now. The only exception is the use of correlation techniques in polarized neutron scattering where one benefits from the great flexibility in generating suitable pseudorandom binary sequences (PRMS) by simple electronic means. For that particular case it is furthermore of importance that there is always a considerable "background" signal present due to the intrinsically imperfect "off" transmission characteristics of such spinflip choppers. It is exactly this back-

ground which favours non-conventional TOF methods as will be discussed later on. Two fundamental reasons are responsible, however, that both the correlation and the Fourier technique have been so largely ignored for all other cases of neutron TOF spectroscopy. First of all, both methods do not directly yield the quantity to be measured but afford either cross-correlation or Fourier transformation procedures to obtain the neutron TOF distribution. Thus there are appreciably more possible sources of systematic errors than with the direct TOF method which need careful precautions to keep them below significance. The second important reason is that the effective gain of statistical accuracy is not simply proportional to the increased duty cycle of a statistical or a Fourier chopper. One finds, instead, that for the Fourier chopper and for statistical choppers with duty cycle 50% only those portions of a TOF spectrum can be measured within given time with better statistical precision as compared to the conventional direct method which lie above the average of that particular TOF distribution /11,15/. This gain depends therefore strongly on the actual shape of the TOF distribution and on the height of the uniformly distributed background intensity. For other duty cycles than 50% things get more complicated, with the consequence that the achievable gain can be optimized in principle by using correlation sequences with appropriately chosen duty cycle. However, with mechanical choppers this is difficult to do in practice since it affords to replace the chopper disk rotor by another one. Furthermore, such an optimization procedure can evidently be done only if the shape of the TOF distribution is either roughly known from previous experiments or can be estimated with some confidence.

But in spite of lacking profound experimental realization a lot of theoretical work has been done for the correlation method to exploit all its features if pulsed neutron sources instead of continuous ones are to be used and if multi-variable correlation procedures are applied to measure inelastic scattering effects with a white incident beam, similar to the conventional double chopper TOF technique. Presumably the most important contributions to this field which actually cover all possible experimental

situations were given by Hoßfeld and Amadori /16-18/ in 1973 already. It would be by far beyond the scope of this short report to summarize all their results. Somewhat subjectively, of course, I will pick out therefore only those few cases which are of actual relevance for the SNQ application.

## 2. Correlation Method

An essential criterion for the judgement whether the correlation method is of advantage at a pulsed neutron source compared to a conventional TOF determination is the duration of the source pulse itself. Depending on whether it is short enough for the required TOF resolution at given length of the flight paths or not, one has to consider two situations:

### a) Primary source pulse sufficiently short

It is trivial that in that particular case the insertion of any type of chopper would be nonsense as long as one works with monochromatic incident neutrons. But with a white beam a correlation chopper placed near the sample which runs asynchronously with the source pulses (i.e. its repetition frequency is coprime to that of the source) yields a degenerate version of a statistical double-chopper spectrometer with a duty cycle of the first chopper  $c_1 \sim 0$  (Figure 1). Defining the gain factor as usual by the ratio of the mean square statistical errors of a conventional double-chopper experiment and that particular correlation technique the gain factor is given approximately as /17/

$$G^2 \equiv \frac{\sigma_{\text{konv}}^2}{\sigma_{\text{korr}}^2} \sim \frac{c_2(1-c_2)N}{1-2c_2 + [\sigma_{ij} + \beta_{ij}]^{-1} c_2(2\beta_{ij} + N)} \quad (1)$$

where  $N \gg 1$  is the length of the pseudorandom binary sequence and  $c_2$  the duty cycle of the correlation chopper and where  $\sigma_{ij} \equiv S_{ij}/\bar{S}_{ij}$  and  $\beta_{ij} \equiv b/\bar{S}_{ij}$  are defined as the relative spectrum height and background values. This gain factor is also shown graphically in Figure 2.  $S_{ij}$  is the discretized two-dimensional time-of-flight

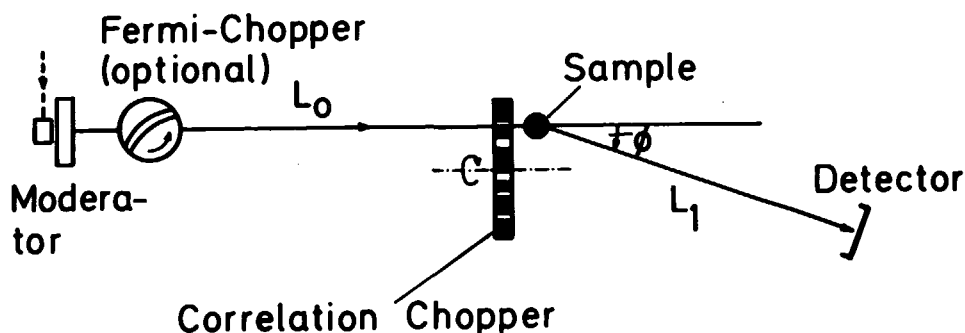


Figure 1: Double chopper neutron time-of-flight spectrometer with a pseudorandom correlation chopper at a pulsed source. The source itself acts as a conventional chopper (duty cycle  $c_1 \sim 0.01$ ) which allows to determine the total time-of-flight along the path  $L_0 + L_1$ . Optionally a Fermi chopper rotating in phase with the source can be used to shorten the source pulse length and to improve the resolution. By means of a two-dimensional analyzer system the response of the detector to the modulation pattern of the pseudorandom chopper is stored separately for each total time-of-flight. From this set of data the neutron time-of-flights along the distance  $L_1$  can be found by cross-correlation of the detector signals and the chopper modulation function.

distribution one would obtain with a conventional double-chopper system in a series of successive experiments in equal total measuring time and where  $i$  and  $j$  refer to the neutron flight times from the source to the sample and from the sample to the detector, respectively. The averaging over the spectrum in defining  $\sigma$  and  $\beta$  is meant there only as the "local" average over a section through the 2-dimensional time-space for which  $i + j = \text{const}$ , i.e. for fixed total time-of-flight. Similar to the univariable correlation technique at continuous sources this gain factor can be larger than unity if either the background is high (naturally only the time-independent background is denoted by  $b$ , any background contribution that is modulated periodically in time according to the source luminosity variation has to be included into the time-of-flight distribution itself) or for those parts of the TOF spectrum which exceed its mean value (see Figure 2). Hence the usefulness of the correlation method just described depends essentially on how well structured the TOF spectrum is. The

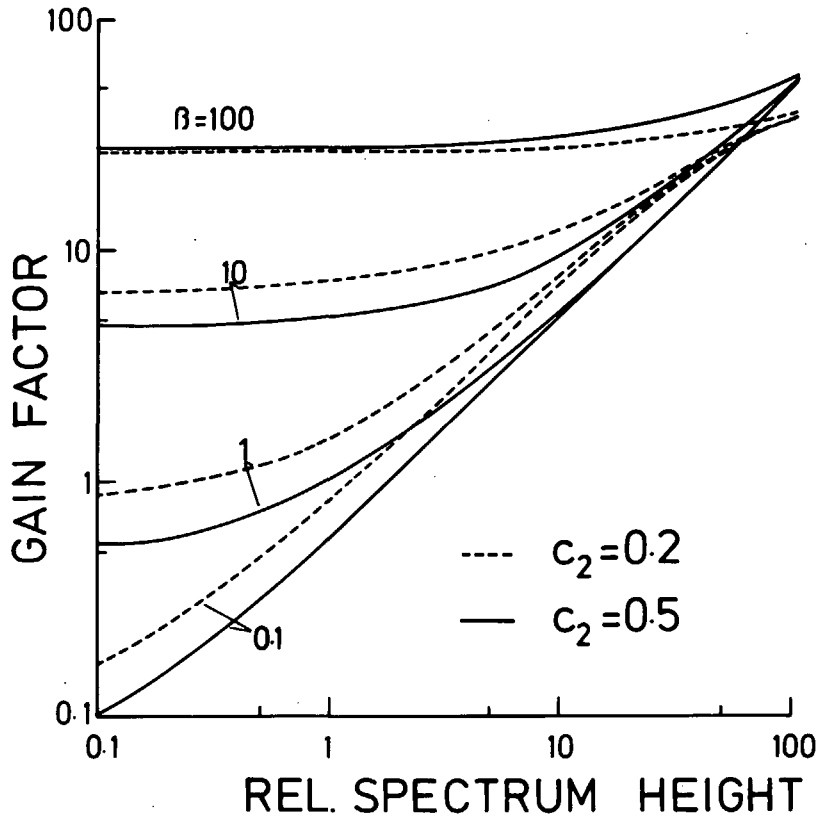


Figure 2: The gain factor of a correlation experiment for two different duty cycles  $c_2$  of the pseudorandom chopper. Parameters are the relative spectrum height  $\sigma$  and the relative background  $\beta$  as defined in the text. A pseudorandom sequence length  $N = 255$  has been assumed. For the duty cycle  $c_2 = 0.5$  the correlation chopper gain factor is approximately equal to that of an equivalent Fourier chopper.

presence of large broad peaks will strongly deteriorate the accuracy of the lower lying parts of the spectrum, exactly as it is the case with the well-known univariable method at continuous neutron sources. Due to the local averaging over the individual subspectras for well-structured two-dimensional TOF distributions it is likely that dominant peaks govern a few of the subspectras only whereas for the remaining ones the local gain factor (1) can reach appreciable values.

b) Source pulse has to be shortened

The length of the neutron pulses of the SNQ (in its LINAC version) will be about 300  $\mu$ s in the thermal wavelength region. This is too long for reasonable TOF applications and hence the pulse has to be shortened. (This is not necessarily so if later on the

neutron pulse length will be reduced to about 120  $\mu$ s by using a proton pulse compressor ring at the accelerator stage of the SNQ /19/). As indicated in Figure 1 the simplest way to shorten the pulse is to use a phased Fermi chopper placed near the source or at some distance from it, thereby defining the wavelength band which will be transmitted. Aside of a mere reduction of intensity the situation is then identical to that described in a) and needs hence not to be treated further.

But for monochromatic incident neutrons there is a very elegant and promising way to use the correlation technique to increase the TOF resolution without losing too much intensity. The idea is simple and applicable both for direct geometry X-TOF (Figure 3a) and inverted geometry TOF-X (Figure 3b) spectrometer arrangements.

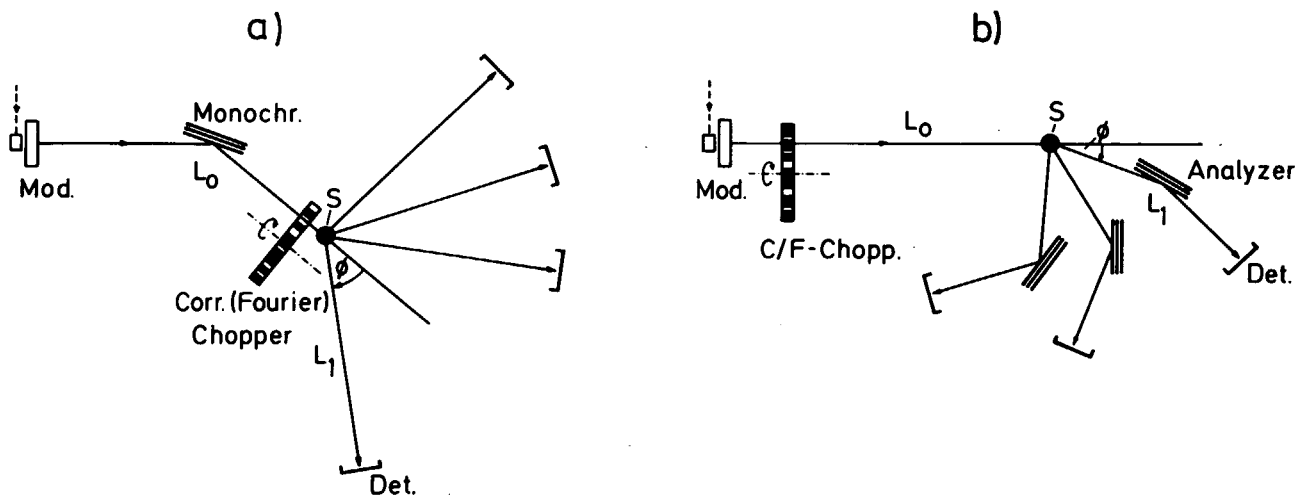


Figure 3: Univariable correlation (or Fourier) spectrometer at a pulsed source with fixed incident (a) or scattered (b) neutron wavelength. The source acts as a conventional chopper of a host spectrometer with coarse time resolution.

The source pulses are asynchronously modulated according to a pseudorandom sequence by means of a statistical disk chopper, but in contrast to continuous sources one can use there the broad initial source pulse for a crude TOF separation of neutrons that were scattered with different energy transfers. Then strong peaks in the TOF distribution do not longer disturb the accuracy of weak ones since the cross-correlation of the recorded intensity with the chopper sequence can be done separately for each time region.



Since, as indicated in Figure 4, for the weak lines (i.e. the inelastic parts of the TOF spectrum) the "locally" averaged value of the TOF spectrum is essential for the relative signal

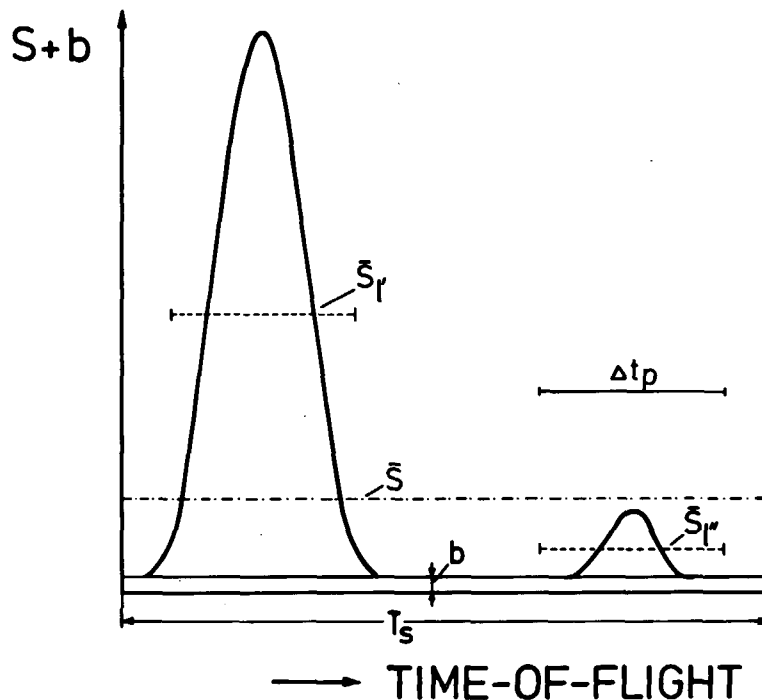


Figure 4: On the definition of local and overall averaging of a hypothetical time-of-flight spectrum  $S$  consisting of a large and a small peak.  $b$  is the time-independent background and  $T_s$  the spectrum length. In an ordinary correlation experiment the small peak would be lower than the mean value  $\bar{S}$  of the spectrum and could therefore not be measured with reasonable statistical accuracy. At the pulsed source only the "local" averages  $\bar{S}_1'$ ,  $\bar{S}_1$  over a time interval  $\Delta t_p$ , which is given by the duration of the primary source pulses, is relevant for the accuracy. This leads to larger gain factors for the low portions of a spectrum. The size of the gain factor depends, however, quite critically both on the shape of the source pulses and the actual form of the neutron time-of-flight distribution /17/.

height, the local gain factors will be appreciably much larger than the corresponding "gain" at continuous source experiments, where weak lines will in general be measured with less accuracy than with conventional choppers in case a strong (usually elastic) line is present.

This method is similar in some respects to the digital filtering techniques developed for continuous source correlation methods /12,18/. There a second statistical chopper, which is asynchronous to the first one, is used in combination with properly delayed gating of the detector to suppress definitely all those neutrons which could have been scattered elastically. But since part of the inelastically scattered neutrons are also not counted and since the insertion of a second chopper causes a further decrease of the duty cycle these filter techniques have been of not much practical use. In most cases they are not worth the effort necessary and frequently they even worsen the achievable accuracy. Essentially this statement seems to be valid also for the statistical double-chopper filter technique at pulsed sources which was implemented by Cser et al. /20/ at the Dubna IBR-30 pulsed reactor. Though these authors could show that their method works in principle their presented results are rather inadequate to allow for a critical verification of any gain in statistical precision of the measured neutron spectrum. The direct TOF filtering, possible at pulsed neutron sources, on the other hand presumably will make the correlation time-of-flight spectroscopy the powerful technique it was intended to be at its very beginnings.

### 3. Fourier Spectroscopy

As mentioned already, Fourier time-of-flight spectroscopy is the other approach that could be followed at a pulsed neutron source. To estimate whether this would be feasible it is necessary to recapitulate the functional principle of a (mechanical) neutron Fourier chopper. Without going into any detail such a chopper consists of a rotor disk with a large number of uniformly distributed neutron absorbing teeth and transparent bridges of equal width and a fixed stator with exactly the same absorption pattern. By this means the neutron beam can be modulated periodically in time at a series of discrete equidistant frequencies  $\omega_k = k\Delta\omega$  ( $k = 1 \dots K$ ) if we neglect for simplicity the odd higher harmonic frequency contributions of the triangular beam modulation that is achieved with such a rotor-stator combination. By means

of a phase-sensitive neutron detection system one measures then for each frequency separately the amplitudes and phases of the detector response oscillations relative to the impressed modulation of the chopper. From these measured Fourier coefficients the time-of-flight spectrum in turn can be calculated by means of discrete Fourier transformation. The duty cycle of the Fourier chopper is  $c_F = 50\%$ , the statistical errors of each point of the TOF spectrum are equal and about the same as those obtained by means of an equivalent correlation chopper experiment /15/.

In spite of its fixed duty cycle and the fact that only half of the beam area is transparent the mechanical Fourier chopper offers some distinct advantages over the mechanical correlation chopper, at least at continuous sources.

First of all, the achievable time resolution and the usable effective beam cross-section are totally independent from each other. All one needs to do to increase the time resolution for given maximum rotational speed of the chopper disk and given beam size is to make the slit pattern as narrow as possible by technological standards. Modulation frequencies of up to 500 kHz corresponding to a time resolution of about 2  $\mu$ s seem to be absolutely feasible.

At a first glance one might believe that one cannot observe the TOF flight spectrum during the run of a Fourier experiment. This is in fact not true since at any moment one can yet perform a Fourier transformation of the set of Fourier coefficients which have already been measured without interrupting the running experiment. This is even an advantage in comparison both with conventional and correlation TOF experiments. With these methods one has to repeat a measurement with changed resolution parameters in case the resulting time resolution was found to be insufficient. With the Fourier method, however, one needs not to repeat the whole measurement but can easily append further coefficients at higher frequencies to the already existing set before an anewed Fourier transformation can take place.

For completeness it should be noticed that apart of the ordinary Fourier method a so-called "reverse" Fourier TOF method /15,21,22/

has been developed, where the chopper speed is swept through continuously between the lowest and the highest frequencies and the Fourier transformation is performed on-line by means of dedicated hardware configurations. But a serious flaw of that method is its considerable amount of dead-time which stems against its application at intense neutron sources.

A further advantage of the Fourier chopper relies on the fact that the periodicity interval of the Fourier synthesized TOF spectrum, which is given as  $2\pi/\Delta\omega$ , can easily be adjusted to the specific requirements of each particular experiment by choosing a proper value of  $\Delta\omega$ . With a mechanical statistical chopper the sequence length is constant and therefore a similar adjustment of the periodicity interval to the length of the TOF spectrum could be achieved only by means of variation of the chopper speed, i.e. by changing the time resolution.

A particularly unique advantage of the Fourier technique is its inherent possibility to concentrate one's attention to certain regions in frequency space, if necessary. For instance, one can easily separate sharp lines from broader distributions since they belong to different frequency regions. Or if one wants to study the variation of the line width of a peak in time space as a function of some parameter as e.g. temperature, one needs not to reconstruct the whole TOF spectrum. Using a model assumption of the line shape it suffices to follow the temperature variation of only very few frequency components in that case, saving thereby a lot of measuring time. The Fourier chopper approach should be obviously the best choice also if one wants to perform line shape analysis of TOF spectra. Since with other TOF techniques one has to Fourier transform these spectra for a proper analysis, one benefits from the fact that a Fourier spectrometer yields data directly in frequency space and one can concentrate to particular details without necessarily measuring over a large frequency region.

Disregarding the just mentioned advantages of the Fourier technique it is known for continuous source applications that for given beam cross-section, time resolution and total measuring time the Fourier chopper yields roughly the same statistical accuracy than the

correlation chopper. It seems to be likely that for pulsed neutron sources the same argumentation holds that was given before for the correlation chopper case. In particular the univariable case with monoenergetic primary beams should be most favourable again, where the time structure of the source can be used as a low resolution host time-of-flight spectrometer with consequently much increased statistical accuracy of the individual local subspectra.

There is one serious technical problem to be overcome, however, if a Fourier chopper shall work properly at a pulsed facility. A phase-sensitive neutron detection system can be realized by subdividing each modulation period of the Fourier chopper into a number (usually 4) of synchronous time channels of equal width. From the contents of these channels estimates of the amplitude and phase of the neutron intensity oscillations at the detector can be derived, provided all time channels are actually equivalent with respect to the incoming intensity /10/. On pulsed sources it will to my opinion be very difficult to fulfil this requirement by proper asynchronism of the chopper and the pulsed source. In particular at low chopper speeds beating effects may occur which lead to systematic errors in determining the Fourier amplitudes and phases. Although the Fourier chopper data acquisition system is particularly easy to realize at continuous sources even for multi-detector arrangements /23/, there will presumably highly sophisticated precautions be necessary to overcome the technical difficulties associated at pulsed neutron facilities.

#### 4. Concluding Comments

The essential question whether the application of correlation or Fourier time-of-flight spectroscopy is of advantage at a pulsed source can be answered with few summarizing statements.

- i) The correlation and the Fourier method are highly equivalent with respect to count rate statistics. However, for mechanical choppers the Fourier technique should be by far the better choice due to its better resolution/beam cross-section per-

formance and its greater flexibility, provided it is possible to avoid systematic errors in the phase-sensitive neutron detection system because of the time structure of the source. With electronic spinflip choppers for polarized neutrons on the other hand the correlation method is well established already and should be preferred therefore.

- ii) Multivariable correlation spectrometers (or equivalent double-Fourier-chopper arrangements) with polychromatic neutron beams would imply an enormous amount of neutron collection and data processing electronics, but it is highly questionable whether they can yield an accuracy gain apart of those few cases with well-structured two-dimensional neutron time-of-flight distributions.
- iii) If the source pulses are too long to allow for a sufficiently high time resolution at reasonable flight path lengths the univariable correlation, respectively Fourier technique with fixed incident (or scattered) neutron energy allows to achieve a considerable accuracy gain as compared to a "conventionally" performed direct TOF experiment of equal duration. Unlike to the situation at continuous neutron sources large dominant peaks contained in the neutron TOF distributions do not longer deteriorate the statistical precision of the lower lying sections of the scattered neutron spectrum because of the possibility of crude direct TOF separation of its different parts.

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ON THE EXPERIMENTAL DETERMINATION OF HYDROGEN MOMENTUM DISTRIBUTIONS  
AT SOURCES PROVIDING HIGH ENERGY NEUTRONS

H. Grimm

Kernforschungsanlage Jülich GmbH  
D-5170 Jülich, P.O. Box 1913

Abstract

With the increased usable flux at high neutron energies of pulsed sources the measurement of momentum distributions enters a new range of feasibility and accuracy. It is shown that for the case of hydrogen which has many applications an additional gain is to be expected due to the insensitivity to monochromization. The optimization of the analyzer is discussed and a test experiment is proposed.



## EXPERIMENTAL BOUNDARY CONDITIONS

For sufficiently large momentum transfers the impulse approximation /1-4/

$$S(\bar{Q}, \omega) = \int \delta(\omega - \frac{Q^2}{2\mu} - \frac{\bar{Q}\bar{p}}{\mu}) f(\bar{p}) d\bar{p} \quad (1)$$

should become valid, i.e. only a "snapshot" of the momentum distribution  $f(\bar{p})$  of the scattering particles (mass  $\mu$ ) is transferred to the scattered particles which experience the momentum- and energy transfer  $\bar{Q}, \omega$ . The scales for measuring the validity of this approximation are the localization and binding energies of the scatterers.

Specializing now to the case neutron-proton scattering and a Gaussian momentum distribution of the proton means:

$$\mu = 1$$

$$f(\bar{p}) = \pi^{-3/2} |\bar{M}|^{1/2} \exp(-\bar{p} \bar{M} \bar{p})$$

Then the scattering function becomes:

$$S(\bar{Q}, \omega) = \pi^{-1/2} (\bar{Q} \bar{M}^{-1} \bar{Q})^{-1/2} \exp \left[ - (\omega - Q^2/2) / \bar{Q} \bar{M}^{-1} \bar{Q} \right] \quad (2)$$

The exponent is merely a function of the direction of  $\bar{Q}$  and the final wave vector  $\bar{k}_f$  of the neutron as can be seen by inserting the energy- and momentum conservation.

$$\omega = \frac{1}{2} \bar{Q} (\bar{k}_i + \bar{k}_f)$$

$$\frac{1}{2} Q^2 = \frac{1}{2} \bar{Q} (\bar{k}_i - \bar{k}_f)$$

Defining  $\bar{Q} = Q\bar{n}$ , one has:

$$S(\bar{Q}, \omega) = \pi^{-1/2} Q^{-1} (\bar{n} \bar{M}^{-1} \bar{n})^{-1/2} \exp \left[ -(\bar{n} \bar{k}_f)^2 / \bar{n} \bar{M}^{-1} \bar{n} \right] \quad (3)$$

The selection of  $\bar{k}_f$  is crucial for the experiment. In order to obtain informations about  $\bar{M}$ ,  $\bar{k}_f$  has to be in the order of  $(\bar{n} \bar{M}^{-1} \bar{n})^{1/2}$ . Note that this quantity means the extension of the ellipsoid  $\bar{p} \bar{M} \bar{p} = \text{const.}$  in the direction of  $\bar{n}$  as sketched below.

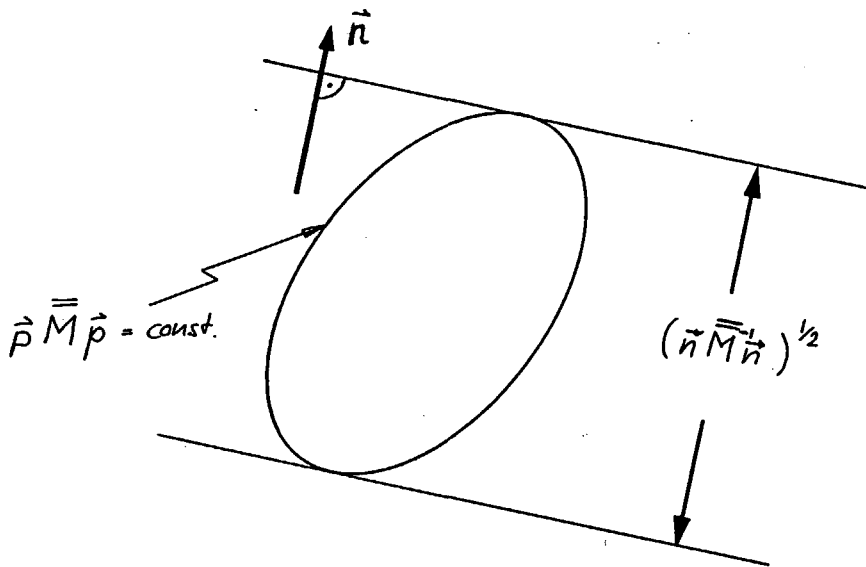


Fig. 1

$\bar{M}$  can be estimated from the knowledge about the Debye-Waller-factor (DW).

Taking e.g. the ground state wave function of an harmonic oscillator

$$\psi \sim \exp \left( - \frac{x^2}{4u^2} \right)$$

one has

$$DW = \left| \mathcal{F} \psi \psi^* \right|^2 \sim \exp(-u^2 Q^2)$$

$$f(p) = \left| \mathcal{F} \psi \right|^2 \sim \exp(-2u^2 p^2)$$

This means that the DW-factor is  $\sqrt{2}$  as broad as  $f(Q)$ . Thus one reaches the  $1/e$ -region of  $S(\bar{Q}, \omega)$  for  $k_f \approx (u \sqrt{2})^{-1}$  where  $u^2$  is the mean squared displacement of the proton.

On the other hand  $k_i$  should be very large in order to reach the necessary momentum transfers for the impulse approximation. G. Reiter /4/ has discussed this range of validity for various examples. A ratio of  $k_i/k_f \geq 10$  is quoted to be sufficient. This ratio restricts  $\bar{n}$  to a cone of  $\leq 6^\circ$  around the  $\bar{k}_i$ -direction and the exponent of equ. (3) becomes only weakly dependent on  $\bar{k}_i$ . Note that this insensitivity of the scattering function is a consequence of the nearly equal masses of neutron and proton. It gives the chance to construct a dedicated instrument for the determination of hydrogen momentum distributions. From the shape of equ. (3) it is obvious that the "monochromator" of such an instrument degenerates to a time gate of the counters. Moreover the analyzer can sum large solid angles easily since  $\bar{n}\bar{k}_f = \text{const.}$  is just the Bragg condition. The arrangement of the analyzer should reflect the symmetry ( $\infty/\text{mm}$ ) of equ. (3) for  $\bar{n} // \bar{k}_i$ , i.e. the cylindrical symmetry around  $\bar{k}_i$  and the mirror plane perpendicular to it.

#### EXPERIMENTAL LAY OUT

On the basis of the above considerations the following schematic lay out is proposed for HIAS (hydrogen impulse approximation spectrometer).

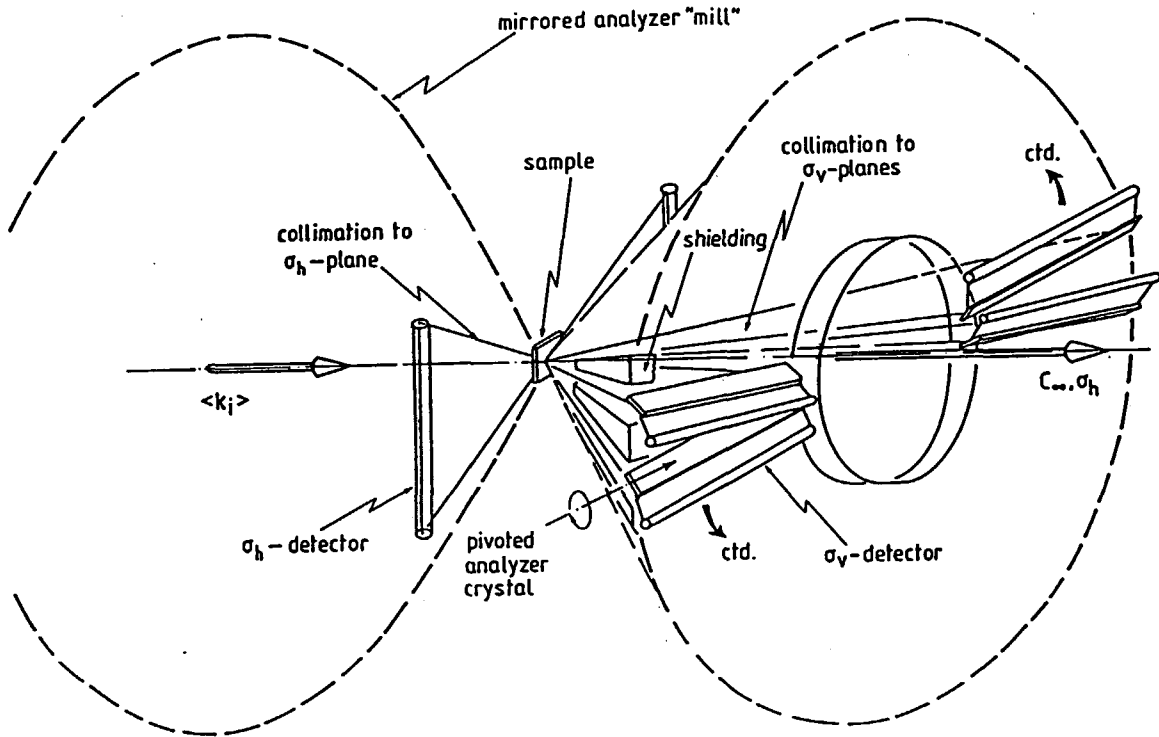


Fig. 2

If the  $\vec{k}_f$ -vectors are collimated to  $\sigma_v$ -planes, i.e.

$$\vec{k}_f \cdot \vec{v} = 0 \quad \text{with} \quad \vec{v} \cdot \vec{n} = 0$$

and the used reciprocal lattice vector  $\vec{G}$  of the analyzer crystal is within the plane containing  $\vec{v}$  and  $\vec{n}$ , i.e.

$$\vec{G} = (\vec{G} \cdot \vec{v}) \vec{v} + (\vec{G} \cdot \vec{n}) \vec{n}$$

then the Bragg condition becomes:

$$\vec{k}_f \vec{G} = (\vec{G} \vec{n}) (\vec{k}_f \vec{n}) = \frac{1}{2} G^2$$

Thus one may scan the exponent of equ. (3) by varying the orientation  $(\vec{G} \vec{n})$  of the "windmill vanes".

The signal for a  $G_v$ -detector is then essentially given by

$$I_v(\alpha) \approx k_f (\vec{n} \vec{M}^{-1} \vec{n})^{-1/2} \int_{\min}^{\max} k_i^{-p-2} dk_i \quad * \quad (4)$$

$$* \exp \left( -\frac{G^2}{4 \cos^2 \alpha} (\vec{n} \vec{M}^{-1} \vec{n})^{-1} \right)$$

where  $\alpha$  is the angle between the reciprocal lattice vector  $\vec{G}$  and the  $\vec{k}_i$ -direction. It was assumed in equ. (4) that the spectrum of the source varies like  $k_i^{-p}$  ( $p \approx 2$ ) in the range (min, max) which is determined by the time gate of the counter.

The necessary values of  $G$  are rather large. Consider e.g. the hydrogen bond in  $\text{KH}_2\text{PO}_4$ . Describing the density distribution by an effective anisotropic oscillator potential, thermal amplitudes of  $u_L^2 \approx 0.03 \text{ \AA}^2$  and  $u_{\parallel}^2 \approx 0.05 \text{ \AA}^2$  perpendicular and parallel to the hydrogen bond have been determined at room temperature /5/. Since

$$(\vec{n} \vec{M}^{-1} \vec{n})^{-1} = 2u_{L,\parallel}^2$$

one needs

$$\frac{G}{\cos \alpha} = \frac{\sqrt{2}}{u_{L,\parallel}} \approx 6 \text{ to } 8 \text{ \AA}^{-1}$$

in order to reach the  $1/e$ -region. Choosing e.g. silicon as analyzer material ( $a = 5.43 \text{ \AA}$ ) this condition becomes for the  $(h,k,l)$ -reflection

$$\cos \alpha = u_{L,\parallel} \frac{\pi\sqrt{2}}{a} \sqrt{h^2+k^2+l^2}$$

From this condition one would recommend Miller indices like (311), (331) and (531) in order to reach convenient Bragg angles ( $\theta=90-\alpha$ ). The  $2\cdot k_f$  neutrons are not reflected and  $3\cdot k_f$  is already suppressed by the scattering function. This removes the main spurions. There remains of course the possibility for "Umweganregung" as usually.

Another problem is the multiple scattering within the sample. This danger demands a plate-like sample with an inserted grid of absorbing material. Note that the measurement does not require a very accurate orientation of the sample. In order to reduce the multiple scattering contribution to an acceptable level of 10%, the thickness of the plates was about 1 mm for Debye-Waller factor determinations /5/. Since the total cross-section decreases from 80 to 20 barns at the  $k_i$ -values considered here, one can relax this condition by a factor of 4. The plate-normal should be tilted off some angle from the  $\vec{k}_i$ -direction in order to allow for scattering to the  $\zeta_h$ -detectors. Their signal is independent of  $\vec{M}$  and essentially given by

$$I_h \approx k_i^{-p} \left| \begin{array}{l} \text{min} \\ \text{max} \end{array} \right.$$

This signal can be used for normalization and the adjustment of the time gate.

## PROPOSED EXPERIMENT

G. Reiter /4/ has discussed the interesting case of a double minimum potential at low temperatures and has shown that even with appreciable noise the oscillatory contribution  $\cos \vec{p} \vec{R}$  to  $f(\vec{p})$  should be recognizable.  $\vec{R}$  connects the two minima. A possible candidate for this type of measurement might be the hydrogen bond of  $\text{KH}_2\text{PO}_4$ . The problem is that the protons order already at  $T_c = 123$  K and therefore one cannot reach the discussed limit. If nevertheless an effective single particle potential governs the dynamics of the proton in the paraelectric phase ( $> 123$  K) one expects the following shape of  $f(\vec{p})$  if one includes the symmetric and the antisymmetric "groundstates". Assuming a symmetric double minimum potential and restricting to a basis of two "pocket" states  $U_0(x)$  centered at  $x = \pm R/2$ , one has

$$\psi_{\pm}(x) = 2(1 \pm S)^{-1/2} (U_0(x - R/2) \pm U_0(x + R/2))$$

$$U_0(x) = u^{-1/2} (2\pi)^{-1/4} \exp\left(-\frac{x^2}{4u^2}\right)$$

$$S = \text{overlap} = \exp\left(-\frac{R^2}{8u^2}\right)$$

Taking the same extension  $u^2$  perpendicular to the bond direction gives:

$$f(\vec{p}) = \left(\frac{2}{\pi}\right)^{3/2} u^3 e^{-2u^2 p^2} (1-S^2)^{-1} \{(1 - S\Delta p) + (\Delta p - S) \cos \vec{p} \vec{R}\}$$

$\Delta p$  denotes the population difference between the two levels. The appearance or disappearance of the  $\cos \vec{p} \vec{R}$ -term depends thus on the competition between overlap and population. Note that this dependence is different from that obtained for the incoherent elastic scattering at small momentum transfers. In the latter case the initial and final states of the proton are the same and one measures the Fourier transform of the density in the position repre-

sentation or equivalently the folding integral in the momentum representation. The impulse approximation however assumes a "freely" recoiling particle, i.e. the recoil energy is much larger than the binding energy and the proton is "ejected" into a free final state. Thus one measures in this case the momentum density of the initial state. This difference between "folding" and "product" is trivial for a Gaussian distribution but for the considered double minimum potential an additional oscillatory contribution with half the argument  $(\cos(\vec{Q}\vec{R}/2))$  results for the incoherent elastic scattering /5/. The dependence of the pre-factors on  $S$  and  $\Delta p$  of the various terms is of course different in the "product"- and "folding"-case. There is the possibility to influence  $T_c$ ,  $S$ ,  $\Delta p$  by pressure. Even if one can not observe the oscillatory contribution (i.e. inadequate description of the proton by effective single particle double minimum potential) one should at least recognize the drastic shrinking of  $f(\vec{p})$  along the bond and the slight elongation along the ferro - electric axis below  $T_c/6/$ .

#### ACKNOWLEDGMENT

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High Resolution Spectroscopy

## Chairman's Report: Working Group "HIGH RESOLUTION SPECTROSCOPY"

Participants: B. ALEFELD, B. EWEN, H. FRIEDRICH, A. HEIDEMANN, A. MAGERL,  
F. MEZEI, R. LECHNER, W. PETRY, D. RICHTER, R. SCHERM,  
A. STEYERL, C. ZEYEN

Chairman: D. RICHTER, KFA Jülich

The High Resolution subpanel discussed neutron spectrometers which on an absolute scale achieve high energy resolution. Such spectrometers use cold neutrons and consequently we discussed only instruments which should be placed at the cold source of SNQ. In order to have a common basis for discussion, the following machine parameters for SNQ were assumed throughout the different contributions:

|     | averaged flux             | peak flux       | pulse length | rep. rate |
|-----|---------------------------|-----------------|--------------|-----------|
| SNQ | $\bar{\phi} = \phi_{ILL}$ | $12 \bar{\phi}$ | $440 \mu s$  | 100 Hz    |

High resolution can in principle be obtained from crystals with Bragg angles close to  $90^\circ$  with time-of-flight techniques using short pulses together with long flight paths (TOF), with neutron spin echo spectroscopy (NSE) and finally with ultra cold neutrons (UCN). For pulsed sources the following spectrometers can be thought of:

|     | Monochromator  | Analyser  | Abbreviation |
|-----|----------------|-----------|--------------|
| (a) | X              | X         | MUSICAL      |
| (b) | TOF            | X         | IRIS         |
| (c) | X              | TOF       | BUNCHING     |
| (d) | TOF            | TOF       | TOF          |
| (e) | Time of flight | Spin echo | TOF-NSE      |
| (f) | UCN            |           | UCN          |

High resolution neutron spectrometers as mentioned above already now have a wide range of applications in solid state physics, physical chemistry, polymer physics and biology. Some examples are given below:

(1) Single particle dynamics as studied by incoherent scattering

- (a) diffusion and tunneling of H in metals
- (b) diffusion in liquid crystals
- (c) self-diffusion in metals below the melting point
- (d) single particle motion in solid electrolytes and intercalates
- (e) reorientation of molecules in plastic crystals
- (f) tunneling motion of molecules in molecular crystals
- (g) motional mechanisms of molecules at surfaces (catalyzers)
- (h) polymer relaxation on a monomer scale

(2) Collective behaviour of matter studied by coherent scattering

- (a) dynamics of polymers
- (b) dynamics of simple biological substances
- (c) critical scattering at phase transitions
- (d) dynamics in spin glasses
- (e) special collective excitation  
(roton in  $\text{He}^4$ )

(3) Hyperfine interactions

From this large spectrum of applications it is clear that high resolution spectrometer will be an important part of the instrumentation at SNQ.

The high resolution working group discussed possible spectrometers at three meetings: During the two general meetings in Bad Kissingen and Maria Laach and at a special meeting in Grenoble which took place at the beginning of June. During these 3 meeting all different instruments outlined in the first table were discussed. Out of these four instruments were considered to be first priority machines (Class A) which should be built in a first stage of SNQ. They are: the backscattering spectrometer MUSICAL /1/, the time of flight backspectrometer IRIS /2/, a double chopper time of flight spectrometer /3/ and one or two time of flight neutron spin echo instruments /4/. The first 3 machines are able to fully utilize the peak flux of SNQ. A future compressor ring would increase their intensity as much as it in-

creases the peak flux at the cold source. They would not suffer from a lower repetition rate as long as the peak flux is raised accordingly. However, longer pulses as a consequence of lower repetition rates would affect these machines negatively. Neutron spin echo as a broad wavelength band method, on the other hand, cannot fully exploit the peakflux of SNQ, since it readily suffers from frame overlap. At 100 Hz gain factors of 3 to 5 over Grenoble are achievable depending on wavelength. From lower repetition rates spin echo would gain essentially inversely proportional to the rate. Longer neutron pulses reduce its q-resolution but basically do not affect its performance.

The other spectrometers (class B) are not worked out enough or contain technically very critical components like fast Doppler drives that a recommendation is not possible at this point. These spectrometers are: a time focusing bunching spectrometer /5/, a backscattering spectrometer using phase space transformation /6/, a crystal time focussing machine /7/ and an IRIS spectrometer working with graphit crystals /2/.

Concerning ultra cold neutrons a time modulated source may allow an enhanced production /8/. All spectrometers dealing with UCN's are identical to those on continuous sources and therefore were not discussed further.

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| /8/ A. Steyerl                           | (this report) |

## The modified Backscattering Spectrometer 'Musical'

B. Alefeld, D. Richter  
IFF, KFA Jülich, Jülich, Germany

A. Heidemann  
Institut Laue-Langevin, Grenoble, France

### I. INTRODUCTION

Neutron spectroscopy with  $\mu\text{eV}$ -resolution has become a well established tool for the investigation of the dynamics in solids and liquids. The backscattering method [1] is particularly well suited to study single particle motions with incoherent neutron scattering. In the past ten years many experiments have been performed with the backscattering spectrometers in Jülich and Grenoble. The range of applications is quite vaste. The following topics can be identified:

#### 1. Single particle motions:

- a) Hydrogen in metals and alloys
- b) Self diffusion processes in metals, alloys, superionic conductors, intercalation compounds and liquid crystals
- c) Reorientational and tunneling motions in molecular crystals
- d) Motions on surfaces
- e) Dynamics of simple biological model substances
- f) Dynamics of polymers

#### 2. Collective phenomena

- a) Phase transitions
- b) Spin glasses
- c) Polymers

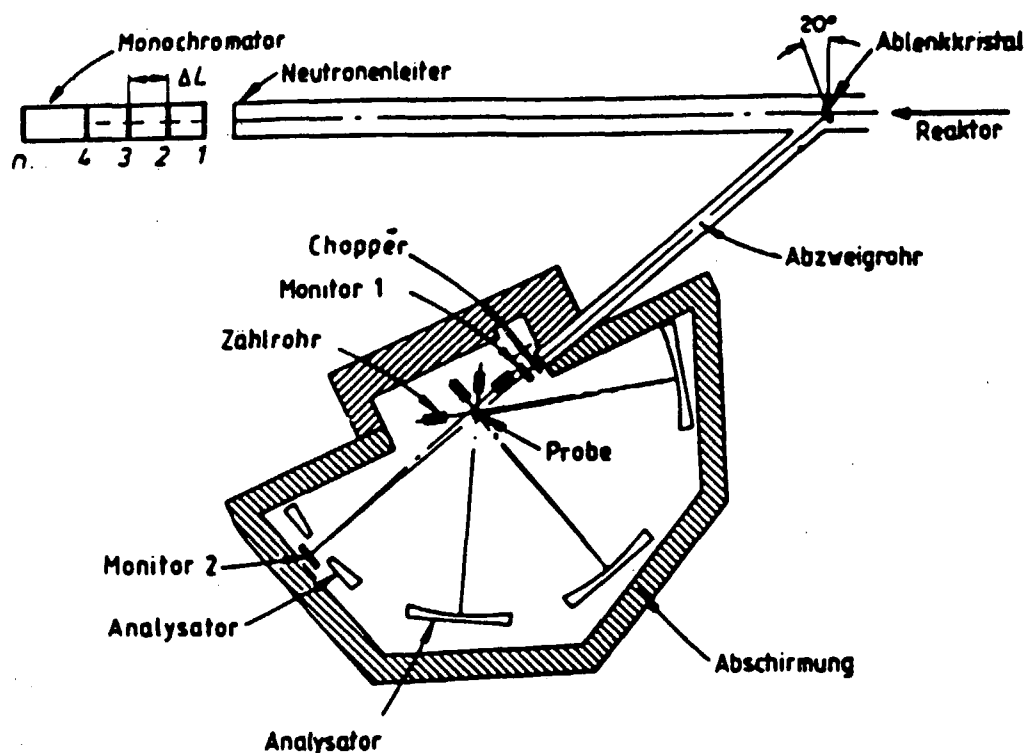
#### 3. Hyperfine interactions

One of the main limitations in high resolution neutron spectroscopy is the intensity even with instruments at a high flux reactor or on future high flux spallation sources. Therefore a very careful and thorough intensity optimisation is primordial. A modified version of a backscattering spectrometer which makes full use of the peak flux of a pulsed source has been proposed recently [2].

In this report we will describe (i) the principle of this spectrometer and (ii) possible layouts on the spallation sources SNS and SNQ. A comparison will be made with the existing backscattering spectrometer IN10 [3] at the ILL and a TOF-backscattering spectrometer like IRIS planned at the SNS [4, 5], both using cold neutrons. The case of a modified backscattering spectrometer for thermal neutrons will also be discussed.

## II. PRINCIPLE

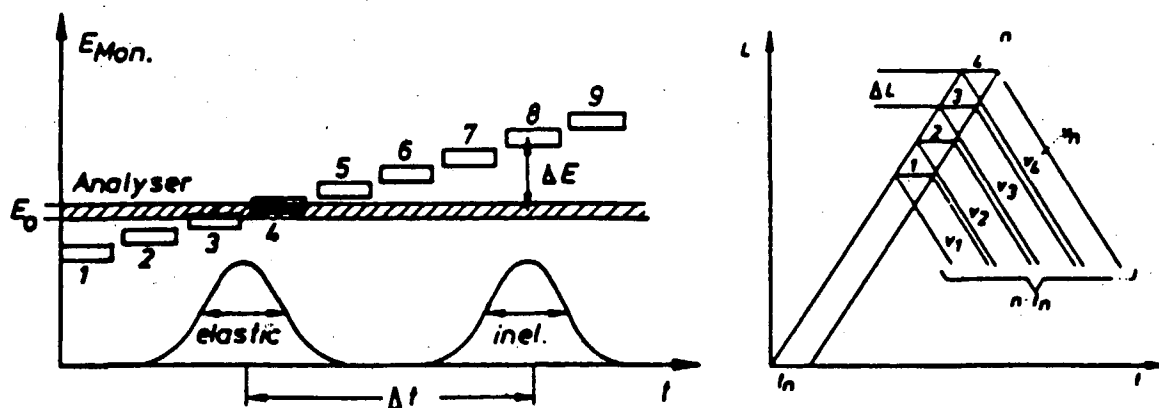
The spectrometer is sketched in fig. 1. Apart from the sophisticated



**Fig. 1** : The multimonochromator backscattering spectrometer 'Musical'.

monochromator the instrument is very similar to IN10: The mono-energetic neutrons, backscattered from the monochromator crystal, are deflected by a graphite crystal towards the sample. The scattered neutrons are analysed for energy and momentum transfer by spherically curved analyser crystals in backscattering geometry. In order to be able to discriminate between neutrons scattered directly into the detectors and neutrons reflected from the analysers the beam has to be chopped with a duty cycle of about 50%. The main difference between 'Musical' and IN10 is given by the monochromator.

On IN10 the monochromator is one Silicon crystal (orientation (111), Bragg angle  $90^\circ$ ) mounted on a velocity drive. On the 'Musical' spectrometer - in order to make full use of the peak flux of a pulsed source - the monochromator consists of several crystals with the same orientation but slightly different lattice spacings mounted behind each other. Each crystal reflects a slightly different wavelength. Due to the path length differences the differently "coloured" neutrons arrive at different times at the sample (see fig. 2).



**Fig. 2** : Time-of-flight diagram of the modified backscattering spectrometer 'Musical'.



The gain factor  $G$  of the modified backscattering spectrometer compared to the conventional one on a pulsed source is equal to the number  $n$  of crystals of the multimonochromator.

In order to achieve a clear separation in time the distance  $s$  between two monochromator crystals has to be

$$s \geq v \cdot \tau / 2 \quad (1)$$

where  $v$  is the neutron velocity and  $\tau$  the pulse length of the source. The optimum number of monochromator crystals is given by

$$n_{\text{opt}} = T / 2\tau \quad (2)$$

where  $T$  is the period of the pulsed source. The factor 2 occurs because of the necessity of discrimination between directly and indirectly (via the analyser) scattered neutrons. This condition correlates also the length  $l_1 = v \cdot T$  of the monochromator with the distance  $l_{\text{SA}}$  between sample and analyser:

$$l_{\text{SA}} = m \cdot l_M = m \cdot n \cdot s, \quad m = 1, 2, \dots \quad (3)$$

$$l_{\text{SA}} = m \cdot T \cdot v / 4 \quad (3a)$$

In order that the neutrons backscattered from the different monochromators do not overlap energetically we postulate that the energy difference  $\delta E$

between neutrons reflected from two nearest neighbour monochromators should be larger than the intrinsic full width at half maximum  $\delta E_M$  of the Bragg reflection of one crystal:

$$\delta E \geq \delta E_M \quad (4)$$

This energy difference can be produced either by the use of thermal expansion or by doping, thereby achieving different lattice parameters  $a$ :

$$\delta E = \begin{cases} 2E\beta\delta T \\ 2E\frac{\delta a}{a} \end{cases} \quad (5)$$

The intrinsic line width  $\delta E_M$  of the Bragg reflection from a perfect crystal is determined by the primary extinction:

$$\delta E_M = 2E \cdot \frac{F_{hkl} \exp(-W)}{V_c |\tau_{hkl}|^2} \cdot 16\pi \cdot 1,061 \quad (6)$$

with  $E$ : Incident energy

$F_{hkl}$ : Structure factor

$\exp(-W)$ : Debye Waller factor

$V_c$ : Volume of the elementary cell

$\tau_{hkl}$ : Reciprocal lattice vector.

For Silicon crystals with (111) orientation  $\delta E_M$  is equal to 0.08  $\mu\text{eV}$ .

Commercially available Silicon crystals with polished surfaces have in fact this line width [6]. Silicon crystals with unpolished surface have an intrinsic

width of about 0.5  $\mu\text{eV}$  [3]. Using eq. (5) we obtain:

$$\frac{\delta a}{a} = \left\{ \begin{array}{ll} 2 \cdot 10^{-5} & \text{perfect Si(111)} \\ 1.25 \cdot 10^{-4} & \text{unpolished Si (111)} \end{array} \right\}$$

or

$$\delta T = \left\{ \begin{array}{ll} 8^\circ\text{C} & \text{perfect Si (111)} \\ 50^\circ\text{C} & \text{unpolished Si(111)} \end{array} \right\}$$

$\delta T$  is the temperature difference between two nearest neighbour crystals.

The total energy band width covered simultaneously by the multimonochromator is

$$\Delta E_{\text{Band}} = n \cdot \delta E_M \quad (7)$$

We will see later that this band width is too narrow with respect to the needs of a high resolution spectrometer. Therefore the center of gravity of this energy band has to be scanned for example by the use of the Doppler effect. On IN10 an energy transfer range of  $\pm 15 \mu\text{eV}$  is available (Doppler velocity  $\pm 2.5 \text{ m/s}$ ). It is possible to extend this range by about a factor of two using (i) high performance drives and (ii) a graphite deflector crystal with a temperature gradient ( $\Delta T \geq 100^\circ\text{C}$ ).

Apart from obvious technical problems of such a multimonochromator mounted on a high speed Doppler drive there is a problem of interference of neutrons traversing an accelerated multimonochromator system. This interference produced by unwanted multiple reflections could lead to a partial

loss of the correlation between the time of arrival and the energy of the neutron. This problem will be discussed in more detail in a later paper. The problem could be avoided by a hydraulic velocity drive capable of producing trapezoidal velocity profiles. Such a device is in development in Jülich [7].

### III. LAYOUT AND TECHNICAL PROBLEMS

Table I shows the parameters of 'Musical' spectrometers adapted to the time structure of the SNQ and SNS. Apparently the time structure of the SNQ is optimal for the spectrometer. This is not quite the case for the SNS : The number of crystals is too high, the sample-analyser distance too long. If one reduces these numbers to more realistic ones reduced gain factors have to be tolerated.

If one produces the lattice parameter differences between the crystals by thermal expansion then non-trivial technical problems can be foreseen. These could be avoided if crystals with slightly different lattice parameters could be produced by doping, for example Silicon doped with Germanium (relative lattice parameter difference between Si and Ge  $\Delta d/d = 0.04$ ).

In order to achieve a shift of 0.5  $\mu\text{eV}$  with the latter system a doping concentration of only 0.3% is required. Therefore for a multimonochromator of 11 crystals (and the 'low' resolution version) the highest doping concentration is only 3.3%. According to recent information from the semiconductor industry good quality Germanium doped Silicon crystals can be produced with these small concentrations [8].

Another side-aspect is to use  $n$  monochromators with the same lattice spacing behind each other on a velocity drive. If the total energy range covered by the Doppler drive is larger than  $n \cdot \delta E_M$  then a gain factor  $1 < G < n$  can also

be achieved compared to a conventional monochromator.

In order to be able to make a clear separation in time of flight between the neutrons reflected from the different monochromator crystals the source pulse shape which has a very long tail on the SNQ has to be tailored resulting in an intensity loss of about 20%.

As mentioned already earlier the source parameters of the SNQ are quite optimal for 'Musical'. However, if the source pulse length  $\tau$  would be shortened by a compressor ring then one could use a higher number of monochromator crystals and gain more intensity.

#### IV. COMPARISON BETWEEN 'MUSICAL' AND 'TOF-BACK'

'TOF-Back' is a combined TOF-backscattering spectrometer [4]. Its great advantage is its flexibility in the energy transfer range. Its disadvantage is the sensibility of the width of the energy resolution function to sample size effects. The properties of 'Musical' are just complementary: Problems with the energy scan range limitation, no sample size problems: An energy resolution of 0.3  $\mu\text{eV}$  (FWHM) is possible.

In order to be able to make an intensity comparison we have to write down the intensity to be expected at the sample position and in the detector.

##### IV.1. Intensity $J_s$ at the Sample Position

$$J_s = \int_{E_i} \frac{d\phi}{dE} f(E_i) dE_i \cdot \frac{\tau}{T} \eta_i F \quad (8)$$

where  $\frac{d\phi}{dE}$  : Differential peak flux

$f(E_i)$ : Transmission function of the primary spectrometer

F: Beam cross section at sample site

$\eta_i$ : Loss factor of the primary spectrometer

a) 'Musical':

This spectrometer should be installed at the end of a bent cold neutron guide of a large cross section (typically  $6 \times 20 \text{ cm}^2$ ). The large beam cross section can be focussed with a super mirror nose and a vertically curved deflector onto the sample. The loss factor  $\eta_i$  of such an arrangement has been discussed in detail by A. Magerl [9]. The integral in eq. (8) can be replaced by:

$$\int \frac{d\phi}{dE} f(E_i) dE_i = \frac{d\phi}{dE} \delta E_M n \cdot f_{\text{Tailor}} \quad (9)$$

where  $f_{\text{Tailor}}$  is the reduction factor due to the tail cutter. Therefore:

$$J_S = \frac{d\phi}{dE} \delta E_M n f_{\text{Tailor}} \cdot F \cdot \eta_i \cdot \frac{\tau}{T} //$$

b) 'TOF-Back':

This spectrometer needs a very long primary flight path of the order of 100 to 200 meters. Therefore not negligible guide losses will occur. Other losses will be produced by imperfect choppers. The integral in eq. (8) can be replaced by

$$\int \frac{d\phi}{dE} f(E_i) dE_i = \frac{d\phi}{dE} \Delta E_{\text{Band}} \cdot \frac{\tau}{T} \quad (10)$$

where  $\Delta E_{\text{Band}}$  is the band width and  $\tau_1$  the pulse width of the incident wave length band. Typical values for the SNQ are  $\Delta E_{\text{Band}} = 60 \mu\text{eV}$  and  $\tau_1/\tau = 0.2$  for a spectrometer with  $1 \mu\text{eV}$  TOF resolution. Therefore

$$J_S = \frac{d\phi}{dE} \Delta E_{\text{Band}} \frac{\tau_1}{T} \eta_i F$$

Comparing the intensities at the sample position of 'Musical' and 'TOF-Back' on the SNQ, we see that apart from loss factors  $\eta_i$  both instruments have about the same intensity:

$$J_S = \begin{cases} \overbrace{\frac{d\phi}{dE} \delta E_M}^{0.5 \mu\text{eV}} \cdot f_{\text{Tailor}} \eta_i \cdot \frac{1}{2} \cdot F \text{ (Musical)} \\ \frac{d\phi}{dE} \underbrace{\Delta E_{\text{Band}} \cdot \frac{\tau_1}{T}}_{0.6 \mu\text{eV}} \eta_i \cdot 1 \cdot F \text{ ('TOF-Back')} \end{cases}$$

#### IV.2 Intensity in the Detector

For the case of a quasielastic scattering law where the width  $\Gamma$  is much smaller than the total energy transfer range, the intensity  $J_D$  in the detector is given by [5]:

a) 'Musical':

$$J_D = J_S \cdot W_{\text{sample}} \cdot \Delta \Omega_a \cdot \eta_f \frac{\delta E_a}{\Delta E} \quad (11)$$

$$\text{or } J_D = \frac{d\phi}{dE} \delta E_M \cdot f_{\text{Tailor}} \cdot \eta_i \frac{1}{2} \cdot F W_{\text{sample}} \Delta \Omega_a \eta_f \frac{\delta E_a}{\Delta E}$$

b) 'TOF-Back':

$$J_D = J_S W_{\text{sample}} \cdot \Delta \Omega_a \cdot \eta_f \frac{\delta E_a}{\Delta E_{\text{Band}}} \quad (12)$$

$$\text{or } J_D = \frac{d\phi}{dE} \delta E_a \cdot \eta_i \cdot F W_{\text{sample}} \cdot \Delta \Omega_a \cdot \eta_f \cdot \frac{\tau_1}{T}$$

where

$W_{\text{sample}}$ : Scattering power of the sample.

Example: Incoherent scatterer

$$W_{\text{sample}} = \frac{\sigma_{\text{inc}}}{4\pi} \cdot \frac{k_i}{k_f} N_V \cdot d$$

with  $\sigma_{\text{inc}}$ : Incoherent scattering cross-section

$N_V$ : Number density of scattering centers

$d$ : Effective sample thickness.

$\eta_f$ : Loss factor of the secondary spectrometer

$\Delta \Omega_a$ : Solid angle of the analyser

$\delta E_A$ : Energy resolution width of the analyser.

The main difference between eq. (11) and (12) apart from the loss factor  $\eta_i$  is given by

$$J_D \sim \begin{cases} 8 \cdot 10^{-3} & < \frac{\delta E_a}{\Delta E} < 0.1 & \text{Musical} \\ \frac{\tau_1}{T} = 10^{-2} & & \text{TOF-Back} \end{cases}$$



Therefore the 'Musical' is superior to 'TOF-Back' if the width of the scattering law is much narrower than the energy window. This window is fixed for the 'TOF-Back' spectrometer. It is adjustable for 'Musical', which is a big advantage.

#### V. Musical for Short Wavelengths

Until now we have only discussed the case of a backscattering spectrometer for cold neutrons like IN10. In this section we will investigate the problems connected with a backscattering instrument for thermal neutrons like IN13 on a pulsed source. If we would use as monochromator material  $\text{CaF}_2$  with (422) orientation then we have:

$$\begin{aligned}\lambda &= 2.23 \text{ \AA} & v &= 1777 \text{ m/s} \\ E &= 16.3 \text{ meV}\end{aligned}$$

From eq. (1) we get the distances between two crystals:  $s = 40 \text{ cm}$ .

Due to the fact a  $\text{CaF}_2$  (422) monochromator reflects also neutrons with  $\lambda/2$ , we have to modify eq. (2):

$$n_{\text{opt}} = T/3\tau \approx 7$$

This yields a distance between the sample and the analyzer of the order of 2.8 m which is too long. We can get rid of this problem by the following trick:

We increase the distance  $s$  by a factor of 3. The resulting situation is shown in a TOF diagram in fig. 3: Between two consecutive pulses

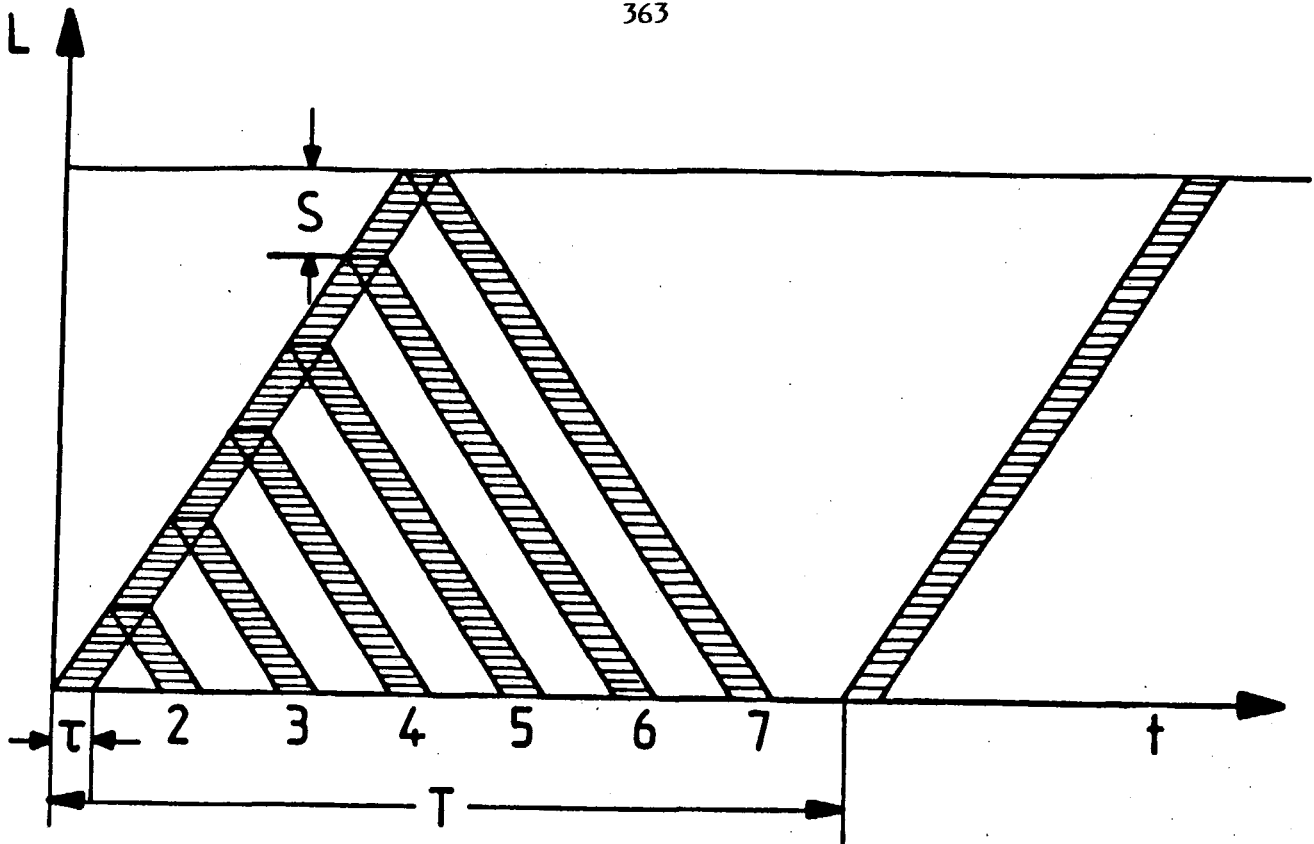


Fig. 3: Time-of-flight diagram of the modified backscattering spectrometer 'Musical' for thermal neutrons.

produced by two nearest neighbour monochromators there is a gap of  $2\tau$  which can be used for the discrimination between directly and indirectly scattered neutrons. The sample analyser distance is now:

$$l_{SA} = 3 \frac{v \cdot \tau}{2} = 1,2 \text{ m.}$$

This is a very reasonable value. The total length of the monochromator system is about 8 meter. It consists of 7 cryofurnaces and neutron guides in between them. The temperature difference between two neighbour furnaces

is obtained from eq. (5):  $\Delta T \approx 5^\circ\text{C}$  corresponding to an intrinsic width of  $3 \mu\text{eV}$  of one  $\text{CaF}_2$  (422) crystal. This measured value is much bigger than the one calculated for a perfect crystal ( $\delta E_M = 0.2 \mu\text{eV}$ ). Each furnace could be scanned between  $-190^\circ\text{C}$  and  $+450^\circ\text{C}$  covering an energy range of  $-300 \mu\text{eV} < h\nu < +120 \mu\text{eV}$ .

One problem with this monochromator is the transmission of the  $\text{CaF}_2$  crystals, which is of the order of  $0.6\%/mm$ . The necessary thickness of one monochromator crystal is between 5 and 10 mm. Therefore the beam is attenuated by a factor between 1.5 and 2.3 after the passage through  $2 \times 7$  crystals. In addition there are furnace heat screens. A very careful design of this monochromator is necessary.

Instead of  $\text{CaF}_2$  (422) one could think of using Ge-doped Si (511) crystals with a lattice parameter gradient ( $\lambda = 2.09 \text{ \AA}$ ). Here transmission problems would be less severe.

Concluding one can say that also for a 'thermal' backscattering spectrometer one can use the peak flux of the pulsed source.

## VI. Conclusion

The modified backscattering spectrometer 'Musical' and the TOF-Backscattering spectrometer are very complementary. Both can use the peak flux of the pulsed source. With a 'Musical' spectrometer for cold neutrons an energy resolution down to  $0.3 \mu\text{eV}$  (FWHM) can be achieved and an energy transfer of  $\pm 30 \mu\text{eV}$  be covered. With 'TOF-Back' an energy resolution of about  $1.5 \mu\text{eV}$  is possible. The energy transfer range is again about  $60 \mu\text{eV}$ . The center of gravity of the latter can be shifted easily into the meV-range. Both spectrometers should be installed on the SNQ. 'Musical' at the SNQ will have a flux at the sample position about 10 times higher than that of IN10 at the ILL. A 'Musical' spectrometer for thermal neutrons at the SNQ can also use the peak flux. The instrument would have an energy resolution of about  $8 \mu\text{eV}$  and an energy range between  $-300$  and  $+120 \mu\text{eV}$ .

| Parameter                                                         | SNQ                  |                        |  | SNS                |                       |                    |                       |
|-------------------------------------------------------------------|----------------------|------------------------|--|--------------------|-----------------------|--------------------|-----------------------|
|                                                                   | Optimum              |                        |  | Optimum            |                       |                    |                       |
| Monochromator                                                     | Si(111)<br>(perfect) | Si(111)<br>unpolished) |  | Si(111)<br>perfect | Si(111)<br>unpolished | Si(111)<br>perfect | Si(111)<br>unpolished |
| Monochromator<br>Energy Resolution<br>FWHM ( $\mu\text{eV}$ ) [3] | 0.08 ... 0.12        | 0.5                    |  | 0.08 ... 0.12      | 0.5                   | 0.08 ... 0.12      | 0.5                   |
| Gain $G = n$                                                      | 11                   |                        |  | 80                 |                       | 40                 | 20                    |
| $s$ (cm)                                                          | 14                   |                        |  |                    |                       |                    |                       |
| $L_M$ (cm)                                                        | 154                  |                        |  | 320                |                       | 160                | 80                    |
| $n \cdot \delta T$ ( $^{\circ}\text{C}$ )                         | 88                   | 550                    |  | 640                | 4000                  | 320                | 1000                  |
| $\Delta E_{\text{Band}}$ ( $\mu\text{eV}$ )                       | 0.88                 | 5.5                    |  | 6.4                | 4.0                   | 3.2                | 10                    |
| Source<br>Period $T$ (ms)                                         | 10                   |                        |  |                    |                       | 20                 |                       |
| Pulse Width $\tau$ ( $\mu\text{s}$ )                              | 440                  |                        |  |                    |                       | 125                |                       |
| Peak Flux<br>(cold neutrons)                                      | 10                   |                        |  |                    |                       | 1                  |                       |
| Realistic/Optimum<br>Peak flux loss factor)                       | 1                    |                        |  | 1                  |                       | 0.5                | 0.25                  |

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Peak Flux at ILL : 1.5  
(cold neutrons)

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## TIME-OF-FLIGHT SPIN-ECHO

F. Mezei

ILL, 156 X, 38042 Grenoble, France\*

D. Richter

Institut für Festkörperforschung, KFA Jülich GmbH, D-517 Jülich,  
Federal Republic of Germany

## I. INTRODUCTION

The basic set-up of a neutron spin echo spectrometer providing ultra high resolution is that of IN11 at the ILL /1,2/. It is characterized by long solenoidal guide fields before and after the sample which principally can be corrected for field inhomogenities. This arrangement allows the best energy resolution which is achievable with neutron spin echo. It is displayed schematically in Fig. 1.

In the following we shall briefly outline the spin echo method on continuous sources. Thereafter we discuss the implications of time-of-flight spin echo and in particular consider large solid angle machines. Finally we compare the performance of neutron spin echo machines on continuous and pulsed sources with special emphasis on ILL and SNQ.

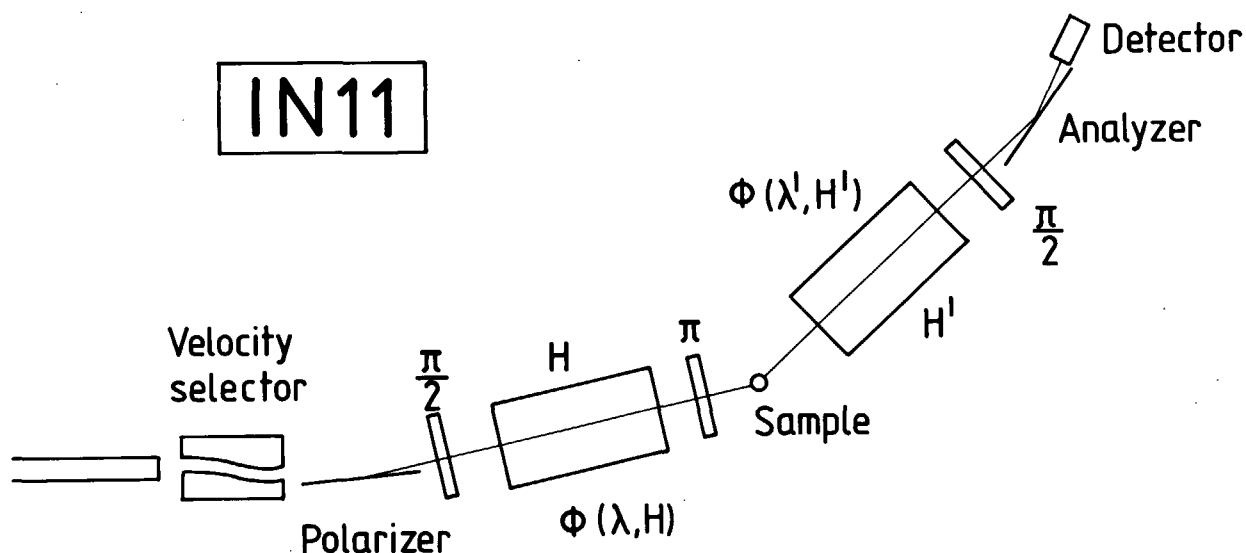


FIG. 1: Schematic sketch of the spin-echo set-up at ILL.

\*present address: Hahn-Meitner-Institut and TU Berlin

## II. BASIC PRINCIPLES

A neutron spin echo spectrometer measures the neutron polarization at the analyzer as a function of magnetic guide field  $H$ . A polarized neutron beam with initial polarization  $P_i$  passes a longitudinal guide field  $H$  performing  $\phi(H, \lambda)$  precessions. Then the neutrons are scattered at the sample, thereby changing their wavelengths by  $\delta\lambda = \lambda' - \lambda$ , and traverse a guide field  $H'$  in opposite direction undergoing  $\phi(H', \lambda')$  precessions. (At IN11 the opposite field direction is achieved by a  $\pi$ -flipper). Thereafter the polarization is analyzed. The final polarization  $P_f$  is given by

$$P_f = P_i \cos(\delta\phi) \quad (1)$$

where

$$\delta\phi = \phi(\lambda', H') - \phi(\lambda, H) = \frac{2|\gamma|\mu_N m}{\hbar^2 2\pi} [H'\lambda' - H\lambda] S$$

where  $S$  is the effective length of the coils,  $\gamma$  the gyromagnetic ratio,  $\mu_N$  the nuclear magneton, and  $m$  the mass of the neutron. For small energy transfers that is in the quasielastic region on which we will restrict ourselves also in the following,  $H$  is chosen to equal  $H'$ . If we further convert the change of wavelength  $\delta\lambda$  to the energy transfer at the sample  $\hbar\omega$  we get for

$$\delta\phi = \frac{2|\gamma|\mu_N m^2 HS\omega \lambda^3}{\hbar^3 8\pi^3} \quad (2)$$

The scattering experiment integrates over all energy transfers

$$P_f = P_i \int_{-\infty}^{+\infty} S(Q, \omega) \cos(\omega t) d\omega \quad (3)$$



Thereby we have introduced the time variable

$$t = \frac{2|\gamma| \mu_N m^2}{8 \pi^3 h^3} \lambda^3 HS = C_t \lambda^3 HS \quad (4)$$

Thus  $P_f$  is essentially proportional to the real part of the intermediate scattering function  $S(Q,t)$ .

Since to first order spin echo decouples the energy resolution from the width of the incident wavelength distribution, a spin echo spectrometer is able to make use of a broad incident wavelength spectrum  $I(\lambda)$  ( $\Delta\lambda/\lambda \approx 20\%$ ) which is provided by a velocity selector.  $I(\lambda)$  is integrated by the analyzing process

$$P_f = P_i \int_0^{\infty} I(\lambda) d\lambda S(Q,t(\lambda)) \quad (5)$$

obscuring thereby the relationship between  $P_f$  and  $S(Q,t)$ . Eq. (4) shows that  $t$  depends linearly on the guide field  $H$ . A scan in  $t$  is performed by varying  $H$ . The number of points taken depends on the problems but 10 to 20 are typical numbers.

### III. TIME-OF-FLIGHT SPIN ECHO

In a time-of-flight experiment we can separate the incoming wavelength by measuring the flight time. For very small energy transfer  $\delta\lambda \ll \lambda$  which we are considering in a high resolution experiment we have

$$T_F = \frac{mL}{h 2\pi} \lambda \quad (6)$$

where  $L$  is the total flight path. The Fourier time  $t$  becomes a function of the flight time  $T_F$ .

$$t = c_t \left( \frac{\hbar^2 \pi}{mL} \right)^3 T_F^3 HS \quad (7)$$

A scan in  $t$  can now be performed by measuring the flight time at a constant magnetic field, which for a given wavelength distribution determines the accessible time range.

Scanning with the wavelength means that not only  $t$  but also  $Q$  is varied.

$$Q = 2 \frac{mL}{\hbar T_F} \sin \theta \quad (8)$$

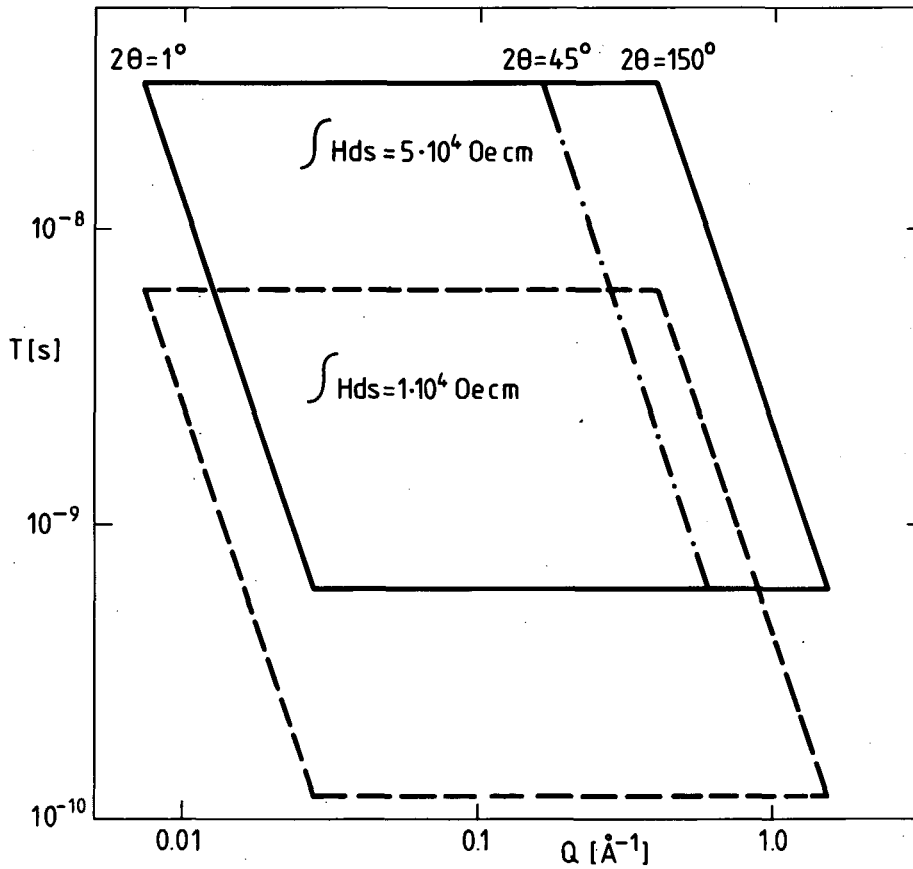


FIG. 2:  $Q$ - $t$  range covered by time-of-flight spin echo using wavelengths between  $4 \text{ \AA} < \lambda < 15 \text{ \AA}$ .

Figure 2 displays the accessible Q-t range for  $H_s = 5 \cdot 10^4$  Oecm and  $H_s = 10^4$  Oecm using scattering angles between  $1^\circ$  and  $150^\circ$ . At each angle scans are performed as indicated by the dashed dotted line.

Thus, in order to reconstruct  $S(Q,t)$  without scanning many magnetic fields, simultaneous measurements over a large solid angle are highly desirable. This can only be achieved, if the cylindrical field symmetry of IN11 is replaced by a plane of symmetry as provided by the vertical field of an iron yoke magnet. Concerning resolution there are two inherent draw backs:

- (1) As a consequence of Maxwell's laws the relative field inhomogeneity in the case of a symmetry plane is 4 times larger than that of the cylindrical arrangement /1/. For cylindrical symmetry it is approximately given by

$$\eta = r^2 / D l_{\text{eff}}, \quad (9)$$

where  $r$  is the radial dimension of the beam,  $D$  the diameter and  $l_{\text{eff}}$  the effective length of the solenoid. For an iron yoke magnet  $\eta$  becomes

$$\eta = \frac{2 z^2}{G l_{\text{eff}}} \quad (10)$$

$z$  being the beam height, and  $G$  the gap width.

- (2) Solenoidal arrangements where the neutrons propagate parallel to the magnetic field can in principle be corrected, while this is not possible for vertical fields. Current loops which are introduced into the beam volume give rise to  $\text{rot } H \neq 0$  for neutrons passing the loop while  $\text{rot } H = 0$  for neutrons travelling outside. In this way spatial inhomogenities are correctable - in case of IN11 such corrections in the form of Fresnel coils /1/ have been applied successfully. For transverse geometry such a scheme is not possible.

Thus, with respect to resolution for principle reasons large solid angle NSE spectrometers are anticipated to be inferior to solenoidal arrangements. With respect to neutron economy, however, analysers with large angular acceptance provide a tremendous gain in intensity which easily could exceed factors of 100. Consequently the construction of two different NSE spectrometers is advisable. (1) A very high resolution version which is most powerful in the study of macromolecular dynamics (polymers, liquid crystals, biological materials, mycelles etc.), and  $Q = 0$  phase transitions in which cases good momentum and energy resolution is required. The interesting  $Q$ -range thereby is limited to  $0.005 \leq Q \leq 0.2 \text{ \AA}^{-1}$ . It would basically look like IN11 which is shown schematically in Fig. 1. (2) A high solid angle spectrometer would be best adapted to the study of relaxation processes at larger  $Q$ 's, which include spin dynamics (spin glasses, low dimensional systems, etc.), and diffusion phenomena in crystals. A schematic layout of such a spectrometer is presented by Fig. 3. While the part in front of the sample (apart from a vertical guide field) resembles that of IN11, large area magnets and a multimirror and multidetector system are characteristic for the analyser part.

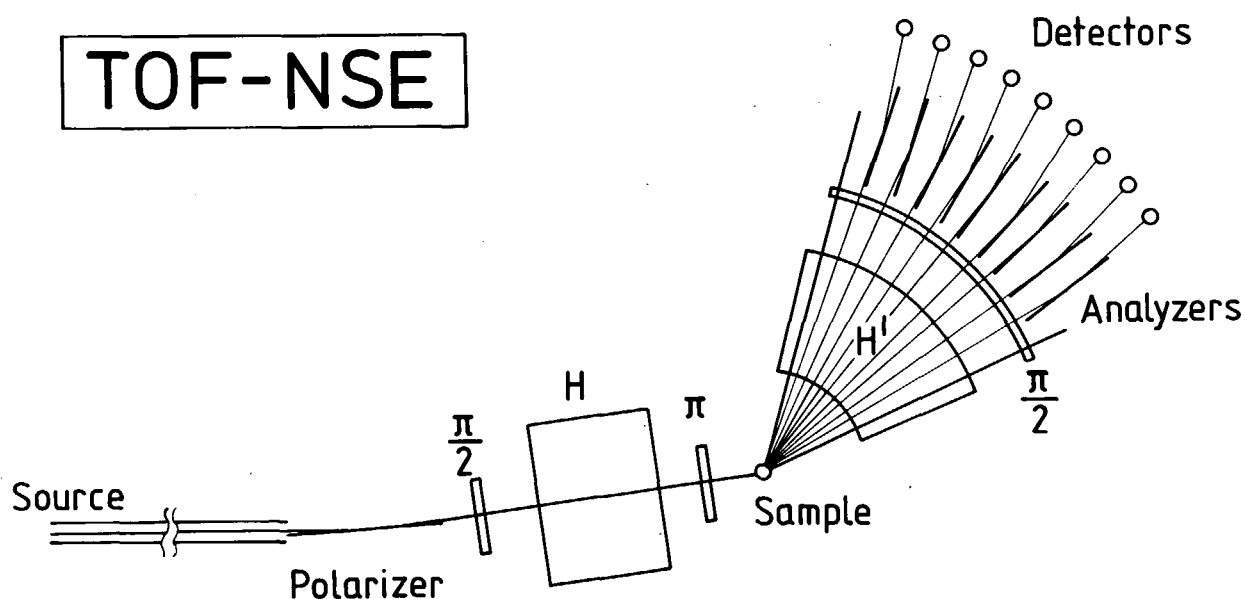


FIG. 3: Principle arrangement for a time-of-flight spin-echo spectrometer using a large solid angle on the analyser side.

## IV. NEUTRON SPIN ECHO AT THE SNQ

To make practical use of neutron spin echo at a pulsed source, the steady-state spin echo spectrometer has to be amended by a course time-of-flight spectrometer which determines the neutron velocity with a few percent precision. It replaces the velocity selector in the steady state version. Immediately we can make the following statements:

- (a) The  $q$  resolution via  $\delta\lambda/\lambda$  can be better on SNQ than that of IN11 (20 %).
- (b) The neutron economy on SNQ can be better: Neutrons out of a wavelength band  $\Delta\lambda$  can be utilized which is determined by the frame-overlap condition

$$\frac{\Delta\lambda}{\lambda} = \frac{v\tau}{L} \quad (11)$$

where  $L$  is the length of the instrument and  $\tau$  the time in between pulses ( $\lambda_{\max} - \lambda_{\min}$  [in  $\text{\AA}$ ] =  $4 \tau/L$  [ $\tau$  in  $\text{ms}$ ] and  $L$  in  $\text{m}$ ]).

Thus the flux which can be used at SNQ is given by

$$\phi_{\text{eff}} = \int_{\lambda_{\min}}^{\lambda_{\max}} \phi(\lambda) d\lambda \quad (12)$$

In the steady state case this integral extends from  $0.9 \lambda_0$  to  $1.1 \lambda_0$  where  $\lambda_0$  is the nominal wavelength. Assuming  $\phi(\lambda) = \text{const}$  within the wavelength window of the instrument, the flux enhancement at SNQ compared to the steady state case can approximately be written as

$$\phi_{\text{eff}} = \bar{\phi} \frac{\lambda_{\max} - \lambda_{\min}}{0.2 \lambda} = \frac{\Delta\lambda\tau v}{L} \bar{\phi} \quad (13)$$

where  $\bar{\phi}$  is the time average flux.

- (c) Defining the dynamic range  $R$  of the spectrometer by the ratio of maximum  $t_{\max}$  and minimum  $t_{\min}$  Fourier times which can be accessed during one scan, we get (Equ. (4))

$$R = \frac{t_{\max}}{t_{\min}} = \frac{H_{\max} \lambda_{\max}^3}{H_{\min} \lambda_{\min}^3} \quad (14)$$

As a consequence of the wide wavelength band  $\Delta\lambda$ ,  $R$  will be considerably larger on SNQ than for a steady state source, where  $R = H_{\max}/H_{\min}$  holds.

In order to maximize the intensity gain, according to Equ. (13), the spin-echo spectrometer has to be as short as possible. Considering 6 m shielding of the source, guide fields of about 3 m each plus polarizer, analyser and detector,  $L = 15$  m appears to be a minimum length of an actual spectrometer at SNQ. In this case we have  $\Delta\lambda = 2.7 \text{ \AA}$  and at  $\lambda = 3 \text{ \AA}$ , the flux enhancement would amount to 4.5. In addition, since no velocity selector is needed, there is another flux gain of 1.5. Thus,  $\phi_{\text{eff}} \approx 6.7 \bar{\phi}$ . The wavelength resolution  $\delta\lambda/\lambda = \Delta\tau/T_F = \Delta\tau v/L$  (pulse length  $\Delta\tau = 440 \text{ \mu s}$ ) amounts to about 4.0 %. With  $H_{\min} = 1 \text{ Oe}$  and  $H_{\max} = 1000 \text{ Oe}$ , the dynamic range would be  $t_{\max}/t_{\min} = 13\,000$ . All these numbers are substantially superior to what can be achieved at the ILL.

It is clear that the constant wavelength band of  $2.7 \text{ \AA}$  in this example becomes a less pronounced advantage around longer wavelength, e.g. at  $\lambda = 8 \text{ \AA}$  it is only about 2.5. Therefore it seems to be more advantageous to use only 50 pulse/sec ( $\tau = 20 \text{ msec}$ ).

Then  $\lambda_{\max} - \lambda_{\min}$  becomes  $5.3 \text{ \AA}$ . This leads to a 50 % loss of flux which is compensated by the twice broader wavelength band. The broader band also provides a more subtle advantage at long wavelength. As can be seen from Fig. 2, the lower  $t$  part, then is collected at shorter wavelength, where the neutron flux is higher (see ref. /3/).

Note that for instruments capable of using a wide wavelength band (e.g. time-of-flight diffraction, small-angle scattering, NSE) a 50 pulse/sec operation of SNQ appears to be more favourable.

Considering a 50 Hz SNQ with an average thermal flux of  $\bar{\phi}_{\text{SNQ}} = 1.2 \cdot 10^{15}$  n/sec cm<sup>2</sup>, 15 m moderator to detector distance, and 440  $\mu$ s pulse length for the cold moderator, table I compares the performance between ideal SNQ and ILL NSE machines.

TABLE I  
Performance of NSE Spectrometer at ILL and SNQ

|                                               | ILL                 | SNQ                       |                           |
|-----------------------------------------------|---------------------|---------------------------|---------------------------|
|                                               |                     | $\lambda = 5 \text{ \AA}$ | $\lambda = 8 \text{ \AA}$ |
| $\phi_{\text{eff}} [\text{n/cm}^2 \text{ s}]$ | $1.2 \cdot 10^{15}$ | $9.5 \cdot 10^{15}$       | $5 \cdot 10^{15}$         |
| dynamic range R                               | 1000                | 34 500                    | 7 900                     |
| Q-Resolution [%]                              | 20                  | 2,3                       | 1,5                       |

We note:

- (a) The dynamic range and the final resolution is a design parameter of the precession fields used in the NSE spectrometer, and the consequences of its choice is identical for both types of sources.
- (b) If  $\bar{\phi}_{\text{SNQ}}$  refers to 100 pulse/sec rate, then the flux figures in the table should be divided by 2, i.e. assuming that we suppress each second pulse.
- (c) In some cases better  $\Delta Q/Q$  resolution is required than the 20 % quoted for ILL in the table. This can only be achieved with a corresponding flux loss at the ILL, while high Q resolution is the natural on SNQ.

## V. CONCLUSION

We have presented a general outline for a spin-echo spectrometer at SNQ. TOF-NSE can utilize wide wavelength bands and gains from the time modulated beam structure of the SNQ. Similar to other wide band techniques like small angle scattering or time-of-flight diffraction it would profit from a lower frequency than 100 Hz. At 50 Hz it gains about an order of magnitude in effective flux compared to an identical steady state version. At the same time its Q resolution is about 10 times better.

In general, there do not seem to be any particular technical difficulties in adapting NSE for use in combination with time-of-flight technique.

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## Backscattering Spectrometer with Adapted Q-Resolution at the Pulsed Neutron Source

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J. Schelten, B. Alefeld, KFA - Jülich

### 1. Introduction

The backscattering spectrometer as installed in Jülich and Grenoble is known to serve the purpose of extremely high resolution neutron spectroscopy. The high energy resolution is achieved on the primary and secondary sides by means of perfect Si-crystals reflecting in the backward direction. The input energy is varied by Doppler shifting. A low Q-resolution is sufficient for the majority of problems involved and is ensured on the secondary side by large-area analyzing crystals. On the primary side, however, there is considerable mismatching due to the low beam divergence of  $\pm 0.01$  rad of the neutron beam guided in the beam tube.

A gain in intensity of about two orders of magnitude could be achieved by increasing the beam divergence so as to be adapted to that on the secondary side.

A method will be described in the following by which such adaptation can be effected at least in one direction. This leads to a gain in intensity by a factor of 15. The method can be applied with particular advantage at a pulsed neutron source.

## 2. Principle of the Modified Backscattering Spectrometer

The beam path is shown in Fig. 1. The neutrons are reflected on a moving imperfect crystal (K) with a mosaic width of about  $18^\circ$ . The crystal moves with a velocity of  $\sim 200$  m/s perpendicular to the reciprocal lattice vector on whose lattice planes the neutron beam is diffracted.

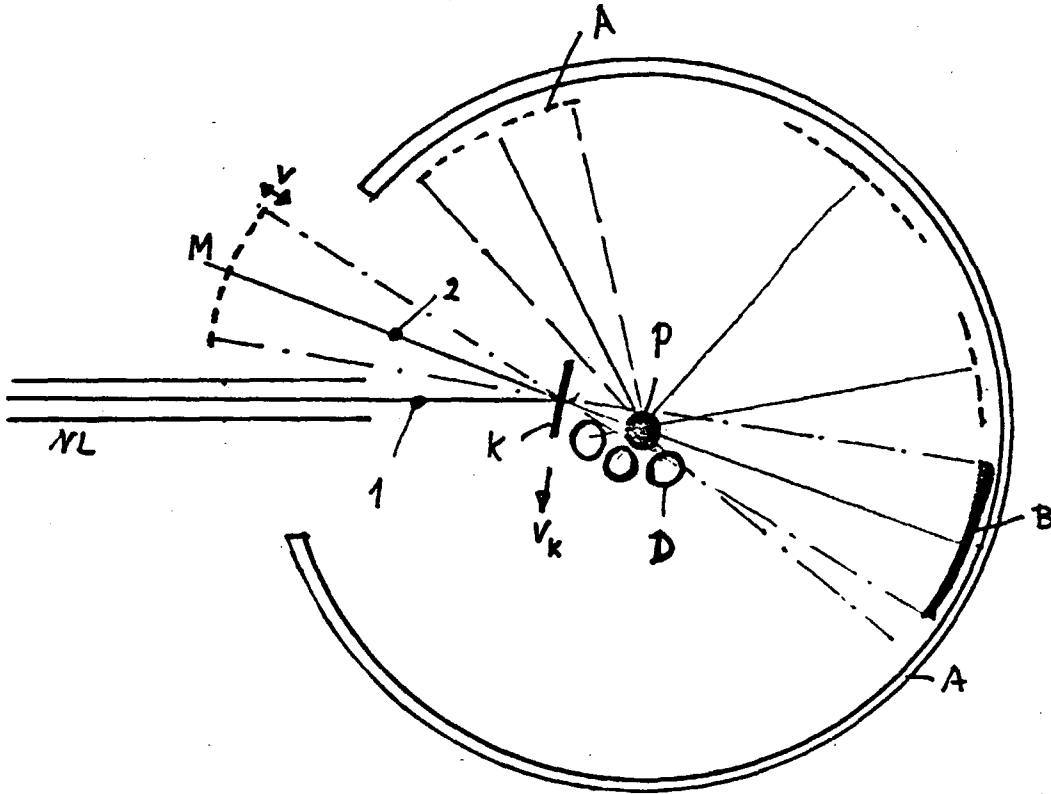


Fig. 1 Set-up of the backscattering spectrometer with moving crystal (K) at the end of a neutron guide (NL).  
P - sample, A - analyzer, M - monochromator, Doppler-moved, D - detector, A - shielding, B - beam stops.

The momentum space elements at 1 and 2 are shown in Fig. 2.

The Bragg angle is about  $75^\circ$ . The diffracted beam is therefore deflected from the backward direction by  $30^\circ$ . At a suitable

velocity of the crystal, a primary cigar-shaped resolution element is transformed into a secondary concavely curved resolution element as indicated in Fig. 2.

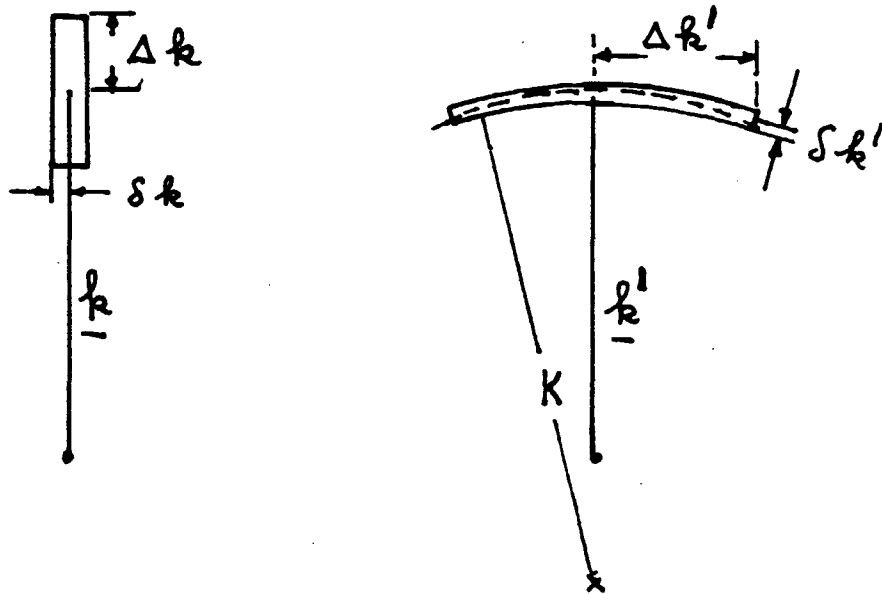


Fig. 2 Resolution element on the primary and secondary sides involving diffraction on the moving crystal.  $K$  is the radius of curvature of the resolution element after diffraction. The selection line of the monochromizing crystal is dashed.

The diffracted beam therefore leaves the moving crystal in the laboratory system with increased divergence and is relatively well monochromized ( $\delta k' \leq \Delta k'$ ). In contrast, the primary beam employed is well collimated with the neutron guide divergence  $\delta k = \eta \cdot k$ , although not monochromatic ( $\delta k \ll \Delta k$ ). The transformation of the moving crystal is, briefly speaking, from "white" to "wide". In the third dimension  $\delta k = \delta k' = \eta \cdot k$  is unchanged.

The divergent beam impinges upon perfect Si-crystals arranged on a cylinder segment (M in Fig. 1). They select an extremely narrow, but wide wave number band out of the secondary  $k'$  resolution element (broken line in Fig. 2) by means of backward

Bragg reflection. The crystals are Doppler-moved. The reflected and slightly Doppler-shifted beam impinges upon the sample of the backscattering spectrometer which from now on is unchanged. The moving crystal is not interspersed a second time by the reflected neutrons because it has meanwhile rotated out of the region at a peripheral velocity of about 200 m/s.

Nothing has changed with regard to the energy resolution and the energy range of the backscattering spectrometer. In terms of intensity, however, a gain of the order  $\frac{\Delta k'}{\delta k} \gg 1$  is achieved. The use of a pulsed source eliminates the need for coarse chopping with 1:1 pulse width repetition ratio, which doubles the gain. In addition, the crystal volume required can be kept smaller at the pulsed source.

### 3. Transformation of the Momentum Space Element by Means of the Moving Crystal

The transformation can be illustrated best in a drawing. The consecutive steps are shown in Fig. 3. The actual Bragg reflection takes place in the crystal system with the wave vectors  $\underline{k}^*$  and  $\underline{k}'^*$ , which are linked with the wave vectors  $\underline{k}$  and  $\underline{k}'$  in the laboratory system by the translation  $\underline{v}_K$ . The slope of the momentum space element after diffraction is obtained by an analytical treatment of the geometrical structure as:

$$(1) \quad \operatorname{tg} \chi = \frac{\operatorname{tg} \theta_0}{2 \operatorname{tg} \theta \cdot \operatorname{tg} \theta_0 + 1}$$

in which  $\theta_0$  is the Bragg angle in the laboratory system and  $\theta$  that in the crystal system. The latter is determined by the crystal velocity.

$$(2) \quad \underline{v}_K = v(k) \sin \theta_0 \left( \frac{1}{\operatorname{tg} \theta} - \frac{1}{\operatorname{tg} \theta_0} \right)$$

$\chi$  can be disposed of in such a way that the resolution element is perpendicular to  $\underline{k}'$ , i.e.

$$(3) \quad \chi = \pi/2 - \theta_0$$

Hence follows from (1) for  $\theta$ :

$$(4) \quad \operatorname{tg} \theta = -\operatorname{ctg} (2 \theta_0)$$

and from (2) with (4) the appropriate crystal velocity.

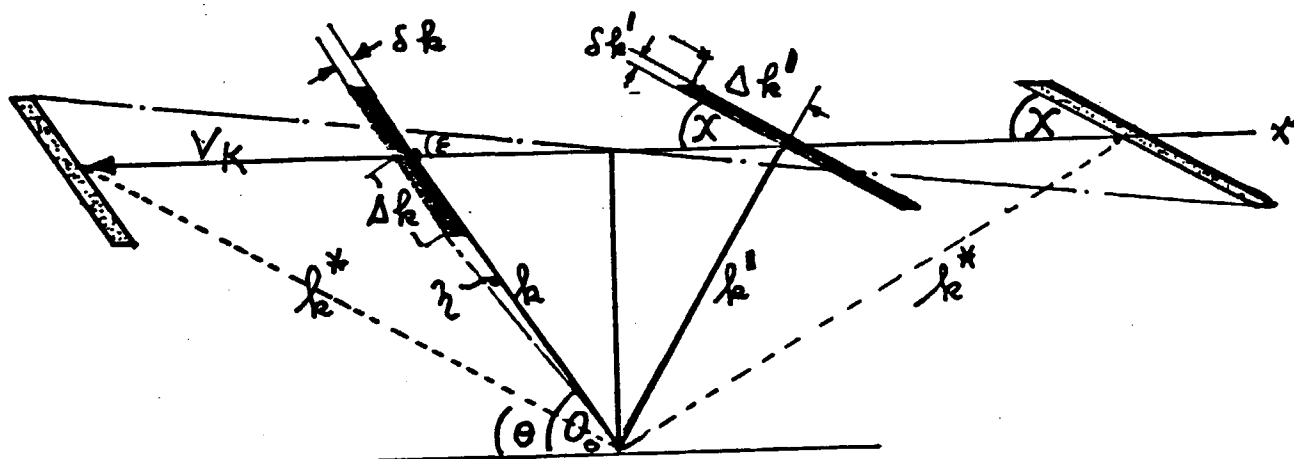


Fig. 3 Diffraction on the moving crystal; wave numbers of the incident and reflected waves are  $k, k'$  in the laboratory system and  $k^*, k^{*'} in the crystal system. The elements of  $k$  and  $k^*$  as well as  $k'$  and  $k^{*'} are always offset by the vector  $\pm v_K$ .  $\theta_0$  and  $\theta$  are the Bragg angles in the laboratory and crystal systems,  $\eta$  - primary divergence angle,  $\epsilon$  - half mosaic width. The concave curvature of the resolution elements after diffraction is not plotted.$$

The lengths and thicknesses of the momentum space elements are determined by mosaic distribution and beam divergence. They will be discussed in the following.

Momentum space elements, which have emerged from each other due to translation by  $v_K$ , are identical. The expansion of the elements in the  $x$ -direction is maintained during reflection.

The following is obtained on the basis of these two principles

| primary side                                                | secondary side                                                                           |     |
|-------------------------------------------------------------|------------------------------------------------------------------------------------------|-----|
| $\Delta k/k = \varepsilon  \operatorname{tg} (2 \theta_0) $ | $\Delta k'/k' = \varepsilon \operatorname{tg} \theta_0  \operatorname{tg} (2 \theta_0) $ |     |
| (5) $\delta k/k = \eta$                                     | $\delta k'/k' = \eta / \operatorname{tg} \theta_0$                                       | (6) |
| $\delta k / k' = \eta$                                      | $\delta k / k = \eta$                                                                    |     |

It may be seen that the momentum space volume is unchanged as must be the case according to Liouville.

The radius of curvature  $K$  of the concavely curved momentum space element was ascertained by computer and by drawing.

A narrow wavelength band of the width  $\Delta k_M$  is selected out of the momentum space element of  $k'$  by the Si-monochromator arranged on a cylinder segment (see Figs. 1 and 2).  $\Delta k_M$  is described, on the one hand, by

$$(7) \quad \frac{\Delta k_M^2}{2k'} = \frac{\Delta k_M^2}{2k} + 2 \delta k'$$

while on the other hand, the condition

$$(8) \quad \Delta k_M \leq \Delta k'$$

must be fulfilled.

Equation (7) assumes that the  $k$ -vector for backward reflection on the Si-monochromator crystal ( $k_M$ ) is adapted to the momentum space element of  $k'$ , i.e.

$$(9) \quad k_M = k' + \delta k' = k' \left(1 + \frac{\eta}{\operatorname{tg} \theta_0}\right)$$

The solution of (7) for  $\Delta k_M$  gives

$$(7) \quad \frac{\Delta k_M}{k'} = 2 \sqrt{\eta} / \sqrt{\operatorname{tg} (\theta_0) \cdot (1 - k'/K)}$$

Due to this adaptation the reciprocal lattice vectors  $\underline{\tau}_K$  of the moving crystal and  $\underline{\tau}_{Si}$  of the monochromizing crystal must be tuned to each other. The following must be applicable according to the Bragg equation:

$$(10) \quad \frac{1}{2} \frac{\tau_K}{\sin \theta_0} (1 + \eta |\operatorname{tg} \theta_0|) = \frac{1}{2} \tau_{Si}$$

i.e.  $\underline{\tau}_K$  must be

$$(11) \quad \tau_K = \tau_{Si} \frac{\sin \theta_0}{1 + \eta / \operatorname{tg} \theta_0}$$

The  $\theta_0$  dependence of all these quantities is specified in Table 1 which will be discussed in the following.

#### 4. Parameters of the Backscattering Spectrometer with Moving Crystal

The Bragg angle  $\theta_0$  for the reflection from the moving crystal in the laboratory system can be freely chosen. According to (4) the Bragg angle  $\theta$  in the crystal system is then given. The changed Bragg angle is a consequence of the motion, the velocity being determined by (2) on the assumption of  $v(k) = v(k')$  = 637 m/s. The expansions of the momentum space elements of  $k'$  and  $k$  result from (5) and (6). The radius of curvature of the element of  $k'$  was taken from the results of numerical calculations.

The pulse width of  $k_M$  results from (7), whereas (8) provides information on the mosaic width  $\epsilon$ .  $\eta$  then has been disposed of. The crystal adaptation (11) leads to the reciprocal lattice vectors  $\underline{\tau}_K$  as specified.

The first three columns are of greatest interest. The Bragg reflection leads to a sufficient beam separation. The velocities are manageable. The width of the processed monochromator momentum space element  $k_M$  is large if the mosaic width is  $\geq 4^\circ$ . This gives rise to an intensity gain of 10 - 18.

|                                       |                       |                      |                       |                      |                      |                    |                      |
|---------------------------------------|-----------------------|----------------------|-----------------------|----------------------|----------------------|--------------------|----------------------|
| $\theta_0$                            | 85                    | 80                   | 75                    | 70                   | 65                   | 60                 | 55                   |
| $\theta$                              | 80                    | 70                   | 60                    | 50                   | 40                   | 30                 | 20                   |
| $v_K/v'_n$                            | 0.089                 | 0.185                | 0.299                 | 0.446                | 0.657                | 1.0                | 1.68                 |
| $v_K$ [m/s]                           | 56                    | 177                  | 190                   | 284                  | 419                  | 637                | 1068                 |
| $\Delta k'/k$                         | $2.02 \cdot \epsilon$ | $2.1 \cdot \epsilon$ | $2.15 \cdot \epsilon$ | $2.3 \cdot \epsilon$ | $2.6 \cdot \epsilon$ | $3 \cdot \epsilon$ | $3.9 \cdot \epsilon$ |
| $\delta k'/k'$                        | $0.087\eta$           | $0.176\eta$          | $0.27\eta$            | $0.36\eta$           | $0.47\eta$           | $0.58\eta$         | $0.7\eta$            |
| $K/k'$                                | 1.4                   | 1.44                 | 1.55                  | 1.75                 | 2.2                  | 3.0                | 5.3                  |
| $\Delta k/k$                          | $0.18\epsilon$        | $0.36\epsilon$       | $0.58\epsilon$        | $0.84\epsilon$       | $1.2\epsilon$        | $1.7\epsilon$      | $2.7\epsilon$        |
| $\Delta k_M/k$                        | $1.11\sqrt{\eta}$     | $1.52\sqrt{\eta}$    | $1.73\sqrt{\eta}$     | $1.84\sqrt{\eta}$    | $1.85\sqrt{\eta}$    | $1.86\sqrt{\eta}$  | $1.86\sqrt{\eta}$    |
| $\epsilon(\eta=0.01)$                 | $\geq 4.2^\circ$      | $\geq 4.6^\circ$     | $\geq 4.6^\circ$      | $\geq 4.1^\circ$     | $\geq 4.1^\circ$     | $\geq 3.6^\circ$   | $\geq 2.7^\circ$     |
| $\Delta k/k$                          | $\geq 0.01$           | $\geq 0.026$         | $\geq 0.040$          | $\geq 0.067$         | $\geq 0.096$         | $\geq 0.107$       | $\geq 0.13$          |
| $\tau_K/\tau_{Si}$                    | 0.996                 | 0.983                | 0.963                 | 0.936                | 0.903                | 0.863              | 0.814                |
| $G = \frac{\Delta k_M}{k \cdot \eta}$ | 11                    | 15                   | 17                    | 18                   | 18                   | 19                 | 19                   |

Table 1: Parameters and results during reflection on the moving crystal and monochromator installed behind.

A perfect crystal adaptation can be achieved with Ge-crystals. Germanium and silicon both have a diamond structure with  $a = 5.658 \text{ \AA}$  and  $5.43 \text{ \AA}$ , i.e.  $a_{\text{Ge}}/a_{\text{Si}} = 0.9597$ . This ratio is very close to the case of  $\theta_0 = 75$  in Table 1 for which the calculated gain in intensity is 17.



## 5. Requirements with Regard to the Moving Crystal

An orientation change of the crystal and a velocity change during its motion will tilt the momentum space element towards its desired position.

### Crystal tilting

Tilting must be small versus the mosaic width  $\epsilon$  so that its influence may be neglected. This requirement can be easily satisfied in the case of large mosaic angle.

### Velocity inhomogeneity

If the crystal is mounted on a rotating disc, the crystal velocity  $v_K = R \cdot \omega$  is position-dependent. The change in velocity  $\delta v_K$  leads to orientation changes  $\delta\chi$  which are calculated from (1), (2) and (3).

$$(12) \quad \delta\chi = 2 \cdot \cos^2 \theta_0 \cdot \tan^2 2 \theta_0 \cdot \sin^{-1} \theta_0 \cdot \frac{v_K}{v(k)} \cdot \frac{\delta v_K}{v_K}$$

An admissible change is  $\delta \leq 0.1 \cdot \delta k' / \Delta k'$  resulting in a velocity homogeneity of

$$\delta v_K / v_K \lesssim 0.01$$

In order to synchronize the crystal motion with the neutron pulses, the frequency of the wheel must be 100 Hz or an integral multiple thereof. In this case, one crystal at the periphery will be sufficient. Two and three crystals are required for the rotational frequency of 100/2 Hz and 100/3 Hz, respectively. In order to attain the velocity of 190 m/s required for  $\theta_0 = 75^\circ$ , the distance  $R$  of the Ge-crystal from the axis of rotation must not exceed 30 cm at 100 Hz. For a 6 cm high crystal this means a velocity inhomogeneity of  $\pm 10\%$  which cannot be tolerated. More likely to be acceptable is  $\omega/2\pi = 33.3 \text{ sec}^{-1}$  and  $R = 90 \text{ cm}$  with three crystals at the periphery. In this case  $\frac{\delta v_K}{v_K} = \pm 3\%$ .

### Rotating wheel with space-fixed crystal

An alternative is a wheel with the crystal itself rotating opposite in direction about its own axis with equal frequency. This keeps the crystal orientation space-fixed, its velocity in the x-direction being

$$v_K = R \cdot \omega \cdot \cos \alpha$$

It will then remain within the defined tolerance for a 3 cm wide reflection range and  $R = 30$  cm. The velocity must be adjusted precisely within 1 %. Since the frequency is fixed at 100 Hz due to the synchronization with the neutron pulses, adjustment can only be effected via the rotating arm  $R$ .

### Crystal thickness

A crystal thickness of a few mm is sufficient for reflecting the neutrons on the imperfect Germanium crystal with  $\pm 4^\circ$  mosaic width at a Bragg angle of  $= 60^\circ$ .

### Crystal width

The crystal width in the direction of motion is determined by the time  $\delta t$  during which reflection must take place. The wave number band to be reflected is  $2 \Delta k/k$ . The transit time of the neutrons from the source to the crystal is  $t_0 = L/v(k)$ . This gives rise to a reflection time of  $\delta t = \frac{2\Delta k^0}{k} \frac{L}{v(k)}$  and a crystal width of  $b = \delta t \cdot v_K$ .

A distance of  $L = 20$  m in conjunction with the values from the third column of Table 1 results in  $b = 50$  cm. This crystal volume, which is not particularly small, is considerably reduced when placing the spectrometer at a distance of only 10 m from the source and adjusting the conditions of the first or second column in Table 1 with  $\theta_0 = 85$  or  $80^\circ$ . The crystal width will then only be 10 cm and less. This can be realized using intercalated graphite as the moving crystal.

## 6. The Moving Crystal as a Monochromator

For high-resolution, although not extremely high resolution, the moving crystal can be used directly as a neutron monochromator. It may serve to reach the range of high energy resolution with low Q-resolution which is otherwise scarcely accessible. Values which can in part be taken from Table 1 and are complemented by others from Table 2 reveal that an energy resolution with  $\delta k'/k'$  of  $1 \cdot 10^{-3}$  is achieved with the neutron guide divergence and that an energy resolution with  $\delta k'/k$  of  $1 \cdot 10^{-4}$  can be attained in the case of additional collimation with  $\eta = 1 \cdot 10^{-3}$ . The divergence of the beam is still wide:  $\Delta k_M/k' \sim 0.05$ .

| $\theta_0$<br>(°) | $\theta$<br>(°) | $\epsilon$<br>(°) | $\eta$<br>(rad) | $\Delta k_M/k'$<br>(/) | $\delta k'/k'$<br>( $10^{-4}$ ) |
|-------------------|-----------------|-------------------|-----------------|------------------------|---------------------------------|
| 85                | 80              | $\geq 3.2$        | 0.01            | 0.11                   | 8.7                             |
| 80                | 70              | $\geq 4.2$        | 0.01            | 0.15                   | 18                              |
| 85                | 80              | $\geq 1$          | 0.001           | 0.035                  | 0.87                            |
| 80                | 70              | $\geq 1.3$        | 0.001           | 0.047                  | 1.8                             |

Table 2: Parameters for the moving crystal as a monochromator,  $\epsilon$ ,  $\eta$ ,  $\Delta k_M$  and  $\delta k'$  are half mosaic and divergence angles, half pulse widths and resolutions.

## 7. Summary

- Using a moving crystal in front of the Si-monochromator, the intensity can be enhanced by a factor of 10 - 18 without impairing the resolution of the backscattering spectrometer (see Table 1).
- The coarse chopper with a pulse width/repetition ratio of 1:1 can be omitted at a pulsed source, which means an additional gain by a factor of 2.
- The expenditure on crystals is not excessive if the spectrometer is placed near the pulsed source.
- The crystal motion at velocities up to 200 m/s does not involve much technical effort.
- Crystals adapted to the problem are Germanium and intercalated graphite crystals.
- The moving crystal can be used directly for high-resolution spectroscopy in the range of  $\delta k'/k' = (1-10) 10^{-4}$ . The momentum space element is still very wide in this case, amounting to  $\Delta k_M/k' \approx 0.05$ .
- The Bragg reflection on a moving crystal transforms a momentum space element of a collimated non-monochromatic beam into a momentum space element of a divergent, but narrowly monochromized reflected beam. The transformation is, briefly speaking, from "white" to "wide".
- The separation between primary and secondary beam is sufficient; between  $10^\circ - 30^\circ$  for  $(\pi - 2\theta_0)$ .

W. Schmatz, KfK Karlsruhe

### Summary of the SNQ-Workshop Special Techniques

Major participants at the two workshop-meetings in Bad Kissingen and Maria Laach have been Alefeld, Badurek, Conrad, Rauch, Steichele, Steyrl, Williams and myself (Schmatz) as chairman, occasional participants have been Bauer, Kley, Kollmar, Prandl, Taylor, Schelten and Windsor. At first we have to thank the organizers (KFA Jülich assisted by Scherm) for the opportunity to spend on an approximately 30 man-days effort (in total) our time for relaxed brain-storming. We tried to follow Scherm's proposal to trace every crazy idea, which may come up. Of course most has had to be put in the trash and more seriously most of what has been said could not be pushed to a point, where I would dare to write it down. Even more, one of the participants reminded me to the fact that there is an absolute danger to interfere with personal interests of researchers by giving written versions on subjects which are just in development by hard efforts. Therefore in opposition to Scherm's idea, that the participants should write many pages on their ideas, this has not been urged by me. Nevertheless there are some written versions. Alefeld, Bauer, Kollmar and Schelten have contributed mainly with elegant ideas for moving crystals and collimators. Badurek and I made a proposal on a time-shaped analyzer. Steyrl made a critical consideration on the possible use of ultra-cold neutrons at pulsed sources.

After this lengthy but necessary introduction I would like to make one general statement: There are many possible ways to come near to the ultimate gain factor  $\hat{\phi}/\bar{\phi}$  at least in a considerable fraction. This means with  $\bar{\phi} \approx 10^{15}$  n/sec cm<sup>2</sup> as planned for the SNQ gain factors ranging from 30 to 1000 in the energy range from 0 to 300 meV are at least thinkable compared to the Grenoble high flux reactor. (I anticipate the

final SNQ with compressor ring.) For  $E > 300$  meV no discussion is necessary in comparison to the Grenoble reactor. The SNQ is the best.

I would like to give a special example: According to a very careful study of the SNQ-workshop group on inelastic scattering (chaired by Reichardt) the conventional triple axis spectrometer hardly has an advantage from the flux peaking. However there might be in future multilayer crystals available with  $\tau \approx 0.1 \text{ \AA}^{-1}$  and  $\Delta\tau/\tau \approx 0.1$  which allow total reflection of a broad band of cold or thermal neutrons. A focussing mirror composed of such multilayer crystals with a length of about 40 m and a height of about 6 m would image the source into a 200 m distant sample, whereby the energy of the incident beam is determined by time-of-flight. The collimation conditions would be the same (vertical and horizontal) as for a conventional triple axis spectrometer at a continuous reactor. The perfection required for the multilayer crystals and the price of such a device is beyond to-day's possibility, but the instrument would beat the conventional triple axis spectrometer.

Another example: Alefeld has reminded us on the possibility of shifting phase space elements ( $\Delta k_0$ ) from low  $k_0$ -values to high  $k_0$ -values. In  $180^\circ$ -scattering (for simplicity) a crystal moving towards the beam with velocity  $v_0$  (not too small) adds a velocity  $2v_0$ . The pulsed source offers the advantage to hit the neutron just at the time where the crystal is in the proper position. In principle the process can be repeated periodically. The problem is to find or to produce the proper set of crystals. Why not! The phase space shifting may be of advantage already at a continuous source. The additional advantage at the pulsed source may approach near to  $\hat{\phi}/\bar{\phi}$ .

Example number three: The magnetic whiggler invented by Drabkin (mirror, spin flipper,  $B_0, B_1$ -spin flip resonance filter, mirror) is a device, which allows to adjust the transmitted energy ( $\Delta\lambda/\lambda \approx 1 \dots 10\%$ ) in rapid time variation (Badurek and Schmatz, submitted to Z. f. Physik). Therefore it would be of special advantage at pulsed sources in inverse time-of-flight geometry. A whole set of such analyzers (similar to the improved D7 with supermirrors) around a sample is thinkable. With to-day techniques the time-adjusted whiggler would make diffuse elastic scattering at the SNQ clearly superior to an instrument at the ILL. With "tomorrow" techniques the magnetic whiggler analyzer could be of interest also for inelastic scattering. The magnetic analyzer is compact. Many of them can surround the sample also for inelastic scattering.

Example number four: Very cold neutrons ( $\lambda = 30 \dots 300 \text{ \AA}$ ) are of principal advantage for inelastic small angle scattering as a simple calculation shows (Steyrl and Schmatz, unpublished). If high energy resolution is required, pulsing is of advantage also for these very slow neutrons. The advantage compared to a small-angle spin echo instrument at a continuous source is the full record even of weak intensities in the time-of-flight spectrum.

The four examples above are illustrating for a whole series. They demonstrate that even a short time of thinking offers new possibilities. We considered also seriously magnetic neutron accelerators, but came to no conclusion. By some remarks from Rauch it seems to be clear however that filling storage rings with neutrons clearly has advantages at pulsed sources. An overwhelming wide variety for the use of moving crystals at pulsed sources seems to exist. Oscillating crystals have been shown to be a real hit in combination with pulsed sources. The next step is to ask why not also oscillating collimators and finally why not an oscillating sample. This is not a joke! With more degrees of freedom the ultimate gain in quality is higher.

We also looked at simple things. Experienced scientists as Taylor, Williams and Windsor convinced us that epithermal neutron background can be mastered at pulsed sources to such a degree that the signal-noise ratio can be pushed to values much lower than for continuous sources. For new ideas in an integrated view we looked also at the source properties. With no doubt the SNQ-concept with choppers near the source is an optimal one in today's technique. Nevertheless it might be possible that magnetic devices or fluid-dynamical ones do a better job than choppers. Also the optimization process for the source itself seems not to be finished. The rotating wheel seems to be ultimate but the moderator philosophy should not be taken as a final one. With focussing devices and for small samples (area  $\leq 10 \text{ cm}^2$ ) in principle one can extract neutrons from smaller source areas. This possibly would allow to install a larger variety of sources along the neutron production zone of the rotating wheel.

Let's try a conclusion: With  $\bar{\phi}$  equal to the value of the HFR Grenoble and ratios  $\hat{\phi}/\bar{\phi}$  ranging from 30 to 1000 in the energy range from 0. to 300 meV I think the average gain factor of about 10, which was evaluated by the other groups of the workshop is a lower limit due to the boundary condition to be honest. The SNQ with compressor ring however is a phantastic source which is in full operation just before the celebration of the year 2000. There is sufficient time for more new ideas and - more important - for hard efforts in corresponding technology. I think the today average gain factor of about 10 is too pessimistic. In my opinion it will move towards 20 to 40 for a broad variety of instruments. In addition there is the new field for  $E_0 > 300 \text{ meV}$ . The situation may be similar to what I experienced in neutron small angle scattering. At 1963 an X-ray small angle scattering specialist looked at our Munich efforts on neutron small angle scattering. At this time resolution and count rate were not very encouraging for a



broad application of this technique and he honestly discussed this point with us. With the HFR Grenoble and a lot of technological efforts latest at about 1980 neutron small angle scattering was a world-wide hit. It is no question to me that a similar situation may arise for the SNQ with compressor ring.

# VERY SMALL ANGLE SCATTERING BY MEANS OF A PERFECT CRYSTAL CAMERA

H.Rauch, Atominstitut der Österr. Universitäten, A-1020 Wien, Austria

D.Schwahn, Institut für Festkörperforschung, KFA, D-5170 Jülich, FRG

## 1. Introduction

The width of the rocking curve of a nondispersive arrangement of perfect crystals is in the order of the Darwin reflection width; i.e. it is in the order of seconds of arc. A sample placed in between causes a broadening of the rocking curve which is directly related to the small angle diffraction pattern. It becomes also popular for neutron small angle scattering to measure very small angular deflections /1,2/, the diffraction pattern of large precipitates or voids /3 - 8/ and the diffraction of magnetic domains or walls /9 - 17/. The angular or momentum ( $Q$ ) resolution is considerably better than in the case of conventional small angle scattering facilities where a long path length is used to achieve the desired resolution (e.g. /18/). The  $Q$ -range which is accessible by this technique is in the order of  $10^{-5} - 10^{-3} \text{ \AA}^{-1}$ . Intrinsically it is a one-dimensional method and relates therefore to the slit geometry often used in X-ray small angle scattering (e.g. /19/). From a quantitative X-ray comparison of the intensities between the conventional and the perfect crystal small angle scattering systems it can be concluded that the perfect crystal system become superior for the investigation of objects or interparticle distances larger than  $7100 \text{ \AA}$  /20/, which can be shifted down to about  $960 \text{ \AA}$  if the resolution is intentionally reduced /21/.

## 2. Motivation

The main motivation to reach the small  $Q$ -range below  $10^{-3} \text{ \AA}^{-1}$  is based on the interest to study large biological macromolecules and the correlation of particles over large distances. Such corre-

lations exist between various components of a biological material /22/, between superparamagnetic precipitates /23/ and inside of lamella and membrane structures. The correlation term yields to a scattering maximum at  $Q \neq 0$  and to a strong  $Q$ -dependence at small  $Q$ -values (e.g. /18/). An example of such behavior is shown for an Al-Al<sub>2</sub>Cu lamella structure with a mean lattice constant of 1.4  $\mu\text{m}$  in Figure 1 /24/. Furthermore, the study of critical phenomena and phase transformations in polymer blends are of great interest /25/. The perfect crystal camera measures in the same  $Q$ -range as the light scattering technique. The advantage of the neutron instrument with respect to the light scattering technique is

- simple scattering law because the first Born approximation is fulfilled in most cases
- large specimen volumens can be used avoiding effects from the walls
- opaque specimens can be studied
- contrast variation method can be applied.

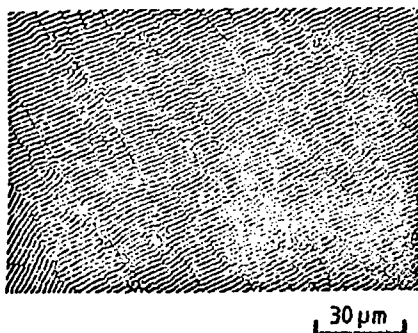
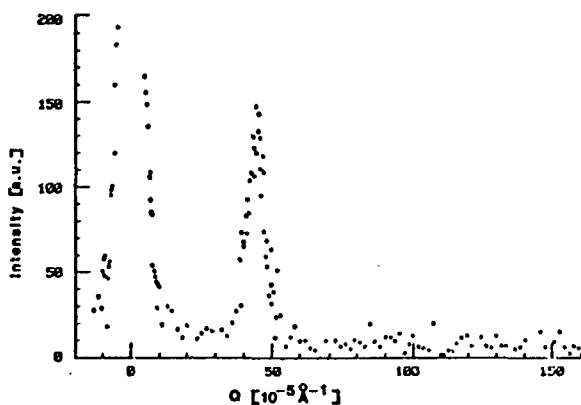


Figure 1: Microscopic view and small angle scattering pattern of a lamella structure



It has to be mentioned that the  $Q$ -resolution of the double crystal small angle camera is strongly decoupled from the wave-length width and the divergency of the incident beam. This is an analogy to the decoupling of the energy resolution from the beam parameters in spin echo spectrometry /26/. The shortness of such devices ( $\sim 1\text{ m}$ ) is an additional

advantage because for any real-time experiment the problem of frame overlap is strongly relaxed compared to conventional small angle scattering facilities. For the situation at the SNQ a time scale

for real-time experiments down to about 10  $\mu$ s can be reached if the relevant external sample parameter - magnetization, pressure, temperature, composition - can be changed rapidly, periodically and synchronously to the neutron bursts. In this case a band width of  $\Delta\lambda/\lambda \sim 2\%$  can be accepted. The pulsed structure of the source can be used advantageously for real-time investigations up to times of about 20 times the pulse repetition time (SNQ:  $20 \times 10$  ms = 0.2 s). In this case flash pictures of the relaxation phenomenon are taken instead of time averaged pictures taken over much longer time intervals. In proper cases the gain of information which can be achieved is in the order of the ratio of the pulse repetition time to the pulse length (SNQ:  $10$  ms/ $0.3$  ms = 33). This time scale between 10  $\mu$ s and about 1 s may be also of interest for the investigation of various fast chemical reactions.

### 3. Intensity consideration

We start with the intensity observed behind the neutron guide and the perfect silicon crystal at ILL which was  $5.6 \times 10^3$  cm<sup>-2</sup>s<sup>-1</sup> at a wave length of about 1.8 Å /27/. For a 10% scattering sample one expects behind the analyzer at the SNQ an intensity of about 50 cm<sup>-2</sup>s<sup>-1</sup> if the scattered intensity is concentrated predominantly in the very small angle scattering regime (10 times the Darwin width of the crystals). This can be increased putting up to penalty of Q-resolution by increasing the reflection width of the crystals, which can be achieved by using a perfect crystal with a temperature gradient /28/, or a curved crystal /29/ or by cutting the crystal in a Fankuchen shape /30,1/. For all three cases the intensity gain times the loss in resolution remains constant. In the first two cases the additional advantage exist that the contribution of the tails of the reflection curves is reduced by about the same member as the width of the reflection curve is increased. In the case of a Fankuchen cut an additional advantage occurs due to the spatial concentration of the intensity at the sample position and, therefore, smaller samples can be used. The Darwin width and, therefore, the width of the double crystal rocking curve (resolution function) depends on the angle  $\chi$  between the reflecting lattice planes and the surface as

$$\Delta\theta = \frac{b_c |F| N \lambda^2}{4\pi \sin 2\theta_B} \sqrt{\frac{\sin(\theta_B - \chi)}{\sin(\theta_B + \chi)}} = \Delta\theta_0 \sqrt{\frac{\sin(\theta_B - \chi)}{\sin(\theta_B + \chi)}} \quad (\chi > \pi/2)$$

where  $b_c$ ,  $|F|$ ,  $N$  are the coherent scattering lengths, the geometrical structure factor and the particle density of the silicon crystal and  $\lambda$  is the neutron wave length and  $\theta_B$  is the Bragg angle. The angular width and the intensity increases by the factor  $\sqrt{\sin(\theta_B - \chi)/\sin(\theta_B + \chi)}$  and the beam cross section reduces by the same factor. Including this possibility the small angle scattering intensity at the SNQ in the very small angle regime is estimated to about  $500 \text{ cm}^{-2} \text{ s}^{-1}$ .

#### 4. Proposed lay-out

In order to achieve an effective use of the available neutron flux we propose a multiple spectrometer system as it is shown in Figure 2. Using such a system a stereoscopic view of the sample

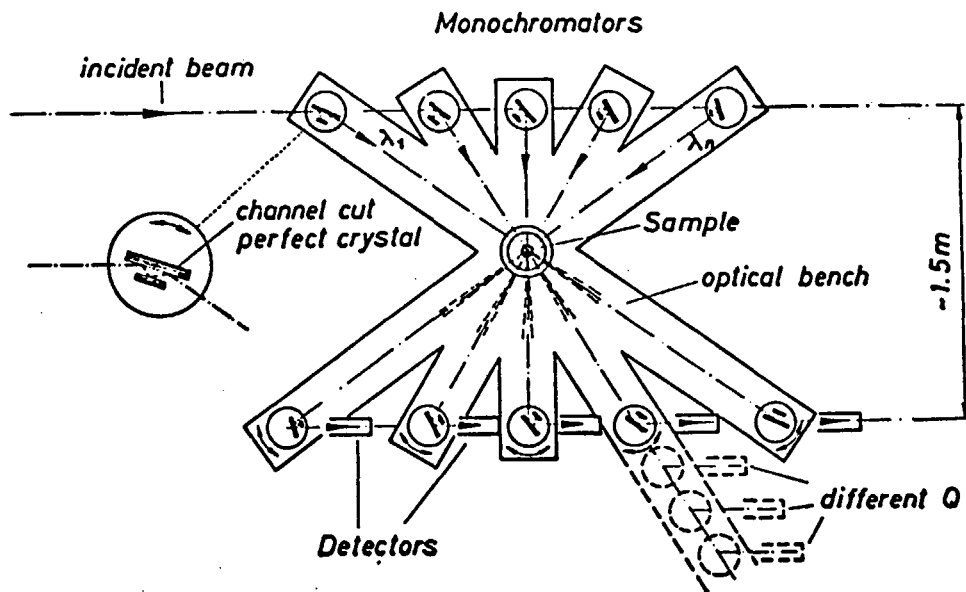


Figure 2: Proposed multiple double crystal small angle scattering facility

and a simultaneous measurement at different Q-values become feasible. The monochromator and analyzer part should be equipped with monolithically designed channel-cut crystals to reduce the tails of the reflection curves /31, 32,24/. Recent neutron tests of such crystals have shown the reduction of the tails but it was less than expected which is attributed to contributions from thermal diffuse scattering and, therefore, the use of thin crystal plates is recommended to reduce this contribution.

The whole apparatus should be equipped for real-time experiments and an optical adjustment system should be included to reduce the time needed for neutronic adjustment.

In future the small angle scattering of polarized neutrons may become of greater interest (e.g. /33/). For cases where the scattered intensity is strongly peaked in the forward direction the double crystal small angle scattering facility can easily be equipped with a polarization supplement by means of prismatically shaped magnetic fields between the monochromator and the sample /34,35/. In order to achieve a sufficient separation, a multistage system will be needed to get an angular separation of the + and - component which is larger than the small angle diffraction pattern. The separation depends on the magnetic induction B, the angle of the magnetic field prism  $\phi$ , its deviation  $\epsilon$  from the symmetric position and from the energy E of the neutron as

$$\delta \simeq 2n \frac{\mu B}{E} \frac{\sin \phi}{\cos \phi + \cos 2\epsilon}$$

where n is the number of stages. The spin-up and spin-down diffraction pattern can be measured simultaneously by using a multistage analyzer system. The number of stages needed will be larger than 20 which can be made of a single proper shaped permanent magnet system.

The sample environment should be designed to permit a variation of the temperature, of the magnetic field, of the pressure and of the chemical composition of the sample.

## 5. Conclusion

To extend neutron small angle and critical scattering to the very small  $Q$ -range a multiple nondispersive double crystal spectrometer is proposed. This instrument will be valuable for the investigation of very large objects, interparticle interference phenomena, and a stereoscopic view of the sample. Distinct advantages exist for real-time experiments for time scales down to about  $10\ \mu\text{s}$ , which is a time scale of great interest for the dynamic behavior of large object and interparticle relaxation. In comparison to steady state sources a gain factor for real-time experiments of 35 is estimated for relaxation phenomena in the time scale  $10\ \mu\text{s} \leq \tau \leq 300\ \mu\text{s}$ , of about 30 for the time scale  $10\ \text{ms} \leq \tau \leq 200\ \text{ms}$ . For longer time scales the gain factor approaches 1.

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## NEUTRON INTERFEROMETRY AT A PULSED SOURCE

H. Rauch, Atominstitut der Österreichischen Universitäten,  
A-1020 Wien, Austria

## 1. Introduction

Motivated by the success of X-ray interferometry /1,2/, neutron interferometry has been developed since 1974 /3,4/. The most effective technique uses monolithic perfect silicon crystals which serve as coherent beam splitter, deflector and phase analyzer. The coherent beams are widely separated and can be influenced separately. Most topics treated by this technique up till now belong to neutron optics and to fundamental physics. Characteristical applications are: the precise measurement of scattering lengths /5,6/, the detection of hydrogen in metals /7/, the verification of the  $4\pi$ -symmetry of spinor wave functions /8,9/, the spin superposition /10,11/ and the observation of the influence of gravitational forces /12,13/. By means of this technique the phase of the wave function becomes a new measurable quantity which can be used for nuclear, solid state and fundamental physics experiments. This new characterization of the beam stands beside the characteristic quantities used up till now: the intensity, the energy and the polarization. It can be modulated in time and space as well.

## 2. Motivation and proposed layout

The reason to propose neutron interferometry to a pulsed spallation source is at least twofold. First of all, the epithermal regime is favored at these machines and the time structure permits a simultaneous measurement at different energies. These

measurements can give new information about the energy dependent phase change which is described by an energy dependent scattering amplitude and can serve as an additional tool to investigate the density of phonon states /14/. Related measurements on  $^{149}\text{Sm}$  at a stationary source have been extended up to an energy of 0.092 eV whereas for a spallation source (SNQ) the energy range up to 1.7 eV can be covered. Figure 1 shows in a most simple and schematic manner such an arrangement.

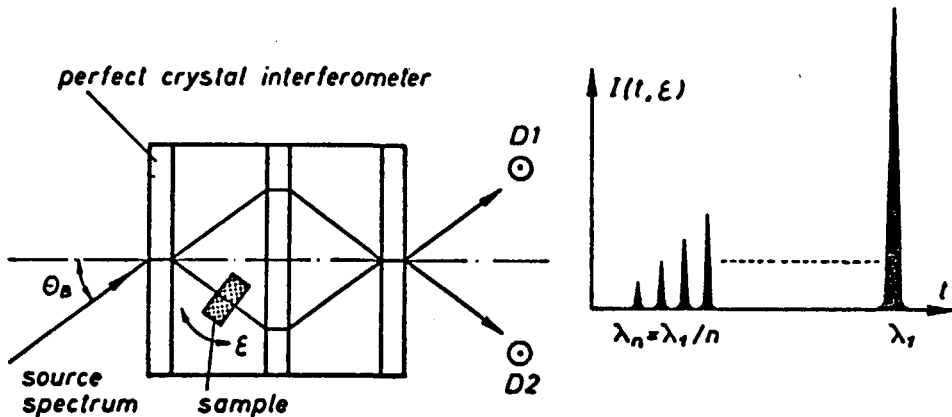


Figure 1: Schematic arrangement of an interferometer set-up at a pulsed source. The measuring principle is indicated at the right side

The optical path length can be varied by rotating the sample and the measurable wave length range can be varied by changing the Bragg angle. Due to the time structure of the source the orders can be measured simultaneously. Starting with the intensity at the interferometer set-up at Grenoble /15/ which amounts at the detector  $1.3 \times 10^3 \text{ cm}^{-2} \text{ s}^{-1}$  and including the flux and reflectivity decrease at shorter wave length  $(\phi(\lambda_1) \exp(-W(\lambda_1)) \lambda_1^2 / \phi(\lambda_2) \exp(-W(\lambda_2)) \lambda_2^2)$  one gets reasonable intensities and statistical accuracies of about 1% up to the 25th diffraction order within a measuring time of about 5 hours. When the first order neutrons are chosen to have an energy of 0.057 eV ( $\lambda = 1.2 \text{ \AA}$ ) the accessible energy range is up to 1.7 eV.

Knowing the wave length dependence of the real and imaginary part of the scattering amplitude a unique phase determination in structure analysis on the basis of Bijvoet ratios become feasible.

The main candidates for such measurements are  $^{113}\text{Cd}$ ,  $^{149}\text{Sm}$  and  $^{157}\text{Gd}$ . A related phase structure analysis für Cd-histidine performed at a stationary source has shown the capacity of this new method /15/. It may become an important new tool for structure analysis at a spallation source where even shorter wave lengths are available.

The second type of experiments which become feasible at a spallation source belong to inelastic scattering. Instead of a plane wave now two coherent plane waves from different directions (same  $|k|$ ) and variable phase relation are incident on the sample placed at the position of the third interferometer plate (Fig. 2).

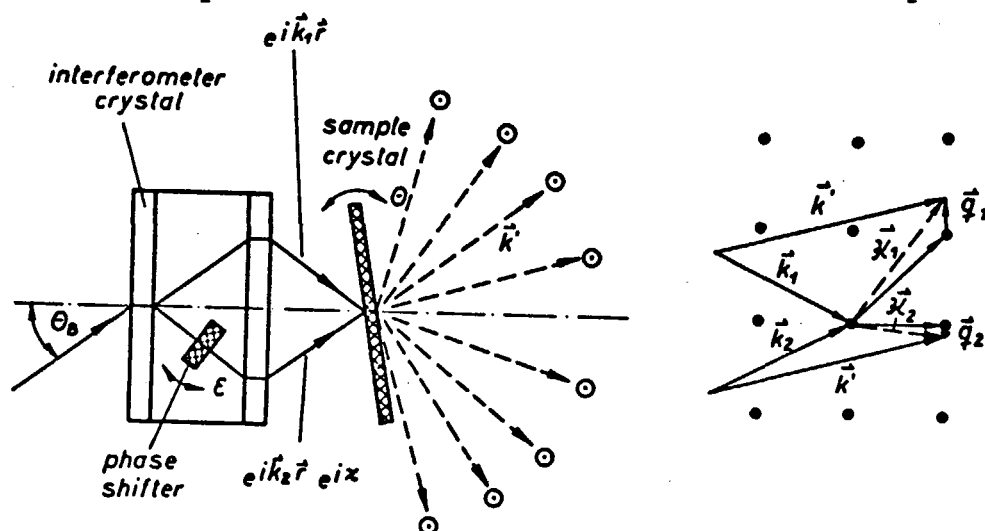


Figure 2: Schematic arrangement of an inelastic scattering experiment using two coherent incident beams

The sample is no longer monolithically connected to the other part of the interferometer crystal but has to be adjusted very precisely to this part. From the inelastic scattering pattern one can extract higher order correlation functions which will give more information about collective phenomena in condensed matter physics. The limitation to perfect crystals will be less restrictive if in future more crystals can be grown with a high prefectness. The basic equations read in the case of two coherent incident waves as /16/:

$$\psi_0 = |k_1\rangle + e^{i\chi} |k_2\rangle$$

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{k'}{2|k|} \left(\frac{m}{2\pi\hbar}\right)^2 S(\vec{k}_2 - \vec{k}_1, \vec{k}, \omega)$$

$$\begin{aligned}
S(\vec{k}_2 - \vec{k}_1, \vec{k}, \omega) = & \sum_{\lambda, \lambda'} P_{\lambda} |\langle \lambda | \sum_j U(\vec{R}) e^{i\vec{k}\vec{R}_j} + \\
& + e^{i\chi} \sum_j U(\vec{k}_2 - \vec{k}_1 + \kappa) e^{i(\vec{k}_2 - \vec{k}_1 + \vec{k})\vec{R}_j} | \lambda \rangle|^2 \cdot \\
& \cdot \delta(\hbar\omega + E_{\lambda} - E_{\lambda'})
\end{aligned}$$

$\vec{k} = \vec{k}_1 - \vec{k}'$  ..... momentum transfer for beam 1

$\chi = \chi_r + i\chi_i$  .... phase difference

where usual symbols of inelastic scattering theory have been used.  
This yields a phonon cross section:

$$\begin{aligned}
\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{\text{inel}} = & \frac{m^2 k'}{4\pi^2 \hbar^3 M k} \left(\frac{V}{(2\pi)^3 N}\right) \text{Re} \left[ a \tilde{U}^*(\vec{k}_1 - \vec{k}') \tilde{U}(\vec{k}_2 - \vec{k}') \exp[-W(\vec{k}_1 - \vec{k}') - W(\vec{k}_2 - \vec{k}')] \sum_{\vec{q}} \int d\vec{q} [\omega_{\vec{q}}(\vec{q})]^{-1} \right. \\
& \times \left( (\vec{k}_1 - \vec{k}') \cdot \vec{\sigma}(\vec{q}) (\vec{k}_2 - \vec{k}') \cdot \vec{\sigma}^*(\vec{q}) [n_{\vec{q}}(\vec{q}) + 1] \alpha(\omega - \omega_{\vec{q}}(\vec{q})) \right. \\
& \times \left| \sum_i \exp[i(\vec{q} - \vec{k}_1 + \vec{k}') \cdot \vec{r}_i] \right|^2 \\
& + (\vec{k}_1 - \vec{k}') \cdot \vec{\sigma}^*(\vec{q}) (\vec{k}_2 - \vec{k}') \cdot \vec{\sigma}(\vec{q}) n_{\vec{q}}(\vec{q}) \alpha(\omega + \omega_{\vec{q}}(\vec{q})) \\
& \left. \left. \times \left| \sum_i \exp[i(\vec{q} + \vec{k}_1 - \vec{k}') \cdot \vec{r}_i] \right|^2 \right) \right].
\end{aligned}$$

A set-up shown in Figure 2 can also be used advantageously for the observation of the neutron Kichuchi effect, which results from a mutual Bragg and phonon scattering inside the perfect crystal. The theoretical background for such experiments have been developed recently /17,18/. Such experiments represent an alternative method for phonon dispersion measurements and for the observation of dynamical diffraction effects in the inelastic regime /19/.

Additional applications of neutron interferometry at pulsed sources are in the field of fundamental physics and deal with delayed choice experiments /20/, the longitudinal and temporal coherence /21,22/ etc. Further topics in this direction are treated in the chapter of nuclear physics.

### 3. Summary

With the advent of pulsed neutron sources on the horizon new neutron interferometer measurements become feasible. The time structure will be used to resolve high order reflections and to measure the wave length dependent scattering amplitudes at epithermal energies. The gain factor in comparison to steady state sources lies in the range 10 and is determined by the time resolution needed to separate the different diffraction orders. Using proper nuclei a direct structure phase determination become feasible which will be of great use for the structure analysis of biological substances.

When high intense spallation sources become available inelastic scattering experiments using coherent incident beams become feasible permitting information on higher order correlation functions and dynamical diffraction phenomena in the coherent inelastic scattering part of the spectrum. For this type of experiments the pulse width has to be reduced to about 10  $\mu$ s and, therefore, the gain factor is only about 5 in comparison to an equivalent steady state source.

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## BUNCHING SPECTROMETER

D. Richter, B. Alefeld  
Kernforschungsanlage Jülich GmbH

D-5170 Jülich, P.O. Box 1913

## I. INTRODUCTION

A time-of-flight focusing spectrometer uses a larger wavelength band than a non-focusing spectrometer with the same resolution. Time focusing is important in rotating crystal spectrometers /1/ and was applied at the crystal-chopper instrument IN6 in Grenoble /2/. If a focusing efficiency is defined as

$$e_f = \frac{\Delta\lambda}{d\lambda}$$

$\Delta\lambda$  wavelength band of the focusing instrument

$d\lambda$  wavelength band of the non-focusing instrument

then  $e_f$  is of the order of two for a rotating crystal spectrometer and it is three at the IN6.

In 1967 Maier-Leibnitz /3/ proposed a high resolution time focusing instrument, also called bunching spectrometer with a much higher focusing efficiency of the order of hundred and more. In this bunching spectrometer a monochromator crystal in back-reflection is accelerated in direction of the incoming neutron beam. As a consequence of the Doppler effect the velocity of the reflected neutrons during an acceleration period increases as

$$v(t) = v_0 + \dot{w}(t) \quad (1)$$

where  $v_0$  is the reflected neutron velocity for the resting monochromator, and  $\dot{w}$  is the velocity of the crystal. If  $\dot{w}(t)$  and  $v_0$  are chosen properly, time focusing can be achieved at the detector which is placed at a distance  $L_2$  from the monochromator and in principle high resolution could be obtained. Such a spectrometer has been considered in detail by Peterlin /4/ and was



promoted later on for the use on pulsed sources /5/. In the following we briefly describe the bunching spectrometer and discuss its possible use on SNQ.

## II. BASIC PRINCIPLES

In order to derive the time focusing condition, we first consider the time dependence of the incoming neutron velocities at the monochromator which is positioned at a distance  $L_1$  from the source. A neutron with a velocity  $v_0$  arrives after a flight time

$$T_F = L_1 / v_0. \quad (2)$$

At later times  $t$  the incoming neutron velocities decrease by

$$v = v_0 - \frac{v_0^2}{L_1} t. \quad (3)$$

To achieve time focusing, these later neutrons have to be accelerated such that they hit the detector at the same time as the initially reflected neutrons. The focusing condition is then:

$$v_0 + \dot{w} = \frac{L_2 - w(t)}{L_2/v_0 - t} \quad (4)$$

where  $w(t)$  is the monochromator position at time  $t$ , and  $L_2$  is the distance from the monochromator to the detector at  $t = 0$ . Neutrons which are reflected at time  $t$  have to travel the distance  $L_2 - w(t)$ . In order to be at the detector at the same time as the first neutrons, they have to cover this distance during a time  $T_F - t = L_2/v_0 - t$ . Finally, we have to account for the fact that also the flight path of the incident neutrons is changed by the monochromator motion. This changes Equ. (3) to

$$v(t) = v_0 - \frac{v_0^2}{L_1} t + \frac{v_0}{L_1} w(t). \quad (5)$$

Equ. (4) combined with Equ. (5) is the differential equation for the monochromator motion  $\dot{w}$ . Focusing is only achieved if  $L_1 = L_2 = L$ . Then Equ. (4) is approximately solved by

$$\frac{d^2 w}{dt^2} = v_0^2 / L. \quad (6)$$

In order to use the time structure of the pulsed source efficiently, the acceleration (6) has to be maintained for a substantial part of the time between two pulses  $T$ . Figure 1 displays the time distance diagram for such a bunching process. At the beginning of a cycle the monochromator moves away from the source and thus reduces the neutron velocities of the incoming pulse. At later times the direction of the Doppler velocity reverses and the slower late neutrons are accelerated, in order to reach the detector at the same time as the first neutrons.

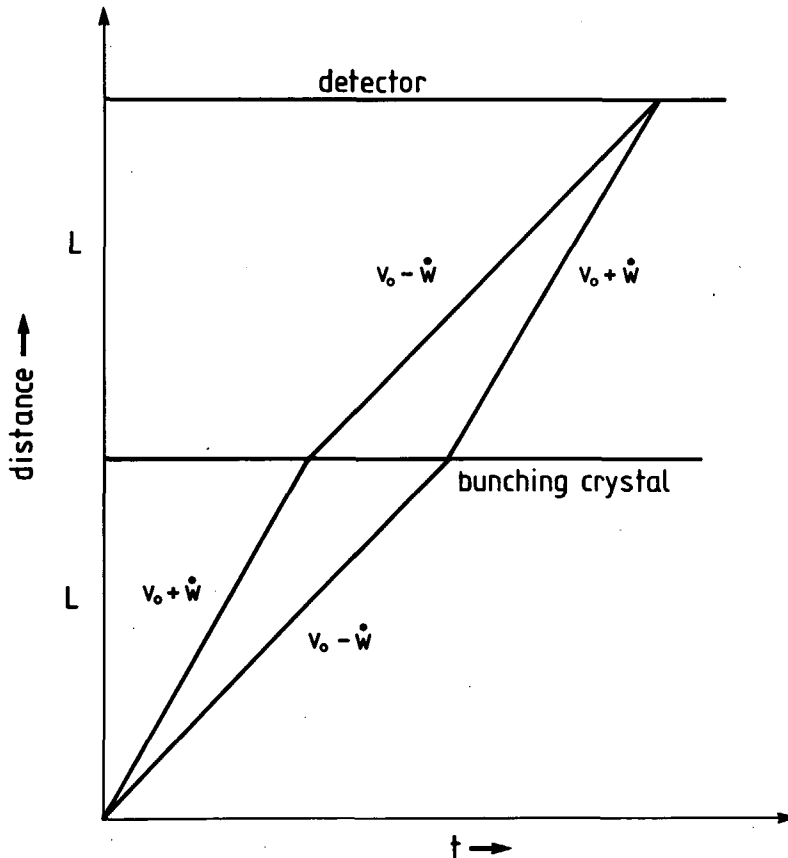


FIG. 1: Time-of-flight diagram for a bunching spectrometer

Now we consider the resolution of the bunching spectrometer. Ideally, for an infinitely sharp monochromator, bunching would be perfect however at the same time the intensity would be zero. For a real crystal providing finite resolution and intensity, the velocity reflected at a given instance  $t$  is affected by an uncertainty resulting from the monochromator resolution  $\delta E/E_0$ .

$$\frac{\delta v}{v} = \frac{1}{2} \frac{\delta E}{E_0} . \quad (7)$$

This velocity spread causes a pulse width  $\delta\tau$  at the detector

$$\delta\tau = \frac{L}{v} \left( \frac{\delta v}{v} \right) . \quad (8)$$

(8) is the leading contribution which determines to a large amount the time-of-flight resolution  $\alpha$  of the instrument, if the sample is placed directly in front of the Doppler drive

$$\alpha = \frac{\delta v}{v} = \frac{\Delta\tau'}{T_F} ; \quad \Delta\tau' = \sqrt{\delta\tau^2 + \Delta T_1^2 + \dots} \quad (9)$$

where the  $\Delta T_1$  are resolution contributions due to finite sample size, detector thickness etc.

For an optimized instrumental design time-of-flight resolution and crystal resolution should be matched. This condition excludes perfect monochromators with resolution widths in the order of  $10^{-4}$  - they never can be matched by a time-of-flight analyser. There remain mosaic crystals like graphite or future gradient crystals like the Si-Ge system /6/.

If the sample is not located close to the monochromator for a given  $\delta\tau$  (Equ. (8)), the time-of-flight resolution becomes poor. One obtains

$$\alpha = \frac{\delta\tau}{T_F} = \frac{L}{v} \left( \frac{\delta v}{v} \right) \frac{v}{L_A} \quad (10)$$

where  $L_A$  is the distance between sample and detector. For a feasible time-of-flight analyzer,  $L_A$  is limited. As an upper limit we consider  $L_A = 6$  m. Thus, the requirements of a short analyzer and a long focusing flight path (Equ. 6), in order to keep the necessary accelerations within technical limits, are contrary. The reconciliation of these opposing demands is the main task for a proper instrumental design.

The parameters which dominantly determine the intensity at the sample position are: the peakflux  $d\phi/dE$  of the source, the energy band selected by the monochromator  $\delta E = 2 (\delta v/v) E_0$ , and the duty cycle  $t_B/T$

$$I_B \propto \frac{d\phi}{dE} \delta E \frac{t_B}{T} = \frac{d\phi}{dE} \frac{L_A}{L} 2 E_0 \alpha \frac{t_B}{T} \quad (11)$$

For comparison we consider a non-focusing double chopper time-of-flight instrument with the same resolution  $\alpha$ . Figure 2 shows the distance time diagram of such an instrument. The time uncertainty  $\delta\tau$  of the neutrons at the detector is composed of two contributions, one from the chopper opening time, the other from the finite neutron velocity band. The matching of both contributions demands

$$\Delta t_{Ch} = \frac{\delta v}{v} \frac{L}{v} = \frac{1}{2} \delta\tau \quad (12)$$

$\Delta t_{Ch}$  is the opening time of the chopper. The resolution is dominated by the pulse width

$$\alpha = \frac{\delta\tau}{T_F} = \frac{\delta\tau}{L_A} v \quad (13)$$

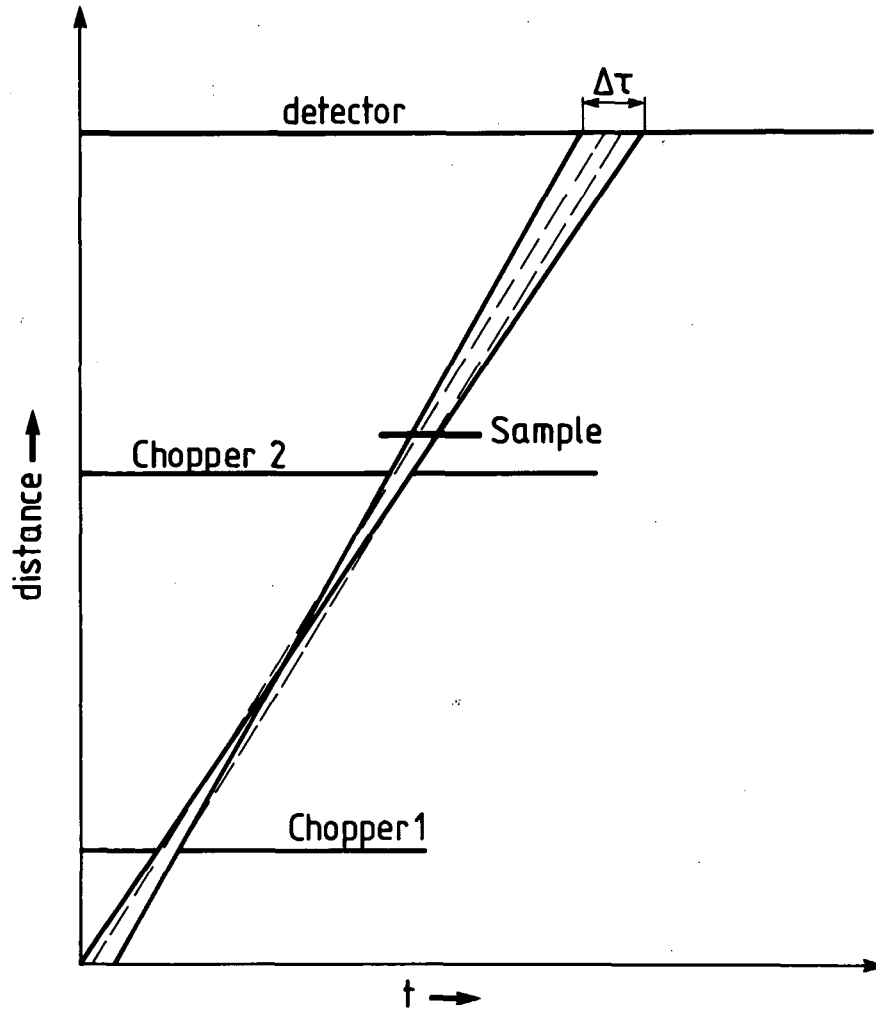


FIG. 2: Time-of-flight diagram for a conventional double chopper TOF spectrometer

The intensity at the sample position is proportional to the peak flux, the chopper opening time and the transmitted energy band

$$I_T \propto \frac{d\phi}{dE} 2 \left( \frac{\delta v}{v} \right) E_o \frac{\Delta t_{Ch}}{T} = \frac{d\phi}{dE} E_o \frac{t_{Ch}}{T} \alpha \quad (14)$$

The relative intensity gain of the bunching spectrometer compared to the non-focusing machine follows from the ratio of Equ's (11) and (14),

$$g = \frac{I_B}{I_T} = \frac{2 L_A t_B}{L \Delta t_{Ch}} = \frac{4 t_B v}{L \alpha} \quad (15)$$

Thus, long bunching times and high resolution lead to large gain factors.

### III. EXAMPLES

The most critical part of a bunching spectrometer is the Doppler drive. Its technical limits to a large extent will determine the design of the whole instrument. Therefore we start our considerations with this component. We assume a sinusoidal velocity profile and use only one tenth of the whole period of 10 ms - the frequency of the Doppler drive has to be synchronized with that of the source -. In this range the sinus is close to linear and the acceleration is constant as demanded by Equ. (6). Thus,  $t_B = 1$  ms. As a maximum velocity which still appears to be feasible technically, we take  $v_{\max} = 30$  m/s [7]. This value corresponds to piston velocities in high performance combustion engines. For the amplitude  $A$  of the Doppler motion we get from  $A\omega = v_{\max}$  ( $\omega = 628 \text{ s}^{-1}$ ),  $A = 4.8$  cm. Considering back-reflection on Si-Ge gradient crystals [6] with  $v = 630$  m/s, the focusing condition (Equ. (6)) determines the focusing length of the instrument

$$\frac{v^2}{L} = A \omega^2 \rightarrow L = 21 \text{ m.} \quad (16)$$

With a maximum secondary flight path of  $L_A = 6$  m, the mismatch of this machine amounts to  $L/L_A = 3.5$ . The maximum velocity range which is bunched during one period follows from

$$\dot{w}_m = \frac{1}{2} A \omega^2 t_B = 9.4 \frac{\text{m}}{\text{s}} \quad (17)$$

and amounts to  $620.6 \text{ m/s} \leq v \leq 639.4 \text{ m/s}$ . The covered energy range is  $\Delta E = 4 \dot{w}_m / v E = 123 \text{ } \mu\text{eV}$ . Finally the intensity gain factor  $g$  (Equ. 15) becomes

$$g = \frac{4 t_B v}{L_1 \alpha} = \frac{0.12}{\alpha} . \quad (18)$$

Thus, in the regime of  $\alpha \approx 10^{-2}$  which is typical for high performance double chopper spectrometers, the intensity gain would still be in the order of 10 opposed to 3 which is achieved by the IN6 in Grenoble. The monochromator resolution necessary in order to achieve  $\alpha = 10^{-2}$  is  $\frac{\delta E}{E} = \frac{2L}{L_A} \left( \frac{\delta v}{v} \right) = 5.7 \cdot 10^{-3}$ . This appears to be possible with Si-Ge gradient crystals /6/. Figure 3 presents a schematic scetch of the bunching spectrometer. The neutrons evolving from the source are traveling 21 m to the bunching crystal, are back-reflected and after further 11 m are extracted from the neutron guide by a graphite crystal which, e.g. by the aid of a temperature gradient, is able to reflect the entire pulse. After 4 more meters the neutrons hit the sample and are analyzed after the secondary flight path of 6 m.

Equ. (18) shows that the intensity of the bunching spectrometer surpasses that of the corresponding double chopper instrument the more the higher the instrumental resolution  $\alpha$  becomes. However, in the very high

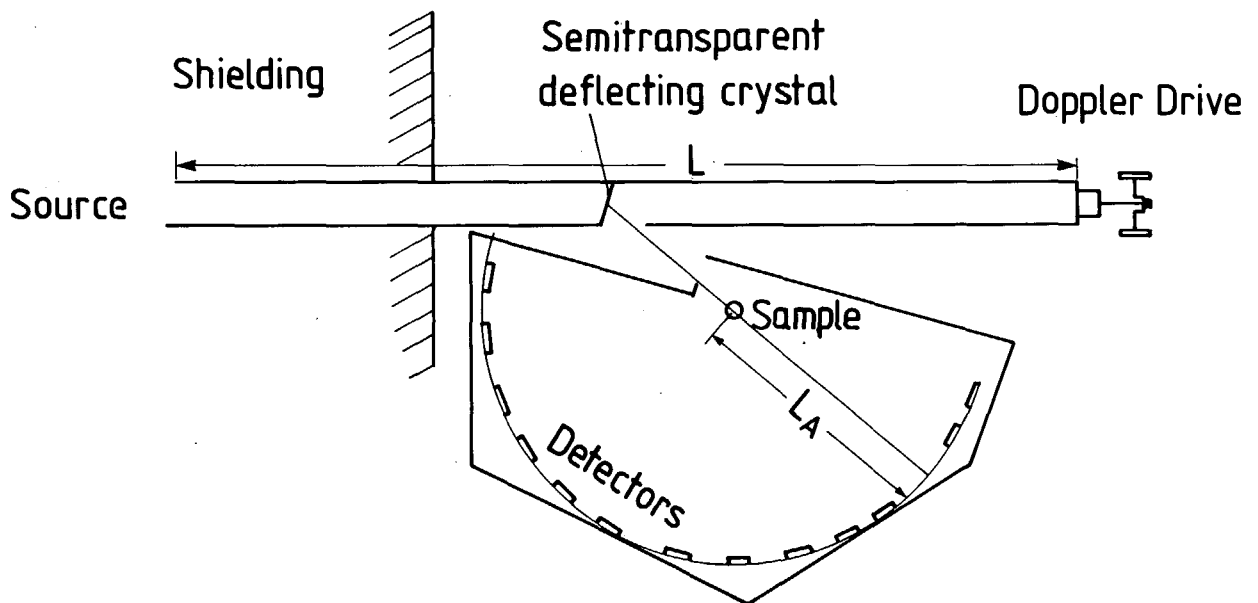


FIG. 3: Schematic drawing of a bunching spectrometer at SNQ

resolution range ( $\alpha \sim \dots 10^{-3}$  and better) double chopper time-of-flight machines are not the appropriate instruments to compare with. Here backscattering spectrometers have to be considered for comparison. As an example we take a time-of-flight backscattering spectrometer (IRIS) operating with graphite crystals. Such a spectrometer is described in the articles of Heidemann et al. /8/ and Scherm et al. /9/ in this report. Using an initial flight path of 130 m, IRIS can use the full pulse width of  $\tau = 440 \mu\text{s}$  provided by SNQ. Thereby it scans an energy window of  $\Delta E = 160 \mu\text{eV}$ . Its intensity at the sample position is given by

$$J_I \propto \frac{1}{2} \frac{d\phi}{dE} \Delta E \frac{\tau}{T}. \quad (19)$$

Comparing Equ. (19) and Equ. (11), the intensity ratio of the backscattering spectrometer and a bunching spectrometer of the same resolution is evaluated to

$$\frac{I_B}{I_T} = \frac{4 L_A E_o \alpha t_B}{L \Delta E \tau} = 0.10 \quad (20)$$

where  $\alpha$  is the velocity resolution of IRIS. Thus, IRIS will be more favourable than a bunching machine provided that the whole scanning range  $\Delta E$  leads to useful information. For the special case of narrow quasi-elastic lines the intensity at the detector of IRIS is reduced from Equ. (19) by a factor  $\delta E / \Delta E$  /8/, while there is no reduction factor for bunching. Then, in Equ. (20),  $\Delta E$  has to be replaced by  $\delta E$  - the resolution width of graphite - and we get 2.0 instead of 0.10. Thus for high resolution purposes, bunching and time-of-flight backscattering yield similar intensities. The actual superiority depends on the physical problem.

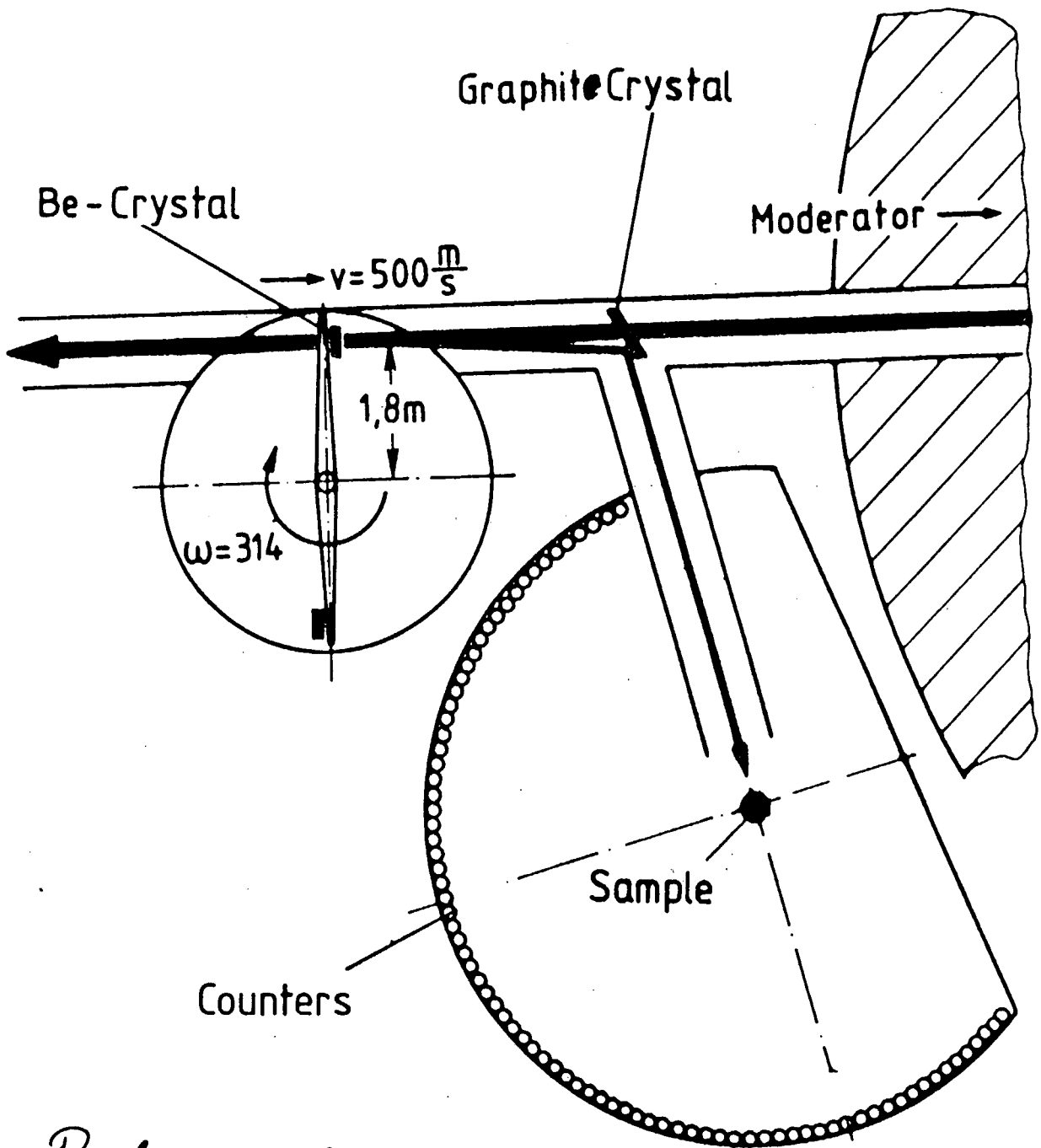


## IV. CONCLUSION

We have outlined the basic principles of a bunching spectrometer which could be used at SNQ. Considering a Doppler drive with a performance within the limits of standard technology, we found that in the regime of high resolution time-of-flight experiments ( $\alpha = 10^{-2}$ ) bunching should gain an order of magnitude in intensity compared to double chopper spectrometers. In the resolution regime of  $\alpha \approx 10^{-3}$  it is far superior to conventional time-of-flight techniques and is comparable with graphite backscattering spectrometers. With a more advanced Doppler drive such as proposed by Friedrich et al. /10/, a matched bunching spectrometer with bunching times  $t_B \approx 1/2 T$  could be possible. It would gain another order of magnitude compared to the instrument discussed here.

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Pulse width

$$\omega = \frac{d\varphi}{dt} \quad d\varphi \approx 10^{-2} \text{ rad}$$

$$dt = \frac{10^{-2}}{314} = 32 \mu\text{sec}$$

Polarization Analysis

V. Wagner, PTB Braunschweig

M. Löwenhaupt, KFA Jülich

Abstract

Polarization analysis (PA) allows the separation of magnetic, nuclear coherent and spin-incoherent scattering in an unambiguous manner. For inelastic neutron scattering studies combined with PA several instruments are considered: a direct geometry TOF spectrometer for cold neutrons, a crystal analyser spectrometer for thermal neutrons. Besides these instruments classical triple-axis-spectrometers with the option of PA will be very valuable.

## Introduction

This paper deals with the use of polarized neutrons and the application of polarization analysis in the field of neutron inelastic scattering as a means of identifying and separating different scattering processes, such as are coherent and spin-incoherent nuclear scattering and magnetic scattering. The paper does not cover the application of polarised neutrons as a tool for measuring inelasticity apart from the reference to the correlation methods which can favorably be adopted in time of flight spectrometers using polarised neutrons. A review on both aspects of polarization analysis based on the experience on reactors was given by Mezei/1/, while polarization spectrometers for a pulsed neutron source, have been reviewed in a recent report by Williams/2/.

In the following sections we recall the essential facts of polarized neutron scattering and outline the achievements and future possibilities of this technique in the field of condensed matter research. There will be a short look at the state of art of the techniques of polarizing, analysing and manipulating polarized neutron beams and then a section on instruments which should be considered for the German Spallation Neutron Source (SNQ).

## Polarized Neutron Scattering

As nuclear forces are spin dependent and the magnetic dipole interaction with matter involves the neutron spin  $\vec{s}_n = \hbar/2 \vec{\sigma}$  directly, neutron scattering does in general depend on the polarization of the beam  $\vec{P} = \langle \vec{\sigma} \rangle$ . In the case, where the sample has a preferred axis of quantization, the cross section itself depends on the polarization and the beam polarization will be changed. In the case of an isotropic sample without preferred direction the cross section does not depend on the polarization, but still the beam polarization may be changed by the scattering. Typically the situation is as shown in figure 1. In order to get all information from the scattering of polarized neutrons one has to determine the cross section  $\frac{d^2\sigma}{d\Omega dE}$ , and the polarization of the scattered beam  $\vec{P}'$ , which is a vector quantity.

The scattered beam polarization allows to identify scattering from different processes. For nuclear coherent scattering  $\vec{P}' = \vec{P}$ , while for nuclear spin-incoherent scattering  $\vec{P}' = -1/3 \vec{P}$ , if the nuclear spins are not aligned. For paramagnetic scattering  $\vec{P}' = \vec{Q}(\vec{P}\vec{Q})/Q^2$ , where  $\vec{Q}$  denotes the scattering vector  $\vec{Q} = \vec{k} - \vec{k}'$ . Scattering from a ferromagnet is more complex. It depends on the three vectors  $\vec{P}$ ,  $\vec{Q}$ , and  $\vec{m}$  (the direction of the magnetization) and may create scattered beam polarization. All cases are treated in detail by Marshall and Lovesey [3].

Many relevant experimental situations are met by the classical polarization analysis [4], which simplifies very much the experiment. The scattered beam polarization is analysed only along the direction of the incoming polarization, which is parallel to the preferred direction, i.e. the magnetization of a ferromagnetic sample or the magnetic guide field applied to an isotropic sample. Only the component  $P'_p = \vec{P}' \cdot \vec{P}/P$  is obtained, which is a scalar quantity. Behind the analyser one obtains directly the non-spin flip intensity.

$$I^{nf} = \frac{1+P'_p}{2} \frac{d^2\sigma}{d\Omega dE'} \quad (1a)$$

when the analyser transmits the spin state corresponding to the incident polarization, and the spin flip intensity

$$I^{sf} = \frac{1-P'_p}{2} \frac{d^2\sigma}{d\Omega dE'} \quad (1b)$$

when upon action of a spin flipper before the analyser the opposite spin state is transmitted.

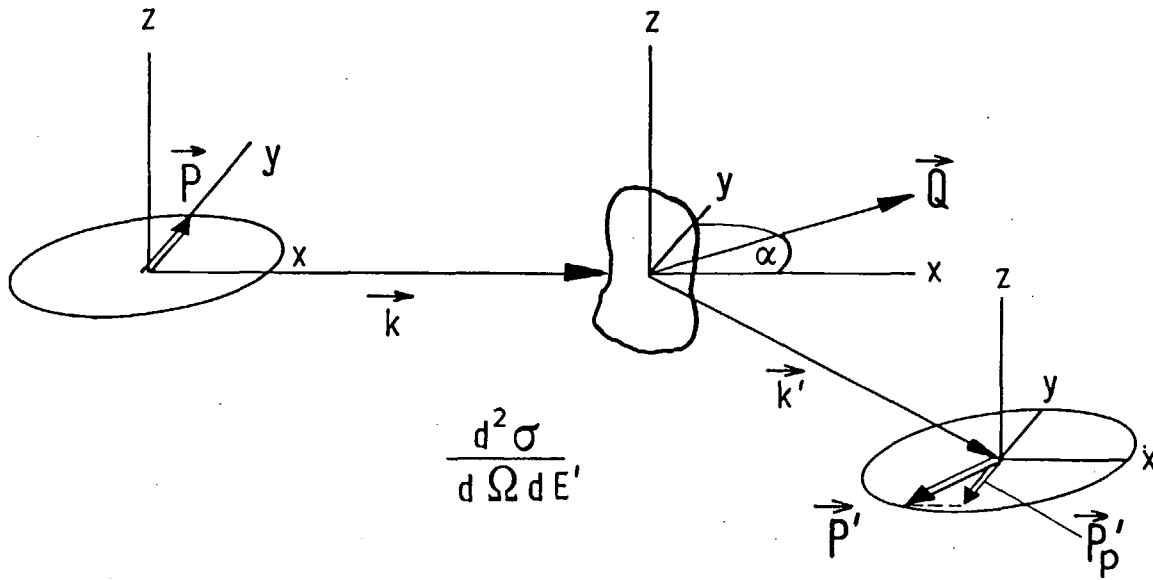


Fig. 1

The sample scatters a polarized beam with cross section  $d^2 \sigma / d\Omega dE'$ .

In general the scattered beam polarization  $\vec{P}'$  differs from the incident beam polarization  $\vec{P}$ .

Only the component  $P'_p$  parallel to the incoming polarization is measured in the case of the classical polarization analysis.

### Separating Different Types of Scattering by Polarization Analysis

In the following we restrict ourselves to the classical polarization analysis and we go to point out how much information may be obtained by this method.

In the case of an isotropic sample there is no preferred axis of quantization, i.e. no magnetization, no nuclear polarization. The cross section does not depend on the incoming beam polarization. It is the scattered beam polarization, which allows to distinguish between nuclear coherent, nuclear spin-incoherent and magnetic scattering. Full information on the different scattering intensities can be obtained by polarization analysis if measurements are taken at different configurations for the orientation of the incident beam polarization  $\vec{P}$  and the scattering vector  $\vec{Q} = \vec{k} - \vec{k}'$ . A small magnetic guide field applied to the sample ( $\mu_0 H = 0.1-0.5$  mT) is used to rotate  $\vec{P}$  at the sample position perpendicular or parallel to  $\vec{Q}$ . The applied field should be small enough to leave the sample

magnetically isotropic. In a TOF experiment  $\vec{P}$  cannot be kept parallel to  $\vec{Q}$ , as  $\vec{Q}$  varies for a given scattering angle with the time of flight. With the coordinate system chosen as in fig. 1 (x,y-axis within the scattering plane and z-axis perpendicular to it), one has for the non-spin flip and spin flip intensities  $I^{nf,sf}$  in the different configurations:

for  $\vec{P} = [0,0,1]$ :

$$I_z^{nf} = 1/3 I_{inc} + I_{coh} + 1/2 I_{mag} \quad (2a)$$

$$I_z^{sf} = 2/3 I_{inc} + 1/2 I_{mag}$$

for  $\vec{P} = [1,0,0]$ :

$$I_x^{nf} = 1/3 I_{inc} + I_{coh} + 1/2 \sin^2 \alpha I_{mag} \quad (2b)$$

$$I_x^{sf} = 2/3 I_{inc} + 1/2 (1 + \cos^2 \alpha) I_{mag}$$

for  $\vec{P} = [0,1,0]$ :

$$I_y^{nf} = 1/3 I_{inc} + I_{coh} + 1/2 \cos^2 \alpha I_{mag} \quad (2c)$$

$$I_y^{sf} = 2/3 I_{inc} + 1/2 (1 + \sin^2 \alpha) I_{mag}$$

$I_{inc}$  and  $I_{coh}$  denote the spin-incoherent and the coherent nuclear scattering intensity respectively. The isotope incoherent nuclear scattering is included into  $I_{coh}$ . The total magnetic scattering intensity is proportional to the fluctuations of the components of the local magnetic moments:

$$I_{mag} \sim 2/3 \langle \vec{M}(0) \vec{M}(t) \rangle.$$

Thus in order to separate magnetic scattering from nuclear scattering at least two measurements are necessary, if the angle  $\alpha$  between the incoming polarization  $\vec{P}$  and the scattering vector  $\vec{Q}$  is well defined in the experiment:

$$P_0 I_{mag} \cos^2 \alpha = 2 (I_x^{sf} - I_z^{sf}) \quad (3a)$$

The overall polarization efficiency  $P_o$  takes account of the non perfect performance of the polarizer and analyser of the spectrometer. A very important application of this method is the separation of paramagnetic scattering as in the case of the high temperature behaviour of itinerant ferromagnets /5/. It should prove very useful in the study of crystal field of spin-orbit levels in cubic symmetry.

In time-focusing spectrometers with relaxed Q-resolution, e.g. a neutron spin echo spectrometer,  $\cos\alpha = PQ$  is not well defined. By a measurement in the third configuration it can be eliminated to give /6/:

$$P_o I_{\text{mag}} = 2 (I_x^{\text{sf}} + I_y^{\text{sf}} - 2I_z^{\text{sf}}) \quad (3b)$$

Polarization analysis can be used to separate in an unequivocal way spin-incoherent and coherent scattering by the use of the relations (2a-2c). This allows to observe the self motion and the collective motion in systems, where the spin-incoherent cross-section is significant and isotope incoherence is negligible (e.g. H,D,<sup>23</sup>Na). Some first experiments carried at the ILL relate to the self diffusion in liquid sodium /7/ and to the orientational motion in the plastic phase of the molecular crystal C<sub>2</sub>Cl<sub>6</sub> /8/.

Among the aligned samples we consider only scattering from magnetic materials in an applied field. Ferromagnets have to be magnetized homogenously by an external field H, as otherwise the beam would depolarize along the passage through different domains. The incident beam polarization  $\vec{P}$  is kept parallel or antiparallel to  $\vec{H}$ , which is the direction of the magnetization  $\vec{m}$  and the preferred axis of quantization. Again the polarization of the scattered beam is analysed along  $\vec{P} // \vec{H}$ . All components perpendicular to  $\vec{H}$  will precess and average out. Because of the preferred axis the total cross-section depends on the incident beam polarization as does the spin-flip scattering as well. In a short motion, where we take the z-axis parallel to  $\vec{H}$ ,

$$I^{\text{nf}} \sim \text{FT} \langle M_{\perp}^z(-\vec{Q},0) M_{\perp}^z(\vec{Q},t) \rangle \quad (4a)$$

$$I^{\text{sf}} \sim \frac{1+P}{2} \text{FT} \langle M_{\perp}^{-}(-\vec{Q},0) M_{\perp}^{+}(\vec{Q},t) \rangle \quad (4b)$$

$$+ \frac{1-P}{2} \text{FT} \langle M_{\perp}^{+}(-\vec{Q},0) M_{\perp}^{-}(\vec{Q},t) \rangle$$



FT means the Fourier transformation and  $\vec{M}_\perp$  stands for the component of the local magnetization  $M$  (from electron spin and orbital momentum) perpendicular to the scattering vector  $Q$  with  $M^\pm = M_{\perp x} \pm i M_{\perp y}$  as usually. The usual orientational factor in magnetic scattering is now contained in  $\vec{M}_\perp$ . In an itinerant ferromagnet both longitudinal and transverse fluctuations are important and they can be individually measured by the non-spin flip and spin flip intensities.

In the most commonly used configuration  $\vec{P} \parallel \vec{H} \parallel \vec{Q}$  all magnetic scattering is spin flip scattering from transverse fluctuations because of  $M^z = 0$ . In this geometry there is no contribution from magneto-vibrational scattering. If the incident beam polarization  $P = 1$ , only the term corresponding to spin-wave creation will remain in (4b), whereas for  $P = -1$  only spin-wave annihilation will be possible. In order to separate magnetic scattering from nuclear scattering it is sufficient to measure the total scattering intensity for polarized neutrons without polarization analysis of the scattered beam for  $P = \pm 1$ . The nuclear scattering intensity does not depend on  $P$  and will cancel in the difference. Equivalent to this technique is the scattering of unpolarized neutrons with analysis of the scattered beam polarization only.

The method may be used to separate magnon scattering from phonon scattering (e.g.  $\text{Pd}_3\text{Fe}$  [9]) or to measure the magnetic content of hybridized spin-wave and phonon modes [10]. The same technique may be used to investigate the high temperature behaviour of ferromagnets. Thus polarized beam experiments on Ni [11,12] have shown the break-down of the above relation between polarization and magnon creation or annihilation. This is evidence that at high temperature the magnetization is not homogeneously aligned parallel to the saturating field, a phenomenon which should be checked in other ferromagnets, too.

The information provided by polarized neutron scattering is welcome to any experimentalist, who has been involved in the problem of separating say magnetic scattering from nuclear scattering in an unambiguous way. But how much is the price he has to pay in terms of counting time? A rough estimate for the increase in counting time is given by the factors

$$F = 2 \cdot n / T_p \quad T_A \simeq 6 \quad n \quad (5)$$

where  $n$  is the number of different partial cross-sections, we have to measure in order to single out the one we are interested in, and the effective transmission

of the polarizer or analyser  $T_p$ ,  $T_A$  is in practice between 0.3 and 0.7. Thus for separating magnetic from nuclear scattering ( $n=2$ ) the counting time has to be increased by an order of magnitude. Only intense neutron sources will render inelastic polarization analysis affordable. Even at the SNQ, polarized neutron scattering will continue to be time consuming and demands for dedicated instruments for a wide range of energy transfer.

### Polarizers and Spin Flippers

The development of efficient polarizers has progressed much for the last ten years /2/. Yet there is still a need for better white beam polarizers for thermal and epithermal neutrons. At present supermirror polarizers (Co-Ti) are available with an angular acceptance of  $\gamma^* = 32 \text{ mrad} \cdot \lambda/\text{nm}$  and a transmittance  $T \geq 0.50$  for wavelengths  $\lambda \geq 0.4 \text{ nm}$  /13/. At shorter wavelengths the transmittance drops sensibly and for  $\lambda \leq 0.3 \text{ nm}$  ( $E_n = 10 \text{ meV}$ ) polarizing crystals like the Heusler alloy  $\text{CuMnAl}_2$ ,  $\text{Fe}_3\text{Si}$  perform better. Peak reflectivities up to 0.50 have been achieved for crystals with a mosaic width at half height of 5 to 8 mrad. Heusler crystals are used as polarizing monochromators and are useful for energies  $E \leq 80 \text{ meV}$ . ( $\lambda \geq 0.1 \text{ nm}$ ). Then the resolution and the reflectivity deteriorate too much. For higher energies polarising resonance absorption filters can be used. Actually a  $^{149}\text{Sm}$ -filter which is again a white-beam polarizer, has been tested successfully at RAL /14/. It attains  $T=0.30$  with a polarization efficiency  $P \geq 0.95$  for energies  $E \leq 160 \text{ meV}$ . At still higher neutron energies white-beam polarizer of comparable performance do not exist. However, promising developments on spin polarized proton filter exist at several places.

There is a variety of spin-flippers /15/. Among these the non-adiabatic DC-coil flipper is most convenient and simple. Although a monochromatic device, it can be used as a wide band device, if the flipping magnetic field is tuned with the time-of-flight of neutrons of different energy.

In summary all components which render an inelastic spectrometer into a polarizing inelastic spectrometer are available in the range of cold and thermal neutrons, which will be the regime, where SNQ in its first stage will be most competitive. Therefore, it is easy to implement polarization analysis to any conventional spectrometer, provided that space is left for the physical size of a polarizer

analyser, flipper and guide fields.

### Inelastic Polarizing Spectrometers

As the field of application of polarization analysis is as large as the field of inelastic neutron scattering from condensed matter, quite different instruments are necessary to meet the requirements of resolution and energy transfer ranging from less than 1 meV say for linewidth of diffusive modes to more than 100 meV for magnetic single particle excitations and crystal field levels. Among the many possibilities we think that the following instruments will best fit to the first stage of SNQ with its rather long neutron pulse ( $\tau_m = 300 \mu s$  for thermal moderator and  $\tau_m \simeq 500 \mu s$  for cold moderator).

#### 1. Triple-axis-spectrometer (TAS) with polarization analysis (PA)

A comparison with TOF instruments shows that for the study of coherent excitations in single crystals TAS are competitive with respect to neutron flux density and resolution/16/. They offer the advantage of a simple control of the scattering vector, and the choice of the best configuration of  $\vec{P}$  and  $\vec{Q}$  for PA. The performance on SNQ will be the same as that of a reactor based instrument, with the advantage of easy discrimination of higher order contamination because of the time structure of SNQ. A dedicated TAS on the cold moderator should permanently have a polarized incident beam and the possibility of PA by a supermirror analyser before the ordinary graphite analyser.

All kind of low energy magnetic excitations and hybridized modes with lattice vibrations could be studied. For the investigation of excitations of higher energy ( $\hbar\omega \geq 5$  meV) the option of PA should exist on a thermal neutron TAS using Heusler crystals as polariser or analyser.

#### 2. Direct geometry time-of-flight spectrometer (X-TOF) with polarization analysis

This instrument is intended for the study of low energy magnetic excitations, mainly in single moment systems (mixed valence systems, crystal field levels etc.) and the investigation of selfmotion in liquids and plastic crystals.

Incident energy is determined by a crystal monochromator (PG) and final energy by time of flight. Beam polarization and analysis is done by supermirror polarizers (SM). Essentially, the instrument is the polarized beam version

of D7 at ILL /8/ transferred to SNQ at the cold source ( $\tau_m \simeq 500 \mu s$ ). Maximum final energy is limited by the decreasing transmission of SM at higher energies ( $E' \simeq 15 \text{ meV}$ ) which might be improved by the development of polarizing microguides. Typical energy resolution is about 4-5 percent.

The instrument can be operated in two modes (cf. fig. 2):

- a) as simple TOF spectrometer with a spin flip chopper,
  - b) with a pseudo-statistical spin flipper for cross-correlation technique.
- In case a) the gain in intensity as compared to the reactor based instrument

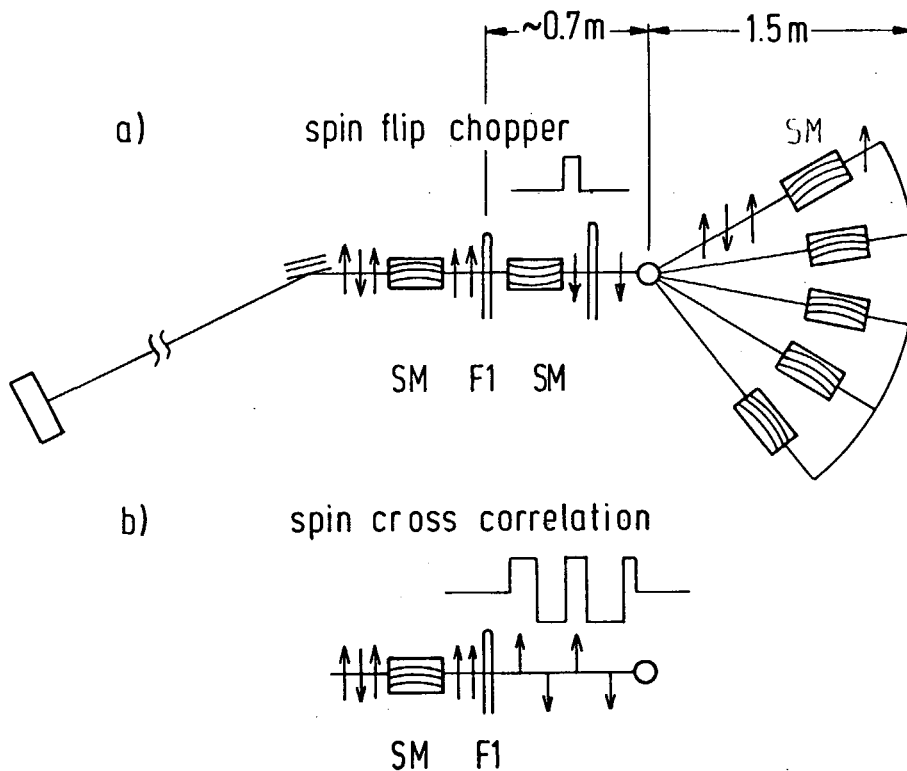


Fig.2 Inelastic TOF spectrometer with polarization analysis by supermirror analysers (cf. ref /8/).

- a) Flipper F1 between two opposite SM-polarizers produces short polarized neutron pulses.
- b) F1 produces a pseudo random sequence of up and down polarized neutron pulses.

is given by the ratio of the peak flux densities times the ratio of the repetition rates ( $f_{\text{reactor}} \geq f_{\text{SNQ}} = 100 \text{ Hz}$ ).

$$\phi_{\text{SNQ}} / \phi_{\text{reactor}} \cdot f_{\text{SNQ}} / f_{\text{chopper}} \simeq 10.$$

In case b) the polarized beam is not modulated in intensity but the polarization is reversed in a pseudo-random manner by the first flipper (F1). The cross correlation of the detected intensity with the pseudo-random sequence of the flipper produces a spectrum of the final flight times, which represents the difference of the non spin-flip and the spin-flip intensity

$$D = I^{\text{nf}} - I^{\text{sf}}.$$

From the difference spectrum obtained in different configurations the individual cross-section for magnetic or nuclear scattering can be obtained as usually.

There is no gain in intensity as compared to the reactor based instrument as long as the time averaged flux densities are the same. However, there is an important gain in quality of the data. Because of the intrinsic TOF resolution given by the moderator pulse length and the final flight time ( $\tau_m/t_2 \leq 0.4$ ), the interesting part of the TOF spectrum can be separated from non interesting parts and the background of ignorance may be improved considerably.

### 3. Crystal Analyser Spectrometer (TOF-X) with Polarization Analysis

This instrument is intended for the study of magnetic excitations in single crystals with energy transfer  $\hbar\omega \leq 100 \text{ meV}$  at scattering vectors  $Q \leq 40 \text{ nm}^{-1}$ . It is an inverted TOF-spectrometer (see fig. 3) characterized by a medium energy resolution. Scattered neutron energy  $E'$  is determined by a polarizing Heusler crystal, the incident neutron energy  $E$  is determined by the total time of flight. To do that to within 5 percent the instrument requires a Fermi type chopper close to the moderator with a burst time of  $\tau_{\text{CH}} = 60 \mu\text{s}$  for lower energies ( $20 \text{ meV} \leq E \leq 50 \text{ meV}$ ) and  $\tau_{\text{CH}} = 30 \mu\text{s}$  for higher energies ( $50 \text{ meV} \leq E \leq 160 \text{ meV}$ ).

The incident beam has to be polarized by a white-beam polarizer, in this case a  $^{149}\text{Sm}$  filter, which polarizes neutrons of energy up to 160 meV, is proposed. The scattered beam is analysed by a polarizing crystal. With  $\text{CuMnAl}_2$  as analyser,  $E' \leq 80 \text{ meV}$  appears to be a reasonable limit. For many

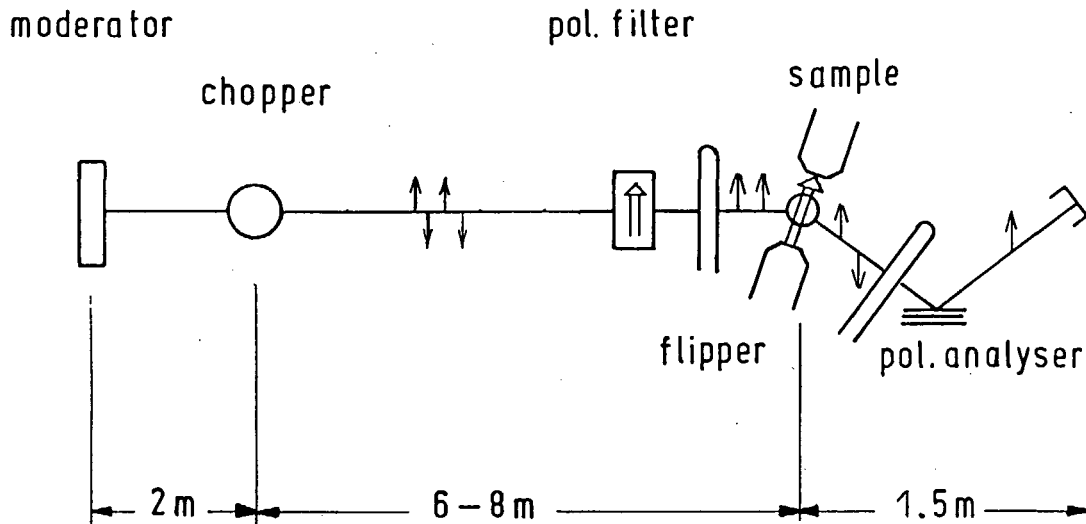


Fig. 3 Crystal analyser spectrometer.

The incident beam is polarized by a polarizing filter ( $^{149}\text{Sm}$  for  $E \leq 160$  meV). The scattered beam is analysed by a Heusler crystal.

applications (aligned sample) polarization of the incident beam only is required, thus leaving the choice of the analyser crystal free for optimization.

In the table 1 the flux density at the sample is compared to that of a TAS with same resolution. As mentioned above, the performance of both instruments is similar in the thermal energy range. At higher energies ( $E \geq 80$  meV) the TAS falls off because of the degrading polarizer reflectivity and narrowing collimation.

The efficiency of the proposed spectrometer can be improved if instead of a single detector several detectors are used. This can be done either in the configuration of a many analyser crystal (MAX) spectrometer /17/ or in the configuration of a constant-Q-spectrometer with one analyser and a multidetector /18/. In view of the selection rule , that all magnetic scattering is spin-flip scattering in the configuration  $\vec{P} // \vec{Q} // \vec{m}$ , the const-Q version appears more interesting. In its usual configuration the

crystal analyser planes are parallel to the incident beam direction  $\vec{k}$ . By this the k-space element  $\vec{\delta k'}$  accepted by the analyser is parallel to  $\vec{k}$  and  $\vec{Q} = \vec{k} - \vec{k'}$  can be kept constant. At the same time the mean scattering angle  $\phi$  equals the mean analyser angle  $\theta_A$ . One can get rid of this undesired restriction by the use of an analyser crystal whose planes are not parallel to  $\vec{k}$  and which is curved within the scattering plane. The curvature allows to rotate  $\vec{\delta k'}$  at will so that again  $\vec{\delta k'} // \vec{k}$ .

Table 1

Comparison of the flux density at the sample for the crystal analyser spectrometer (TOF - X) with a single detector and a triple-axis spectrometer (TAS) at  $E = 40$  meV,  $E = 160$  meV (values in brackets) and  $\delta E/E = 0.06$ . Distance moderator - sample  $d_{ms} = 8$  m and 7.5 m for TOF - X and TAS. Moderator width 0.1 m.

Time averaged flux density  $\sim \phi \cdot \delta t_{CH} \cdot f \cdot \Delta\Omega \cdot T_c \cdot T_p$

|                                                                  | TOF-X<br>$\delta t_{CH} = 60 \mu s$<br>(30 $\mu s$ )<br>$a_1 = 6$ m | TAS<br>Monochromator<br>$Cu_2AlMn(111)$          | relative<br>efficiency<br>TOF-X/TAS |                  |
|------------------------------------------------------------------|---------------------------------------------------------------------|--------------------------------------------------|-------------------------------------|------------------|
| transmission of<br>chopper or collimator $T_c$                   | 0.8                                                                 | 0.9                                              | 0.9                                 | } 0.37<br>(0.75) |
| accepted solid angle<br>$\Delta\Omega = \alpha_H \cdot \alpha_V$ | 43' x 43'                                                           | 21' x 46'<br>(10')                               | 1.9<br>(4.0)                        |                  |
| transmission of filter $T_p$<br>or reflectivity of Heusler $R_p$ | 0.3                                                                 | 0.4<br>(0.2)                                     | 0.75<br>(1.5)                       |                  |
| duty cycle $\delta t_{CH} \cdot f$                               | $6 \cdot 10^{-3}$<br>( $3 \cdot 10^{-3}$ )                          | 1                                                | 0.29<br>(0.14)                      |                  |
| flux density $\phi$                                              | $4.8 \cdot 10^{16} \text{ cm}^{-2} \text{ s}^{-1}$                  | $1 \cdot 10^{15} \text{ cm}^{-2} \text{ s}^{-1}$ |                                     | } 1.8<br>(3.8)   |
| number of useful<br>spectral elements                            | 5                                                                   | 1                                                | 5                                   |                  |
| gain by vertical focusing                                        | 1                                                                   | 2                                                | 0.5                                 |                  |
| gain due to compressor ring                                      | 2.5                                                                 | 1                                                | 2.5                                 |                  |

The crystal analyser spectrometer has a large potential for improvements, which would come with the installation of the compressor ring and a fast moderator at SNQ. It will gain in efficiency in the thermal neutron range due to the increased peak flux density by a factor of 2.5 and even more at higher energies. However, the extension of the incoming neutron energy beyond 160 meV would require a polarizing filter for these energies. A possible candidate is a  $^{151}\text{Eu}$ -filter, which still has to be developed [2].

#### 4. Resonance absorber spectrometer (TOF - TOF)

This is a vague suggestion for a polarized beam inelastic spectrometer at high energies. It requires a short pulse of epithermal neutrons ( $\delta t = 2 \mu\text{s}$ ), which will be available with reasonable intensity only after the installation of the compressor ring at SNQ. Incident beam polarization only is intended for the study of magnetic excitations in an aligned sample ( $\hbar\omega \leq 0.3 \text{ eV}$ ,  $Q \leq 50 \text{ nm}^{-1}$ ). A fast Fermi chopper and background choppers are necessary for primary energy definition. Polarization could be done by a resonance filter ( $^{163}\text{Dy}$ ).

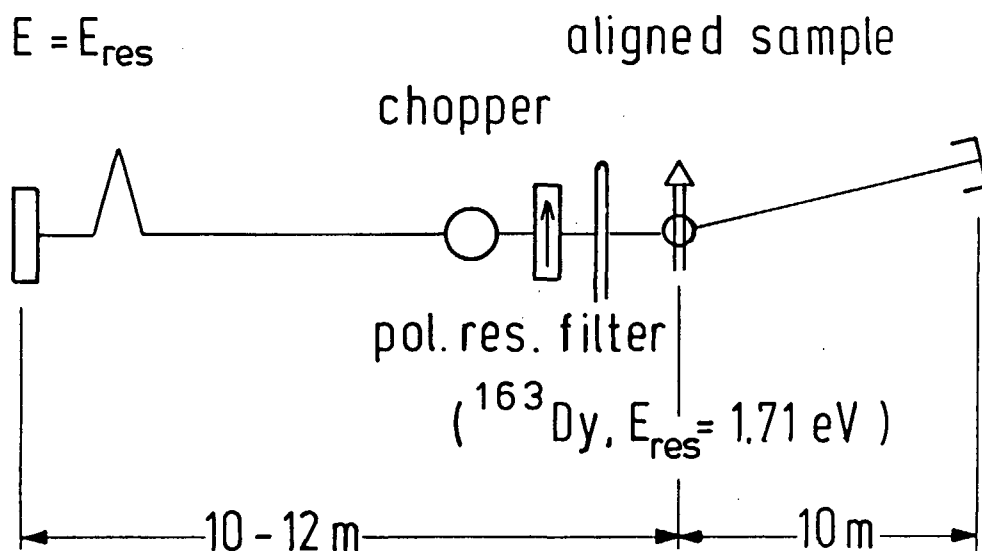


Fig.4 Resonance absorber spectrometer on a fast moderator.  
The moderator pulse and the chopper define  $E = E_{\text{res}}$ .  
Only the incident beam is polarized by the resonance filter.



### Conclusion

Polarization analysis is a unique tool for separating magnetic, nuclear coherent and nuclear spin-incoherent scattering. The success of the method depends much on the performance of the polarizer and analyser. At present the loss of intensity in a complete polarization analysis experiment is roughly one order of magnitude or worse. From this fact comes the need for dedicated inelastic polarization analysis spectrometers and also for a continuous improvement of the polarizers themselves.

For the cold neutron range a direct geometry TOF instrument is proposed, which offers the largest intensity gain as compared to a reactor-based equivalent. For thermal neutron energies the crystal analysers spectrometer will have a performance comparable to that of a TAS instrument as long as the thermal neutron pulse width is 300  $\mu$ s. It will be clearly superior, when the compressor ring will be installed at SNQ.

The authors are indebted to Dr. W.G. Williams for stimulating discussion and many suggestions.

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# A POSSIBLE NEW METHOD FOR OPTIMAL USE OF THE PEAK FLUX OF PULSED NEUTRON SOURCES

G.Badurek\* and W.Schmatz

Institut für Angewandte Kernphysik, KFK Karlsruhe, Fed.Rep.Germany

\*Institut für Kernphysik, Technische Universität Wien, Austria

## 1. Introduction

Neutron scattering experiments on pulsed neutron sources, as e.g. the planned spallation source SNQ of KFA Jülich, in general, will require spectrometers which differ from those installed at continuous neutron sources. There is a big challenge to the experimentalists and technicians both to appropriately modify existing spectrometer types and to conceive novel instruments to make optimal use of the time structure of pulsed neutron sources and consequently of their very high available peak flux. In the thermal wavelength range, for instance, this peak flux will exceed the time averaged flux by a factor of about 40. It is evident that time-of-flight techniques at pulsed sources play an essential role in defining either the incident neutron energy or the energy of the scattered neutrons or both. The usual supplementary method is given by crystal diffraction and we may refer to the several possible combinations of crystal (X) and time-of-flight (TOF) arrangements before and after the scattering target as TOF-X, X-TOF and TOF-TOF geometries. All of these combinations have their particular merits (and flaws) which we do not aim to discuss here; we just mention, however, that for the special situation given at a spallation source an "inverted" geometry TOF-X spectrometer seems to be particularly adequate. There the pulsed white beam is scattered by the sample sufficiently far away from the source to allow for a time-of-flight separation of the incident energies. The energy analysis of the scattered neutrons is done by means of single crystal analyzers

adjusted to fixed energies of as many scattering angles simultaneously as possible. It is evident, however, that from the white spectrum of incident energies only those energies can be used to which the analyzer crystals were adjusted.

In what follows we aim to show that it is in fact possible to conceive a novel neutron spectrometer of that inverse geometry type which makes use of actually all primary energies. But it is beyond the scope of this preliminary report to give a detailed construction recipe of such an instrument including a thorough quantitative examination of all its performance features. We rather intend here to merely explain its functional principles and to stimulate broader discussions about eventual further improvements.

## 2. Essential Components

More than two decades ago Drabkin /1/ has first proposed to use spatially alternating magnetic fields oriented perpendicular to a homogeneous magnetic field as a wavelength-dependent spinflip device for polarized neutron beams. On passage through such a magnetic wiggler-type flipper which is shown schematically in Figure 1 in its own frame of reference each neutron "sees" a time-dependent rf field with a basic frequency component (higher harmonic contributions will be neglected for simplicity)

$$B_1'(t) = B_1' \sin\left(\frac{\pi v}{a} t\right) = \frac{4}{\pi} B_1 \sin\left(\frac{\pi v}{a} t\right) \quad (1)$$

where  $B_1$  is the amplitude of the alternating field,  $2a$  its spatial period and  $v$  the neutron velocity. In direct analogy with conventional rf spinflippers the spinflip probability of Drabkin's device is then given as /2/

$$W_{\text{flip}} = \frac{\left(\frac{B_1'}{2B_0}\right)^2}{\left(\frac{v_0 - v}{v}\right)^2 + \left(\frac{v_0}{v} \frac{B_1'}{2B_0}\right)^2} \cdot \sin^2\left(\frac{\pi \ell}{2a} \left[\left(\frac{v_0 - v}{v}\right)^2 + \left(\frac{v_0}{v} \frac{B_1'}{2B_0}\right)^2\right]^{1/2}\right) \quad (2)$$

where  $\ell$  is the overall length of the flipper and  $v_0 = |\gamma|B_0 a/\pi$  is that neutron velocity which produces a frequency equal to the Larmor frequency  $\omega_0 = |\gamma|B_0$ , with  $\gamma = -1.83 \times 10^8$  rad/(s.T) the neutron gyromagnetic ratio. This spinflip probability is very sensitive to deviations  $\delta v = v - v_0$  of the neutron velocity from the resonance value  $v_0$ . If the amplitude condition for optimal flipping

$$\frac{\ell B_1}{2aB_0} = \frac{|\gamma| \ell B_1}{2\pi v_0} = 1 \quad (3)$$

is fulfilled for the neutron velocity  $v_0$  one finds for the full half-width of the velocity dependent spinflip probability resonance curve

$$\left(\frac{\delta v}{v_0}\right)_{1/2} \sim 1.6 \frac{a}{\ell} \quad (4)$$

Drabkin and co-workers /2/ were also the first to recognize that such a device in combination with magnetic mirror reflection polarizers cannot only be used to realize a novel neutron velocity selector without mechanically moving parts but can actually replace single crystal energy analyzers in conventional neutron spectrometers. However, this method remained as long "exotic" and of not too much practical use as no powerful magnetic mirrors had been invented. Nowadays the situation is totally different due to the availability of high quality supermirror /3,4/ and multilayer /5/ neutron polarizers which can handle appreciably large angular beam divergencies even for thermal neutrons.

What makes such a magnetic neutron energy analyzer system so exciting? In particular for pulsed source applications it is the possibility to adjust the desired energy simply by varying the size of the static magnetic field  $B_0$ . If we assume, for instance, a period  $2a = 1$  mm then even for  $1 \text{ \AA}$  neutrons this field would be as moderate as 680 G and its spatial extension for a wavelength resolution of 1% not larger than about 10 cm\*. Therefore, it is absolutely feasible without any serious experimental difficulty to be overcome to vary this field quickly enough in time at a pulsed neutron source to collect all neutrons that have been scattered into a

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\*Footnote: According to eqs. (1), (2) and (3) the ratio  $B_1/B_0$  has to be constant for optimal flipping and therefore  $B_1$  has to be changed proportional to  $B_0$ , which is even much less a problem.

certain direction with the same energy transfer irrespective of their primary energy. In the next chapter we try to sketch a neutron spectrometer that is based on this concept.

### 3. Proposed Spectrometer Design

Consider the arrangement sketched in Figure 2 where for simplicity only one scattering angle is indicated. For the beginning let us assume we want to single out the purely elastic scattering contribution, as e.g. is the case for diffuse scattering investigations. Let us further assume that the source pulse is tailored short enough by means of a phased chopper near the source to yield sufficient TOF resolution at moderate flight paths. From the space-time diagram of Figure 3 it is seen that this polychromatic pulse spreads out in space during its propagation to the sample. Therefore, every incident wavelength hits the sample at different times. If a crystal analyzer would be used behind the sample to filter out the elastic contribution, the time-of-flight  $t_1 = \hbar k_0 L_1 / m$  along the distance  $L_1$  would be fixed and hence only a very small portion of the incident neutron spectrum actually be useful. If instead the magnetic analyzer system shown in Figure 2 is used, the situation becomes completely different. This analyzer consists of the velocity-dependent flipper described in the preceding section, placed between two supermirror polarizers, plus an additional spinflipper whose action is independent of the neutron wavelength. Since there are a lot of rather simple options to realize such a type of flipper, it shall not be further treated in our present context. It is just necessary to achieve that without the wiggler flipper in action (nearly) no neutron can reach the detector because its polarization is then aligned antiparallel to the orientation of the second supermirror.

Now suppose that the magnetic analyzer is adjusted to transmit neutrons of a given wavelength  $\lambda$ . It is straightforward to derive /6/ that for such an inverted geometry spectrometer then a total flight time  $t$  corresponds to an energy transfer

$$\Delta E = \frac{m}{2} \left[ \frac{L_0^2}{\left(t - \frac{L_1}{v}\right)^2} - v^2 \right] \quad (5)$$

As indicated in Figure 2,  $L_0$  and  $L_1$  denote the respective flight paths from source to sample and sample to detector. From this equation and from the resonance condition  $v = |\gamma| B_0 a / \pi$  it follows then that all elastically scattered neutrons (i.e.  $\Delta E = 0$ ) irrespective of their initial velocity have a chance to reach the detector if the value of the homogeneous field  $B_0$  of the Wiggler flipper decreases linearly in time according to

$$B_0(t) = \frac{\pi v_{\max}}{2|\gamma|a} \left[ \frac{v_{\max}}{L} t + 3 \right] \quad \text{for} \quad \frac{L}{v_{\max}} \leq t \leq \frac{L}{v_{\min}} \quad (6)$$

Here  $L = L_0 + L_1$  is the total flight path and  $v_{\max}$  and  $v_{\min}$  are the highest and lowest velocities present in the incident neutron spectrum.

From the reciprocal space diagram in Figure 4 one sees qualitatively the improvement compared with a crystal analyzer. Since with the latter the scattered wave vector  $\vec{k}_1$  is fixed and independent of time only a small volume in  $\vec{k}$ -space whose actual size is determined by the chosen momentum resolution can contribute to the detected scattering events. (In Figure 4 this resolution has been assumed to be infinitely high, hence the volume is degenerated into a single line.) With the novel analyzer system, however, all scattered wave vectors within the hatched area yield an intensity contribution. The intensity gain of the time varying analyzer as compared to a conventional crystal analyzer is thus obviously the more pronounced the better the resolution is. But aside of reflectivity and angular acceptance considerations which have to be taken into account for a thorough quantitative comparison of both methods one has to bear in mind that neutrons of wrong wavelength have a finite chance to reach the detector because of imperfections of the supermirror polarizers and the spinflippers. A reasonably realistic assumption for the effective beam polarization product is  $e_1 e_2 P^2 \sim 0.97$  ( $e_1, e_2, \dots$  efficiencies of the two flippers,  $P$  ... polarization efficiency of the supermirror). If the incoming neutrons are sorted into different time channels

according to their total time-of-flight each channel belongs to a specific incident wavelength which has been scattered elastically. The average probability for an inelastically scattered neutron belonging to any incident wavelength to be counted in that particular time channel is then about 3% for the assumed performance of the analyzer. For elastic scattering investigations such a background, in general, can be tolerated. But with the present state of the art of supermirror polarizer technology it turns out that for inelastic scattering investigations the application of the proposed new method will not be recommendable because of the much more severe background problematic. On the other hand, if the relative wavelength resolution of the analyzer can be as relaxed as e.g. 5 - 10%, which frequently occurs in elastic diffuse scattering experiments, one needs not even to sort into different time channels. There the integral intensity is a sufficient measure of the elastic scattering contribution and the imperfections of the analyzer transmission characteristics with regard to the inelastic background can be neglected.

#### 4. Concluding Comments

Aside of detailed luminosity and resolution considerations in this preliminary report we also did not treat eventual applications of electronically tunable monochromators to others than inverted geometry neutron spectrometers. In particular, we also did not consider which influences might arise on the design of such spectrometers due to the fact that the wavelength resolution of such monochromators can be altered almost instantaneously without any variation of the scattering geometry. All one needs to do for that purpose is to shortcircuit part of the magnetic wiggler oscillations while simultaneously maintaining the amplitude condition (3) by proper increase of  $B_1$ .



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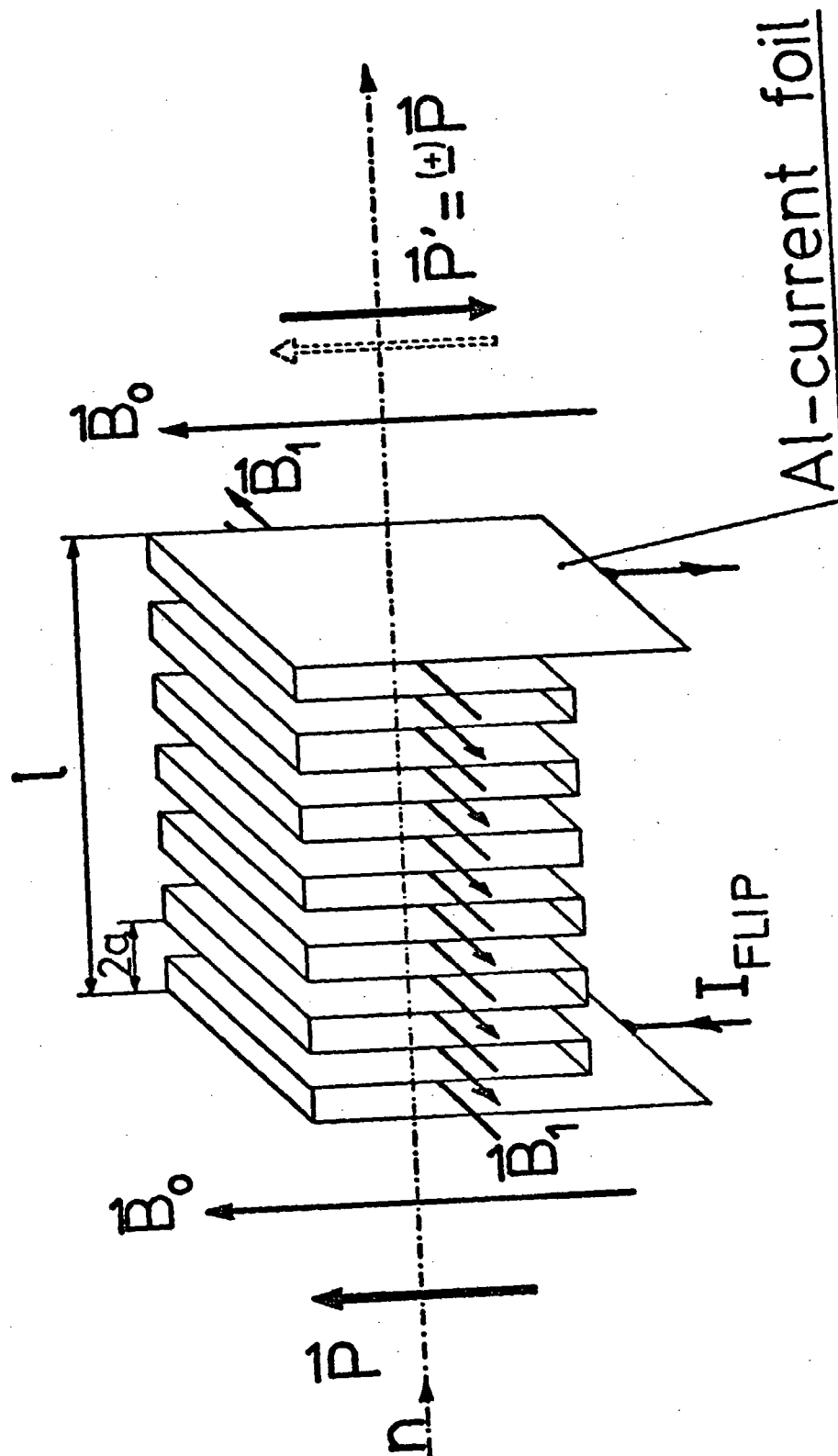


Figure 1: The magnetic wiggler neutron spinflipper.

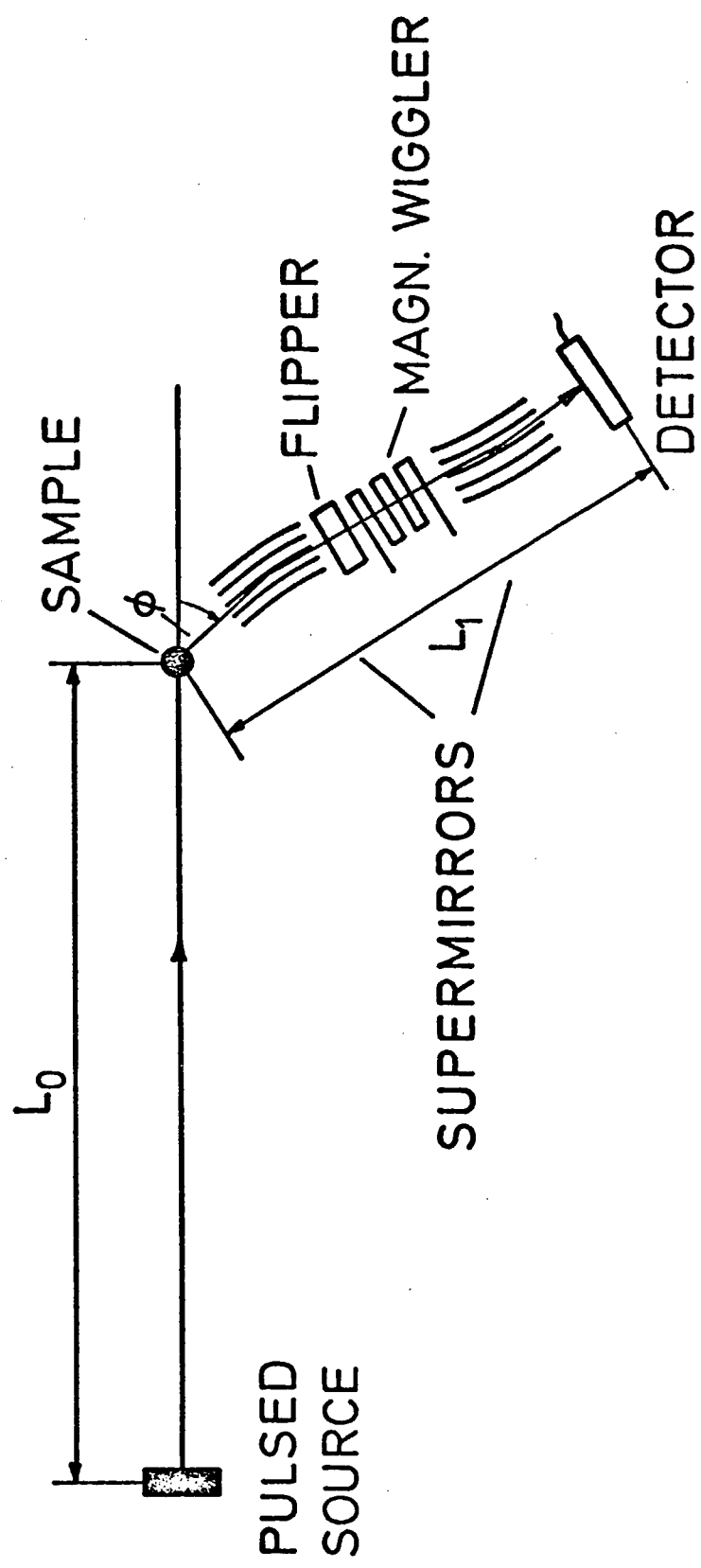


Figure 2: Schematic arrangement of the proposed new time-of-flight spectrometer.

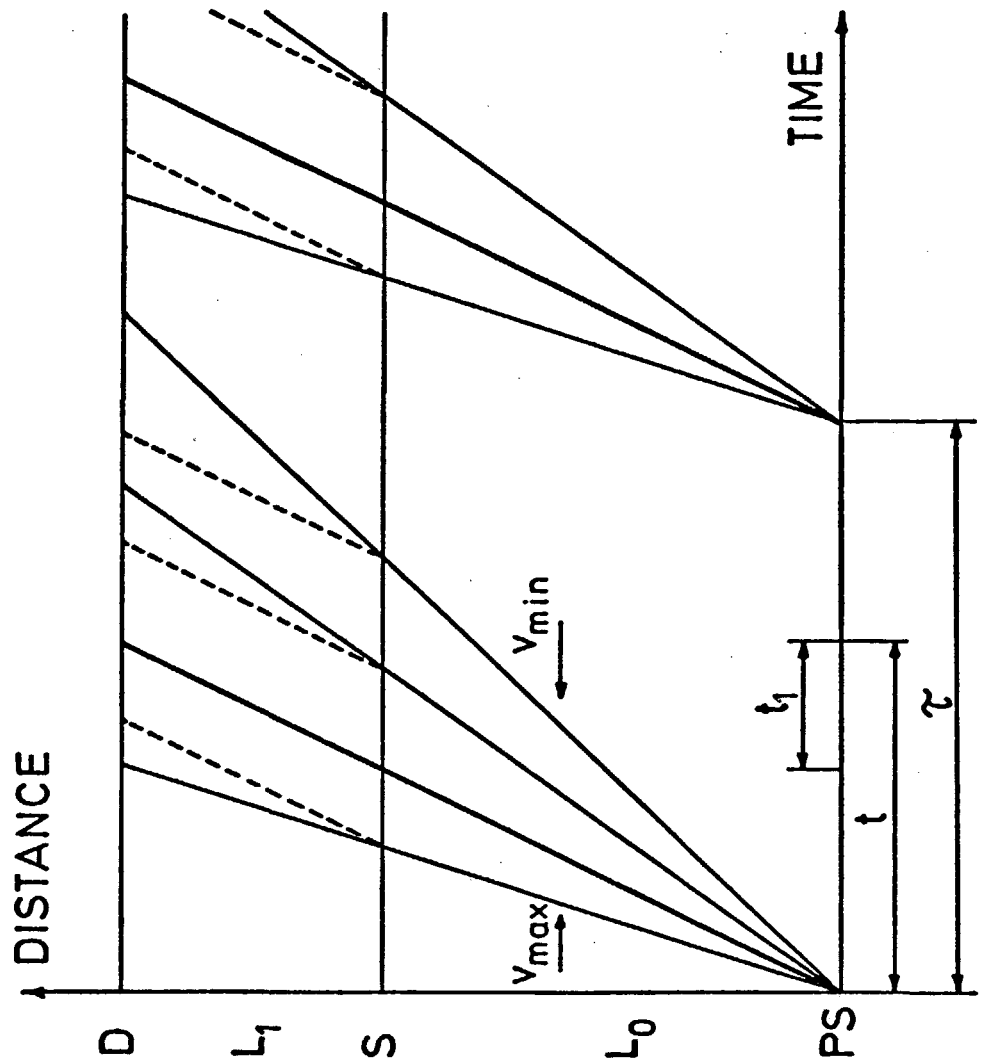


Figure 3: Space-time diagram of the neutrons propagating from the pulsed source (PS) to the detector (D). The broken lines correspond to the case of a crystal analyzer which selects out those scattered neutrons with a velocity  $v_1 = L_1/t_1$ . For elastic scattering this corresponds to one fixed incident velocity  $v = v_1$  (thick line).

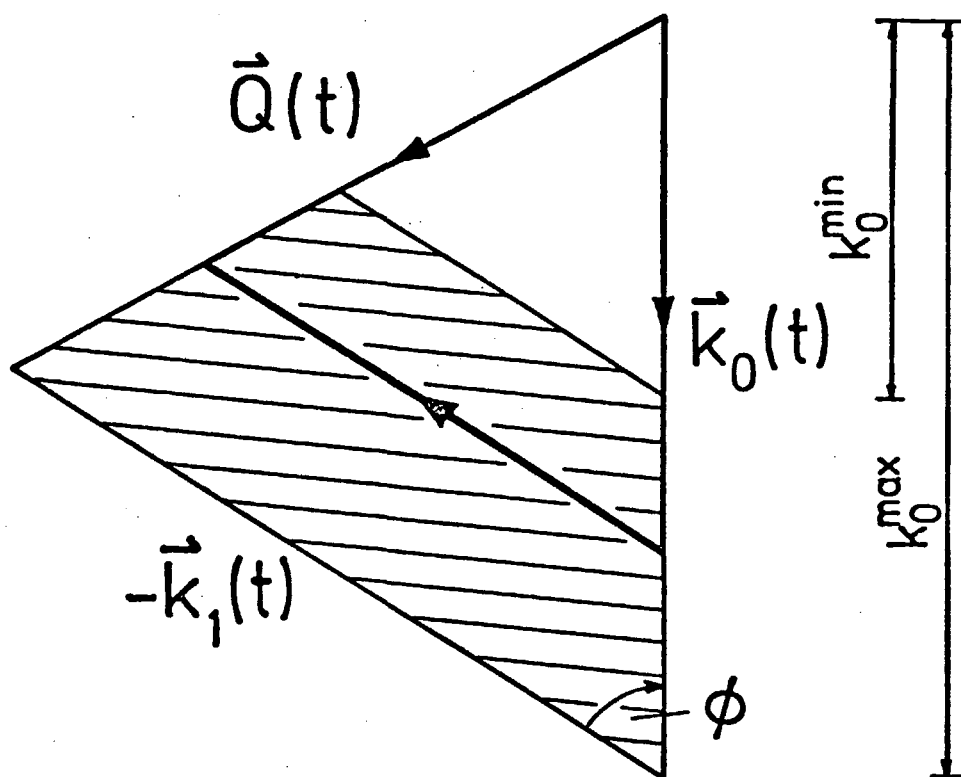


Figure 4: Reciprocal space scattering diagram of the proposed spectrometer for elastic scattering. Due to the variation in time of the scattered momentum vector all elements of the  $\vec{k}$ -space within the hatched area contribute to the detected intensity.

Polarizing Filters

by W.G. Williams, RAL Chilton, U.K.

These are based on the preferential absorption or scattering of one of the neutron spins by polarized nuclei. They all have the property that their polarizing efficiencies may be increased by increasing the filter thickness, though this is at the expense of a loss in transmittance.

a) Absorption Filters

The cross-sections for the absorption of the two neutron spin states  $\pm$  by a resonantly absorbing nucleus with polarization  $p_N$  is given by

$$\sigma_{A(\pm)} = \sigma_{A(0)} (1 \pm \eta p_N)$$

where  $\sigma_{A(0)}$  is a large unpolarized beam absorption cross-section and  $\eta$  is a statistical weighting factor which takes the values  $\eta = -1$  for  $(I - 1/2)$  compound nuclear spins and  $\eta = I/(I + 1)$  for  $(I + 1/2)$  compound nuclear spins. Any material containing a nucleus which can be highly polarized, and which has  $\eta \rightarrow 1$ , acts as an effective polarizer over its nuclear resonance energy regions since is generally the dominating cross-section at these neutron energies. This method relies on the ability to generate a large nuclear polarization, a non-trivial experimental problem, and its feasibility was first demonstrated by Freeman and Williams. Their method was to statically polarize  $^{149}\text{Sm}$  nuclei which were substituted as an approx. 6% dope in the paramagnetic insulating material cerous magnesium nitrate; the large hyperfine field at the nucleus ( $\sim 300$  T) was sufficient to create a high nuclear polarization ( $p_N \sim 0.9$ ) when the material was cooled to approx. 16 mK in a dilution refrigerator with a modest applied field ( $H \sim 0.1$  T). Measurements showed that a neutron polarization  $\sim 99\%$  was achieved near the resonance maximum at 98 meV at a transmittance  $\sim 30\%$  for the required spins. This filter performs well over the energy range 0-150 meV. Subsequently attempts have been made to produce a metallic Sm filter and extensive measurements on polycrystalline  $\text{SmCo}_5$  (permanently magnetized) have been carried out at the Harwell Linac. These have only been partially successful and an analysis of the data has shown that there was a considerable grain boundary depolarization in this material. More recently a paramagnetic filter

PdSm with Sm 2% has been tested; initial results are encouraging though the sample available had too low a Sm atomic thickness.

Other absorbing nuclei can be used e.g.  $^3\text{He}$ ,  $^{151}\text{Eu}$ , and  $^{165}\text{Ho}$  and many of these are effective in the epithermal neutron region. A detailed

evaluation of the potential offered by this technique has been published by Cywinski et al..

#### b) Scattering Filters

Polarized proton targets, which have been used by high energy physicists for nearly 20 years, can provide efficient polarizing filters for neutrons over a very wide energies range (E up to keV). The polarized proton scatters the neutron spins differentially by a factor  $\sim 6 : 1$  and for this application a proton polarization greater than 70 % is required. The most common method is that of dynamic polarization using some variant of the 'solid effect'. This utilizes the dipolar coupling between the electron spins in a paramagnetic dope and the proton spins in the host material and the experimental method is of the magnetic double resonance type i.e. it requires extremely stable applied fields and pumping (microwave) frequencies. The technique is difficult and expensive to run (it is at least beyond the capabilities of conventional neutron scattering teams), though it is being pursued at the Japanese pulsed source KENS. An alternative polarized proton target which uses the spin refrigerator' method has been investigated as a neutron filter at the Argonne National Laboratory. This uses a single crystal of yttrium ethyl sulphate (YES) doped with highly anisotropic ytterbium paramagnetic ions. The crystal is cooled to approx. 1 K and rotated in a magnetic field at a rate ( $\sim 100$  Hz) which allows the high electron polarization which is established at one crystal orientation to be transferred to the proton spin system as the crystal rotates. So far the method has only given low proton polarizations ( $\sim 42$  %).

A third type of polarized proton target uses the brute force polarization method and nuclear polarizations  $\sim 60\%$  have recently been achieved by Maschow et al. at Karlsruhe. The material was  $\text{Ti/H}_2$  which was cooled to  $\sim 10$  mK in an applied field  $\sim 9$  Tesla. The advantage of this static method is its experimental simplicity in comparison with dynamic methods.



# AN ACTIVE MONOCHROMATOR WITH MOSAIC CRYSTALS

Hans Friedrich

Physikalisch-Technische Bundesanstalt, 3300 Braunschweig

Triple axis spectrometers at a pulsed source have only a gain factor of 1. If it were possible to change the different velocities of the incoming neutrons to a common value and to a common direction, this would considerably increase the spectral intensity. Where mosaic crystals can reflect a large volume in  $k$ -space, moving crystals can change the velocity of neutrons. So we intend to combine these two elements.

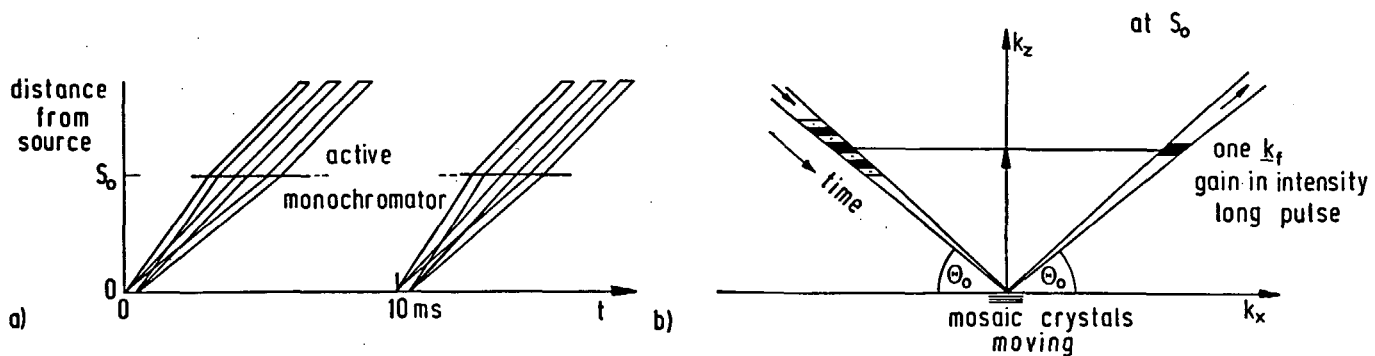


Fig. 1 Acceleration of white neutrons into a monochromatic and collinear beam. a) in space-time, b) in momentum space  $\underline{k}$

The action of a moving ideal Bragg-crystal is given by the following transformation rules. The  $\underline{\tau}$ -vector, here expressed as a velocity, is perpendicular to the lattice planes and defines the  $z$ -direction. As Doppler velocity  $v_D$  we denote only the  $z$ -component of the crystal velocity, because components parallel to the lattice planes have no effect.

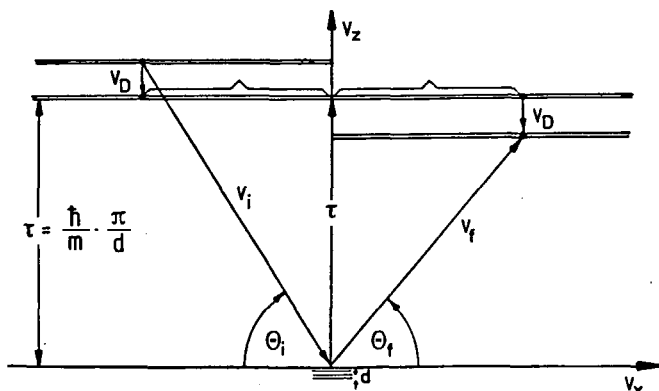


Fig. 2 A crystal moving with  $v_D \parallel$  to its reciprocal vector  $\tau$  transforms  $v_i$  into  $v_f$

## Transformation of neutron velocity

$v_i \rightarrow v_f$ :

$x$  component  $v_x$  conserved  
 $z$  component  $v_z$  reversed +  $2 v_D$

$\theta_f \neq \theta_i$

With these rules it is possible to transform any point in v-space to any other point if all 3 following parameters are free (fig. 3):

- 1. the lattice spacing :  $\tau$
- 2. the crystal velocity :  $v_D$
- 3. the orientation of  $\tau$  in v-space :  $\alpha$

The problem shown in fig. 1b, with fixed  $\theta_0$ , can only be solved if unfortunately all 3 parameters are free, i.e. many different crystals and a fast crystal changer are needed.

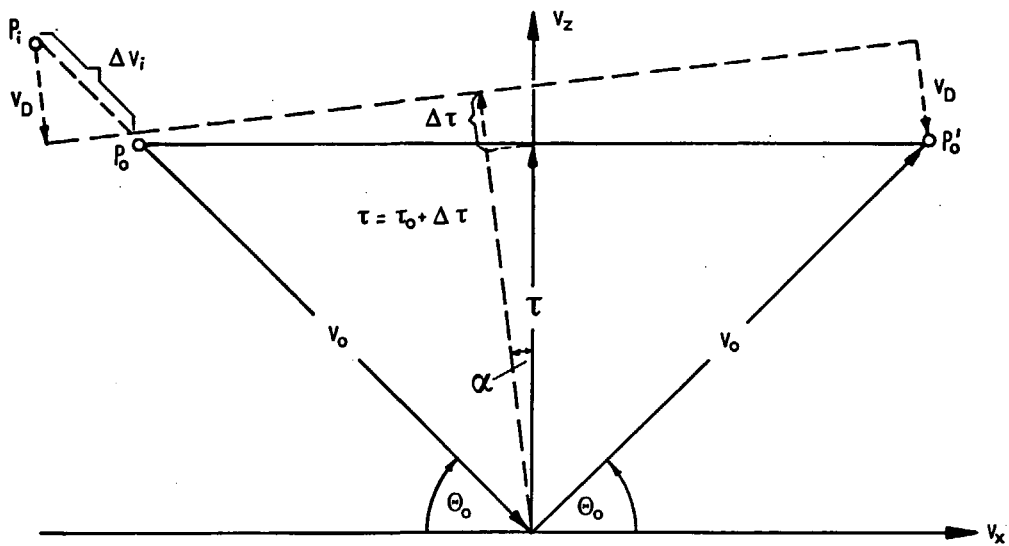


Fig. 3 Bragg condition for a crystal at rest ( $P_0 \rightarrow P_0'$ , solid line, index<sub>0</sub>) and for a moving crystal ( $P_i \rightarrow P_0'$ , broken line) able to transform any point  $P_i$  of a fixed incoming direction to the same  $P_0'$  of a fixed outgoing direction in space.

Applying the transformation rules of fig. 2 in a v-frame tilted by an angle  $\alpha$  (broken line in fig. 3), one gets approximately linear relationships (symbols shown in fig. 3):

Table 1

|                                                                        | $\theta_0 = 30^\circ$                 | $\theta_0 = 45^\circ$              | $\theta_0 = 90^\circ$  |
|------------------------------------------------------------------------|---------------------------------------|------------------------------------|------------------------|
| $\Delta v_i = \frac{2}{\sin \theta_0} \cdot \Delta \tau$               | $4 \cdot \Delta \tau$                 | $2,83 \cdot \Delta \tau$           | $2 \cdot \Delta \tau$  |
| $v_D = - (1 + \frac{1}{\text{tg}^2 \theta_0}) \cdot \Delta \tau$       | $-4 \cdot \Delta \tau$                | $-2 \cdot \Delta \tau$             | $-1 \cdot \Delta \tau$ |
| $\alpha = \frac{1}{\text{tg} \theta_0} \cdot \frac{\Delta \tau}{\tau}$ | $1,73 \cdot \frac{\Delta \tau}{\tau}$ | $1 \cdot \frac{\Delta \tau}{\tau}$ | 0                      |

The fast crystal changer consists of 2 wheels. Wheel  $W_A$  of radius  $R$  rotates around the axis  $A$  and carries on its periphery the axis  $B$  of wheel  $W_B$  (fig.4).  $W_B$  has the same radius  $R$  and rotates with the same angular speed in opposite direction.

A series of 10 crystals or more, mounted on the rim of wheel  $W_B$ , will move through the point  $A$  with their orientation fixed in space. Neutrons impinging on  $A$  see these crystals consecutively, whose Doppler-active component  $v_D$  of the crystal velocity varies sinusoidally with time  $v_D = \omega R \cdot \sin \omega t$ .

The plane of the wheel should be perpendicular to that defined by the neutron beams, i.e. the rotation axis should be horizontal to avoid hindrance of different crystals.

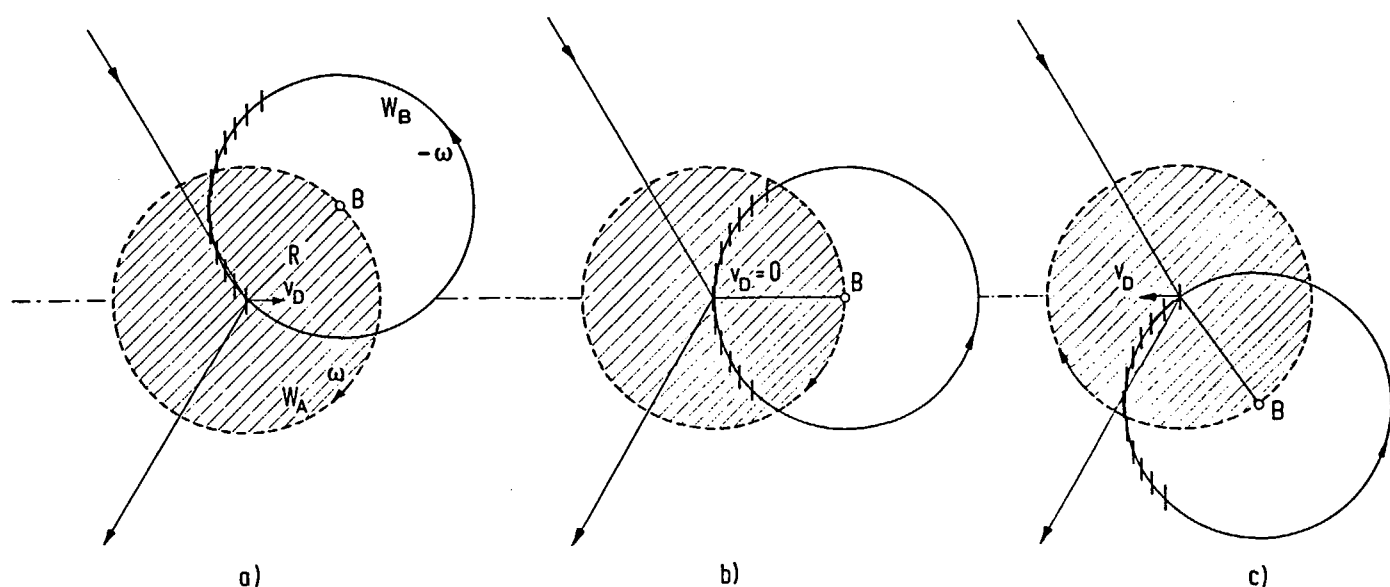


Fig. 4 Doppler-device and fast crystal changer shown at 3 extreme positions.

At the extreme position fig. 4b  $v_D = 0$ , i.e. the crystal velocity is perpendicular to  $\tau$ , the neutrons of velocity  $v_0 = \tau_0 / \sin \theta_0$  are reflected as if the crystal were at rest. Here  $\alpha = 0$ ,  $\Delta\tau = 0$ . At the time of position 4a (4c), where faster (slower) neutrons of a pulse arrive, the neutron velocity is changed to the same  $v_0$  if the Doppler velocity  $v_D$ , the lattice spacing, given by  $\tau + \Delta\tau$ , and the orientation  $\alpha$  of that individual crystal fulfil the relations of table 1.

A more or less realistic situation is given in fig. 5. We choose  $\text{Si}_x\text{-Ge}_{1-x}$  [111] mosaic crystals, which may become available in the future /1/. Any  $\tau$  between  $\tau_0 = 356,78 \text{ m/s} \pm 2,1 \%$  will then be available. Furtheron we choose  $\theta = 30^\circ$ . The  $v$ -resolution of the subsequent triple axis spectrometer is assumed to be  $\delta v/v = 0,5 \%$ , a typical value /2/, a resolution which can easily be realized with mosaic crystals and collimation.

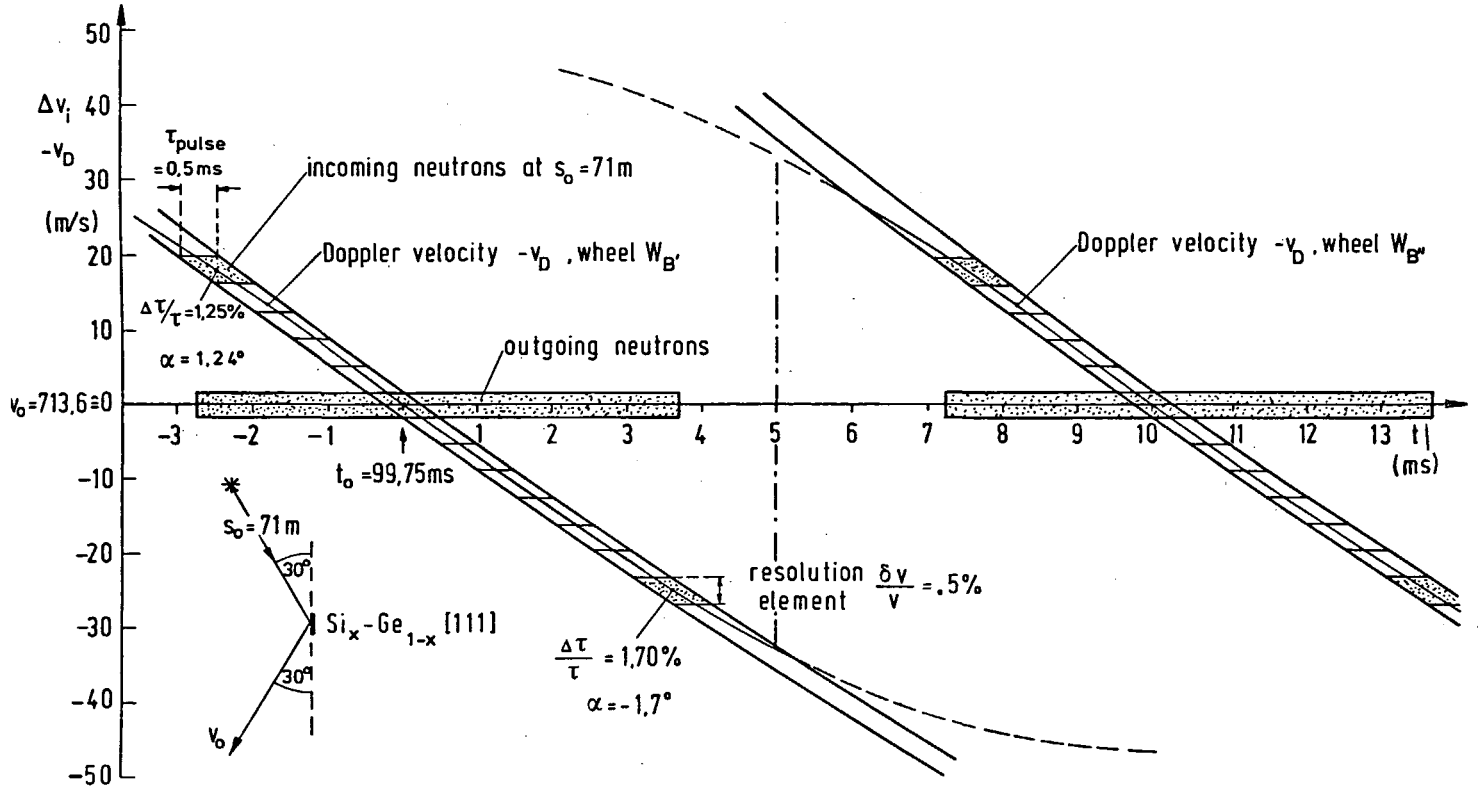


Fig. 5 Representation of incoming and outgoing neutrons in a  $v$ - $t$  diagram. The sinusoidal line represents the Doppler-velocity  $v_D(t)$  which transforms a wide velocity band into a final  $v_0$ . The variation of the lattice spacing  $\Delta\tau$  and the orientation  $\alpha$  for the first and for the last crystal is given. 65 % of the time between 2 pulses can be filled with monochromatic neutrons.

At a distance  $s_0 = 71 \text{ m}$  neutrons of  $v_0 = 713.6 \text{ m/s}$  arrive at  $t_0 = 99,75 \text{ ms}$ . The incoming neutron band in the  $t$ - $v$  diagram (fig. 5) is divided in several resolution elements defined by the individual mosaic crystals. As a criterion for a proper distances  $s_0$  we postulate, that the time distance between the centers of two adjacent resolution elements should be equal to the pulse width  $\tau$  pulse of the neutron pulse. This leads to

$$s_0 = v_0^2 \cdot \frac{\tau_{\text{pulse}}}{\delta v}$$

The Doppler velocity- $v_D(t)$  should fit for a long time into the band of incoming neutrons (for  $\theta = 30^\circ$   $\Delta v_i = -v_D$ ), which favours a low frequency  $f$  of the wheels. If, for example, the repetition rate of neutron pulses is 100 Hz and  $f = 25$  Hz, then by applying 4 wheels  $W_B$  instead of one (radius  $R = 30$  cm), each neutron pulse could be handled (fig. 6). No collisions of crystals occur, if the angular range, where crystals are mounted on each wheel  $W_B$ , is  $< 90^\circ$ .

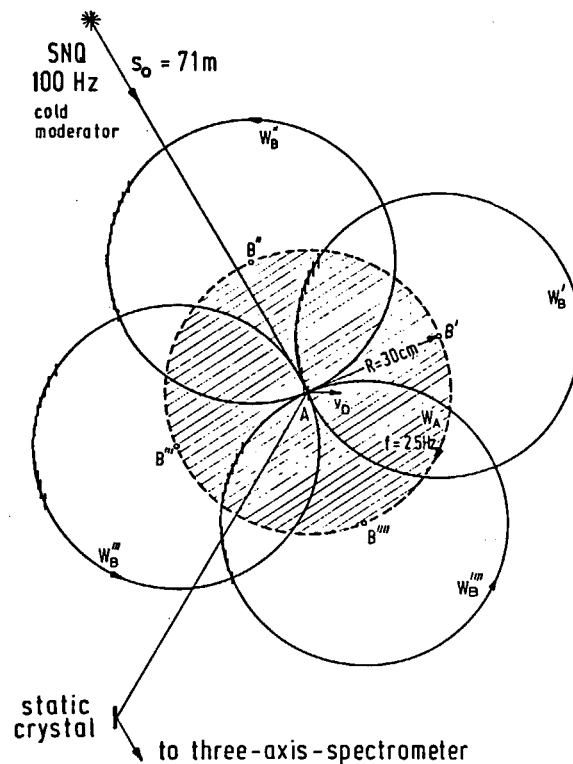


Fig. 6 4 wheel arrangement in a position comparable to fig. 4a.

The amplitude of the Doppler velocity needed in our example is 45,4 m/s. A better fit for the first and the last crystal can be achieved if the amplitude of  $v_D$  is slightly increased by about 2,5 %. In that case 13 crystals can be installed, the values of  $\tau$  and  $\alpha$  ranging linearly from  $\Delta\tau/\tau = 1,25$  % to -1,70 % and from  $\alpha = 1,24^\circ$  to  $-1,70^\circ$ . In this way monochromatic neutrons are produced during  $13 \times 0,5$  ms = 6,5 ms which is 65 % of pulse interval of 10 ms. Over- or undertuned neutrons can be cut away by one subsequent crystal at rest ( $\Delta\tau = 0, \alpha = 0$ ).

The radius  $R$  limits the useful height  $h$  of each crystal, in our example unfortunately only  $h \approx 2,5$  cm. So a suitable place for the instrument should be the end-position of a neutron guide where only about 2,5 cm useful height of the beam is left, due to other preceding users. If the full height of a neutron guide of about 10 cm is available then focussing by a large monochromator is by far easier.

As far as mosaic crystals with  $\Delta\tau/\tau \geq \pm 2\%$  are available and Doppler-machines with  $v_D \approx 45$  m/s are possible, the gain factor relative to 1 crystal of the same size at rest should be 13 in the ideal case or rather 7 in a more realistic case.

- /1/ Magerl, A., Heidemann, A., Hahn, C., Sirtl, E. ILL int. Scient. Rep. Nr. 84MA24T
- /2/ Dolling, G. in Dynamical Prop. of Sol. Vol.1, Horton u. Maradudin Eds, NH 1974, p. 550

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# POSITION-SENSITIVE DETECTORS FOR NEUTRON SCATTERING DEVICES AT THE SNQ

by W. Schäfer

Mineralogisches Institut der Universität Bonn  
Außenstelle für Neutronenbeugung in der KFA Jülich

## 1. Introduction

The detector system available for recording the scattering process is of decisive significance for the efficiency of a neutron scattering device. It is essential to collect the scattering information transmitted from a sample i.a. into the entire solid angle  $4\pi$  as quickly and completely as possible in terms of space and time. This can be accomplished by position-sensitive neutron detectors. Quantitative neutron detection is effected indirectly through conversion reactions initiated by neutron capture. The sufficiently energetic secondary products of these reactions will cause either ionizations or scintillations manifesting themselves as gas discharges or flashes of light. Position-sensitive detectors of adequate detection efficiency make use of the following nuclear reactions:  $^3\text{He}(n, p)^3\text{H}$ ,  $^6\text{Li}(n, \alpha)^3\text{H}$  and  $^{10}\text{B}(n, \alpha)^7\text{Li}$ . Decisive for the performance of a detector is the product of cross-section and density of conversion atoms as well as the efficiency of the read-out system for detecting the conversion reaction.

The level of development reached for position-sensitive neutron detectors to date is extensively described in the volume "Position-Sensitive Detection of Thermal Neutrons" published by P. Convert and J.B. Forsyth in 1983 /1/. It summarizes the results of the ILL Workshop held in October 1982 under the same title, which was attended by a large number of working groups concerned at present with the development and application of position-sensitive neutron detectors. In addition, the volume contains detailed information on the basic data, technical realization and application of these detectors and their coupling to data storage systems in conjunction with

a comprehensive bibliography. The aim of this paper, which was stimulated by the SNQ workshop in Bad Kissingen in 1984, is a critical review of position-sensitive detector systems with regard to the specific circumstances and requirements of their use in connection with SNQ neutron scattering devices. Emphasis will be placed on the possibilities attained to date and those realistically achievable on the basis of current results, specifying as well the intrinsic limits of position-sensitive gas and scintillation detectors. Particular attention will be given to scintillation detectors whose development has only been intensified by several working groups in the last five years. This will be compared with the development and experience made with gas detectors for more than 30 years, which more or less have reached their technological peak level today.

## 2. Requirements to be Met by Detectors

Position-sensitive neutron detectors can be characterized by a large variety of typical features. As far as the physical specifications are concerned, the following properties are of major importance: detection sensitivity, position resolution, uniform performance across the entire detector surface, stability, time resolution of successive events and the handling of high count rates as well as gamma sensitivity and other background effects. Of further practical significance is the geometrical design, mechanical reliability, low maintenance and repair and last but not least the cost of manufacture.

The suitability of position-sensitive detectors for SNQ experiments is primarily governed by their suitability for measurements in a pulsed white neutron beam. Their detection sensitivity should extend over a broad energy spectrum ranging from about 0.001 eV to 10 eV ( $\approx 9$  Å to 0.09 Å). A new quality feature as compared to the experiments with reactor neutrons is, in particular, the suitability for detecting short-wave neutrons up to the epithermal energy range.



SNQ experiments will be essentially time-of-flight experiments. Detectors suited for time-of-flight analyses should be as thin as possible (a few mm) in the direction of flight in order to eliminate inaccuracies in the length of the neutron flight path; moreover, the decay times for the neutron pulses in the detector should be short and identical for all amplitudes in order to avoid inaccuracies due to different retardation effects.

The demand for short decay times or minimum dead time is also decisive for the time resolving power of a detector which must satisfy high requirements due to the high neutron count rates in the pulsed SNQ peak flux and which will also govern the degree of SNQ utilization. A processing capacity of  $10^6$  n/s will certainly not be the maximum requirement for single crystal structure analyses.

High-precision measurements with position-sensitive detectors require a high angular resolution with a variable spacing between sample and detector. A value of 1.0 to 0.5 mm should be aimed at for the position resolution of a detector in order to achieve sufficient resolution even at short distances. The detector surface for a given angular scattering range can be reduced as the position resolution is improved.

Well aware of the different physical requirements to be met in terms of quantity and weight by position-sensitive detectors for the different neutron scattering experiments - small-angle scattering, liquid scattering, powder diffractometry, single crystal structure analysis, elastically and inelastically diffuse scattering, neutron spectroscopy - the feasibility of detectors with the following specifications will be examined below:

|                        |               |
|------------------------|---------------|
| detection efficiency:  | 50 % at 0.3 Å |
| position resolution:   | below 1 mm    |
| count rate processing: | $10^6$ n/s    |
| sensitive depth:       | below 5 mm    |
| gamma sensitivity:     | $10^{-6}$ .   |

### 3. Performance Spectrum of Gas Detectors

#### 3.1. Detection sensitivity

Gas detectors operated as proportional counting chambers exploit the absorption cross-sections of 3000 (5400) barn for  $^3\text{He}$  gas and 2100 (4000) barn for  $^{10}\text{BF}_3$  gas - related to neutrons of 1.0 Å (1.8 Å). The cross-sections are proportional to  $1/v$  in the thermal range. The detection efficiency is determined by the product of gas pressure (atm), gas volume (= depth (cm) of the detector chamber) and neutron wavelength (Å). A value of 21 atm · cm · Å and of 30 atm · cm · Å is required e.g. to achieve an 80-percent neutron detection for  $^3\text{He}$  gas and  $^{10}\text{BF}_3$  gas, respectively (Fig. 1). High detection probabilities therefore require high gas pressures (and possibly the admixture of slowing-down gases) or deep detector chambers. Larger position-sensitive  $^3\text{He}$  detectors are built up to pressures of about 15 atm, which is approximately the intrinsic technological limit because of the window thickness required (10 mm). The intrinsic restriction for  $^{10}\text{BF}_3$  detectors is about 3 atm due to the beginning recombination of ion pairs produced after neutron absorption and required for detection. Typical values for the detection efficiencies of existing gas detectors are 50 % at 2.5 Å or 30 % at 1.2 Å in the  $^{10}\text{BF}_3$  case and 60 - 70 % at 1.2 Å in the  $^3\text{He}$  case.

#### 3.2 Position resolution

The intrinsic restriction of position resolution for gas detectors is the range of the charged secondary particles produced during the neutron-capture reaction. The limit for the so-called ionization centroid is about 3 mm for  $^{10}\text{BF}_3$  gas of 1 atm (1.5 mm at 2 atm). The proton produced in  $^3\text{He}$  gas has a substantially larger range - about 80 mm at 1 atm; the range is drastically reduced by higher gas pressures and the admixture of slowing-down gases (e.g. Xe,  $\text{CF}_4$  or  $\text{C}_3\text{H}_8$ ). These foreign gases, however, will cause at the same time a reduction in detection sensitivity due to the recombination of charge carriers and lead to a certain gamma sensitivity (cf. chapter 3.4). The range in  $^3\text{He}$  at 10 atm is about 1.5 mm with added gas and 3 mm without gas addition.

For gas detectors designed as multi-electrode chambers, the electrode spacing achievable is an additional technological barrier to position resolution. The older "banana" design operated with  $^{10}\text{BF}_3$  gas of 1 atm and featuring 400 cells has an anode wire spacing of 5 mm with an effective position resolution of 5.3 mm; the wire spacing of the new "banana" consisting of 200 cells and operated with 2 atm  $^{10}\text{BF}_3$  gas is 2.5 mm with an effective position resolution of 2.62 mm (Roudaut /2/). The multiwire detectors with  $^3\text{He}$  also have a position resolution of about 3 mm (Jacobé et al. /3/). A better resolution can be achieved with the analog resistance wire technique. A two-dimensional detector with RC position coding at 5 to 6 atm  $^3\text{He}$  gas developed in Brookhaven has a position resolution of 1.26 mm (Fischer et al. /4/); a similar value is aimed at in Oak Ridge with a  $^3\text{He}$  detector in which the position analysis is effected by time difference measurement of the pulse rise times (Valentine et al. /5/).

### 3.3 Time resolution and time-of-flight suitability

The processing time required for a single event in the gas detector is governed by the charge collecting and discharge time and by the speed of the read-out system. The dead times for one-dimensional multi-electrode chambers are between 2 and 6  $\mu\text{s}$  and in the range of 10 to 20  $\mu\text{s}$  for two-dimensional systems with resistance wire technique. The intrinsic counting rate restriction is around  $10^5$  events/s. The suitability of gas detectors for time-of-flight measurements is limited by the detector depth required for an appropriate detection sensitivity. The detector chambers working with  $^3\text{He}$  pressurized gas require a gas depth of 10 to 20 mm involving certain inaccuracies as to the actual detection position and the flight path covered by the neutron (parallactic error).

### 3.4 Background and gamma discrimination

The signal/background ratio of gas detectors is governed by the charge produced per neutron capture, i.e. the number of

electrons, and by the discrimination capability in the pulse height spectrum. Particularly favourable are the ratios for  $^{10}\text{BF}_3$  gas with 60,000 electrons per absorbed neutron ( $\approx 10^{-14}$  Cb) and a deep and wide gap between the amplitudes from background and signal electrons. Approximately 1/3 less electrons are produced in  $^3\text{He}$  gas, but there is an excellent discrimination capability between neutron and background pulses in this case, too. An appreciable gamma sensitivity is caused for  $^3\text{He}$  detectors by the admixture of slowing-down gases, by wall effects in the detector casing and by the relatively thick windows. The gamma sensitivity is  $10^{-5}$  for  $^{10}\text{BF}_3$  detectors and  $10^{-6}$  for pure  $^3\text{He}$  gas. Background values of 1 event/h  $\cdot$  cm<sup>2</sup> are achievable.

### 3.5 Other properties

The position-sensitive gas detectors also possess a homogeneous and uniform sensitivity across the entire detector surface ( $\pm 3\%$ ) and a good long-time stability with regard to position and intensity measurements. Roudaut /2/ measured a relative stability of  $2 \cdot 10^{-3}$  between the cells of a multi-electrode detector over a period of six months. Technological restrictions are encountered for gas detectors in connection with the dimensioning of the high-pressure chambers (roughly 60 cm) and the neutron window thickness required (10 mm); the geometrical flexibility is relatively small. The detectors do not easily lend themselves to repair due to the gas technology involved, but the systems are well understood and operate reliably as a result of many years of experience.

## 4. Performance Spectrum of Scintillation Detectors

### 4.1. Schematic design principle

There are two techniques of position resolution for scintillation detectors which are shown schematically in Fig. 2. In analogy to the technique of gas detectors, a distinction is made between

digitalized multi-element systems composed of many identical single elements, corresponding to the multi-electrode gas detectors, and the analog signal system of a detector of continuous design derived from the Anger technology of X-ray detectors, corresponding to the resistance wire technique of gas detectors.

In the basic single-element version, each scintillator strip (=resolving element) is connected to a separate photomultiplier via a light guide, and the scintillation light produced by neutron absorption in the detection medium is identified in this photomultiplier (cf. 4.2). However, this design is unattractive for a position-sensitive detector with many single elements due to the large number of photomultipliers required. The number of photomultipliers can be drastically reduced by means of an optical coding system with several optic fibres outgoing from each single element. The correlation between the number of resolving elements  $m$  and the number of multipliers required  $n$  is

$$m = n! / 2! (n-2)!$$

for dual coding and

$$m = n! / 3! (n-3)!$$

for triple coding.

Triple coding could theoretically serve to resolve 1140 detector elements with 20 multipliers. Such optimum coding is not practicable because of the complex light guide system required - 171 light guide bunches would have to be conducted to each photomultiplier in the case of triple coding with 1140 elements. It is therefore necessary to optimize the number of photomultipliers as well as the quantity and routing the light guide bunches for a given number of single elements (the more multipliers, the less light guides to a single multiplier). A double coincidence technique proposed by Kurz et al. /7/ basically permits position coding with  $2n$  multipliers for  $n^2$  elements; a simplified light guide system requires 60 multipliers for 400 detector elements.

This technology is basically characterized by a high degree of flexibility for the detector design and the read-out electronics required is relatively simple. On the other hand, the mechanical detector setup is very sophisticated because of the complex light guide system of either rigid plexiglass rods or flexible fibre glass bunches. Prior to considering in detail the physical detector properties in the chapters below, the essential physical problem involved in this type of construction may already be outlined here as follows: the percentage of light produced by neutron capture in the scintillator and reaching the photomultiplier via the light guide system is relatively small, which drastically reduces the efficiency and gamma discrimination in the detector.

The alternative technology is based on a linear scintillation band or, in the two-dimensional case, on a continuous scintillation surface applied onto a glass light coupler (disperser), behind which a most densely packed arrangement of photomultipliers contacts optically with their cathode surfaces. Position analysis of the scintillation event succeeding each neutron capture is effected by measurement of the corresponding distribution of light in the photomultipliers. First of all, the central multiplier, i.e. the multiplier with the highest anode current, is determined for each event, and the exact position is then determined from the intensity distribution of the immediately adjacent multipliers according to

$$q_k(x) = \frac{I_{k-1}(x) - I_{k+1}(x)}{I_{k-1}(x) + S_k I_k(x) + I_{k+1}(x)},$$

in which  $k$  is the multiplier with the highest anode current,  $x$  the scintillation position,  $I$  the anode current and  $S$  a correction factor for linearizing the position analysis. A high light output and a concentration (bunching due to total reflection at the air gap) of the scintillation light onto a limited number of photomultipliers is achieved by means of a light reflector at the front of the scintillator, an air gap between the scintillator and the glass light coupler

and by an optimum adaptation of the disperser thickness and cathode surface of the multiplier (cf. Fig. 2b). This detector design is mechanically rather simple, but requires sophisticated and high-speed electronics. The physical performance features will be discussed in the following chapters.

Within the scope of cooperation between the KFA Jülich (R. Kurz, ZEL and J. Schelten, IFF/SNQ) and our Institute (cf. Annual Reports /6/), one prototype detector each was built according to the single-element principle /7/ and with the continuously operating analog signal system /8, 9, 10, 11/ and tested at the DIDO reactor in Jülich. Based on this experience, a large linear position-sensitive detector system was then built using the continuous detection and signal processing technique for the powder diffractometer of our Institute in Jülich. The detector system has been in routine measurement operation for a good twelve months now /12, 13, 14/ (Fig. 3). A two-dimensional scintillation detector also of analog design is installed on a Laue single-crystal diffractometer in Argonne /15, 16/ (Fig. 4); moreover, this technology is being pursued and further developed in Los Alamos /17/. A working group in Rutherford has been engaged for many years in the development of scintillation detectors /18/; their studies relate to the single-element design with flexible light guide coding.

#### 4.2 Detection sensitivity

As far as scintillation detectors are concerned, whose development has only just begun, two solid-state scintillators have been presented as candidates to date for position-resolving neutron measurements: amorphous Cer-doped  $^6\text{Li}$  silicate glass (density  $2.5 \text{ g/cm}^3$ ) and europium-doped  $^6\text{LiI}$  in single-crystal form. Most experience has been gathered so far with  $^6\text{Li}$  glass.  $^6\text{Li}$  glass and  $^6\text{LiI}$  have comparable linear attenuation coefficients for neutrons (for  $^6\text{LiI}$   $\Sigma = 17 \text{ cm}^{-1}$ ). The basic constituents of the scintillator glass are  $\text{SiO}_2$ ,  $\text{CeO}$  and  $\text{Li}_2\text{O}$  /19/, the  $^6\text{Li}$  fraction being about 7 to 8 wt %. The absorption cross

section for  $^6\text{Li}$  is 520 barn (910 barn) for neutrons of 1.0 Å (1.8 Å). The high absorber atom density in the solid (as compared to the gas) leads to the neutron detection probabilities plotted in Fig. 5 for glass thicknesses of 1 to 4 mm.  $^6\text{Li}$  glass of 2 mm thickness still has e.g. a detection probability of 60 % for neutrons of 0.5 Å. The detector installed in Jülich uses 1 mm glass with a detection probability of 70 % for 1.3 Å. Conditions are even more favourable for the  $^6\text{LiI}$  single-crystal scintillator with an even higher  $^6\text{Li}$  atomic density.

As far as the light output is concerned, amorphous  $^6\text{Li}$  glass is a poor scintillator. Approximately 5300 photons with a light wavelength of 400 nm are produced per captured neutron; in comparison, this only accounts for 2.6 % of the light output of an NaI(Tl) crystal for gamma rays. Conditions are more favourable for monocrystalline  $^6\text{Li}$  glass /20/. Considering the quantum efficiency of the photomultiplier cathode surface, which is in the range of 28 %, a maximum of approx. 1500 electrons per neutron are available in the case of  $^6\text{Li}$  glass.

Since the scintillator light is emitted to the full solid angle  $4\pi$ , only part of this maximum possible number of electrons can be realized on account of the detector geometry. About 700 to 800 electrons are produced per neutron in the case of continuously operating detectors with  $\text{Al}_2\text{O}_3$  reflector and an air gap between scintillator and disperser (see Fig. 2b) /16/. The resulting anode currents of the photomultipliers permit an adequate background and gamma discrimination (cf. section 4.5) and ensure that the absorption probability of the neutrons in the scintillator is also identical with the detection sensitivity of the detector.

Conditions are much more unfavourable for the single-element design with light guide coding. Only 17 % of the photons produced during neutron absorption are logged in the case of dual coding with a relatively large cross-sectional area



of  $1 \text{ cm}^2$  of a rigid light guide. Added to this are further light losses on the partially very long (1 m and more) light guide paths to the photomultiplier, on the coupling pieces between scintillator and light guide and between light guide and photocathode of the multiplier (cf. Fig. 2a). Only about 10 electrons per neutron were ultimately available in the prototype detector with rigid plexiglass light guides tested by us. Conditions would deteriorate even further in the case of triple coding because of the additional separation of the light. The use of flexible fibre glass light guides only provides very small light guide cross sections (approx. 1 to  $3 \text{ mm}^2$ ), so that an extremely unfavourable conversion of scintillator light into electrons will also result in this case (approx. 1 electron per neutron). The detection sensitivity of the detector achieved with this technology may be lower than the absorption probability in the scintillator.

#### 4.3 Position resolution

The intrinsic physical limit of position resolution in a scintillation detector is governed by the range of the ionizing particles generated during the neutron reaction process in  ${}^6\text{Li} - {}^6_3\text{Li}(n,\alpha){}_1^3\text{T}$ . Triton and  $\alpha$ -particles move apart at an angle of  $180^\circ$ , the triton with its energy of 2.72 MeV covering a path length of  $40 \text{ }\mu\text{m}$  and the  $\alpha$ -particle with an energy of 2.04 MeV a path length of  $7 \text{ }\mu\text{m}$ . The path lengths of the electrons produced by both particles until absorption in the activator atom are  $0.0648 \text{ }\mu\text{m}$  and  $0.0196 \text{ }\mu\text{m}$ , respectively /21/. The intrinsic resolution limit is therefore around  $50 \text{ }\mu\text{m}$  as compared to about 1.5 mm in the case of gas detectors. While the particle range is the limiting element in a gas detector, this intrinsic restriction is of no significance for the required resolving power of a scintillation detector.

The technologically achievable position resolution is governed by the size of the photomultipliers (the half-width of the signal function is proportional to the dimension of the multiplier), by the total amount of photoelectrons produced

(depending on the quantity of light in the scintillator, on the optical transmission of the scintillator and disperser, on the photocathode coverage of the scintillator surface and on the quantum efficiency of the photomultipliers) and by the distribution of the total light to the individual multipliers (distribution to as few multipliers as possible). The statistics of photon detection limit position resolution to about  $1/45$  of the multiplier spacing /17/.

The linear scintillation detector installed on the powder diffractometer in Jülich has a resolution (half-width of the signal function) of 2.5 mm; it is equipped with round multipliers of 30 mm diameter and 1 mm thick  $^6\text{Li}$  glass. At present, a new prototype detector is under construction which, for the first time, will be provided with a  $^6\text{LiI}(\text{Eu})$  single crystal as the scintillator. Owing to the higher light output, the resolution will be better by about a factor of 3.5, amounting to 0.7 mm, while using the same multipliers (30 mm).

The two-dimensional scintillation detector operated in Argonne covers an area of  $30 \times 30 \text{ cm}^2$  and uses a  $7 \times 7$  photomultiplier arrangement with square cathode surfaces of 51 mm edge length. The scintillator is 2 mm thick  $^6\text{Li}$  glass. The position resolution measured is 2.7 mm. The resolution could be improved by a factor of 2 when using multipliers with 28 mm cathodes; if a  $^6\text{LiI}(\text{Eu})$  scintillator were used, the resolution would be between 0.5 and 1 mm with the 51 mm multipliers installed /16/.

The plane detector under construction in Los Alamos has been designed for a size of  $25.5 \times 31.0 \text{ cm}^2$  and will be equipped with 49 hexagonal multipliers of 60 mm diameter. This detector will have 15,000 resolving elements of  $2.3 \text{ mm}^2$  each when  $^6\text{Li}$  glass is used /17/.

In the single-element system operated in Rutherford with light guide coding the resolution is governed by the dimension of the individual element. Each single element requires at least 2 (dual coding) or 3 light guide outlets, so that it is hardly possible to remain below a minimum size of 3 mm.

A linear prototype detector was built from 100 single elements of 3 mm width each and 0.1 mm spacing comprising two 1 mm thick  $^6\text{Li}$  glass strips each. Flexible fibre glass light guides of 1 mm diameter each were used for triple coding with a total of 12 photomultipliers /18/. A two-dimensional detector with triple coding and scintillator elements of  $10 \times 10 \text{ mm}^2$  is under construction for a single-crystal diffractometer at KENS, Japan.

#### 4.4 Time resolution and time-of-flight suitability

The intrinsic limit for the duration of a single event or for the dead time of two consecutive single events is the duration or decay time of the scintillation pulse. It is essential for a good scintillator that the excitation energy from the activator atom is given off immediately as a scintillation, that the major portion of the excitation energy is emitted as light and that the scintillator material is transparent for scintillator light, i.e. that e.g. the ground and excitation states of the activator are situated in the band gap between the valance and conduction bands of a crystal. The neutron scintillators examined to date (single crystals, mineral glasses, gases, organic scintillators) present an unfavourable relation between light output and decay time of the light in that shorter decay times also involve smaller light output. Consequently,  $^6\text{Li}$  glass has a decay time of about 60 ns, whereas the monocrystalline  $^6\text{LiI}(\text{Eu})$  has a decay time of 1.4  $\mu\text{s}$ .

In addition to this intrinsic pulse duration, the sequential electronics plays a decisive role for the actual pulse processing time in scintillation detectors. According to the high-speed process electronics proposed by Seeger /17/ for a two-dimensional Anger detector, the time for the formation of the analog sum signal (cf. the formula in 4.2) for the position analysis is 150 ns and the time for digitalizing the signal with flash ADCs is 100 ns. By dividing the multipliers into several subunits, events in separate subunits can be processed simultaneously, so that the time for a multifield analysis is approx. 400 ns in total.

For  ${}^6\text{LiI}$  with its relatively long decay time of 1.4  $\mu\text{s}$ , the anode signal can be shortened to 150 ns by means of a shaping amplifier with integral and differential time constants of 20 ns. After test measurements by Kurz and Schelten in Jülich /22/, a double-pulse time resolution of about 150 ns was achieved with the aid of a high-speed window discriminator. Count rates of  $5 \cdot 10^5/\text{s}$  can be processed without appreciable dead time losses. A dead time of 100 ns was estimated for a triple coincidence arrangement in connection with the electronically less sophisticated light guide technology.

The particular suitability of scintillation detectors for time-of-flight technology is due to the electronically feasible low dead time in analyzing time-peaked spectra as well as to the thin scintillation layer (approx. 2 mm) with negligible errors concerning the detection position of the neutron in the direction of flight.

#### 4.5 Background and gamma discrimination

In addition to the neutrons which, after their interaction with the  ${}^6\text{Li}$  atoms, produce electrons through the ionizing effect of  $\alpha$ -particles and triton as triggers for the scintillation capability of the activator atoms, the gammas also interact with all the atoms in the scintillator and produce electrons (1 to 2 KeV energy with gamma energies of a few MeV) which can also excite the activator centres for light emission. In the multiplier pulse height spectrum, which is characterized by different light outputs ( $\hat{\text{amplitude}}$   $\hat{\text{multiplier current}}$ ) in the various interaction processes, the amplitude of the "neutron light" still overlaps with the amplitude of the "gamma light" in the case of energetic gammas (Fig. 6). A good scintillator is distinguished by a narrow neutron peak of high intensity, which is shifted to highest possible amplitudes in the pulse height spectrum in order to eliminate background and gamma pulses at the

foot of the neutron peak by a pulse height discrimination. The high-purity  $^6\text{Li}$  glass available today involves a neutron peak width of approx. 12 to 15 % (FWHM) and the pulse height corresponds to an electric charge of approx.  $0.05 \cdot 10^{-14}$  Cb with approx. 1500 electrons/neutron in the photomultiplier; in comparison, the charge produced in gas counters is by more than one order of magnitude higher (cf. 3.4). The gamma sensitivity of  $^6\text{Li}$  glass was reduced to a value of about  $10^{-4}$  by means of the pulse height analysis of the scintillation events (threshold at the foot of the neutron peak). This figure relates to gammas of 1.2 MeV of a  $^{60}\text{Co}$  source and was realized on the detector in Jülich as well as in Argonne. Due to the light output being by a factor of 12 higher ( $\hat{=}$  pulse height) in the  $^6\text{LiI}$  scintillator, the neutron peak is shifted to greater pulse heights providing a considerably more favourable possibility for gamma discrimination through the pulse height. Signals of gammas with energies up to 4 MeV can be discriminated by the neutron signals /22/.

A further, although electronically much more sophisticated, method for gamma discrimination is the pulse shape analysis of the scintillation pulses. This method makes use of the different pulse shapes of neutron- and gamma-induced events; gamma-induced pulses are about 6 % longer with an initial intensity of approx. 10 % /19/. However, the relevant development is still in the initial stage; when applying this method to  $^6\text{Li}$  glass, which is relatively low in light, about 50 % of the total events are lost according to the present state of the art along with a reduction of the gamma sensitivity by another two orders of magnitude. The pulse shape technique is promising, however, for the  $^6\text{LiI}$  scintillator where 550 effective photoelectrons are still left after shaping with a relative half-width of the neutron peak of 15 % /22/.

The pulse height discrimination practised on the detector operated in Jülich with a residual gamma sensitivity of the order of  $10^{-4}$  leads to a gamma background of 2 c/min  $\text{cm}^2$  with

a gamma dose rate of 1 mrem/h on the detector. The background caused by cosmic radiation is below  $0.5 \text{ c/min cm}^2$ .

The light guide coded scintillation detectors possess a high gamma sensitivity due to the small light and electron yield per absorbed neutron (cf. 4.2) and the resultant restriction for pulse height discrimination. A gamma sensitivity of 0.5 % at gamma energies of approx. 1.2 MeV is reported for the Rutherford prototype detector. An improvement to 0.04 % is possible in conjunction with a counting restriction to triple coincident events (simultaneous events through three light guides in three different multipliers). This method causes a further reduction of the detection probability to approx. 70 % /18/.

#### 4.6. Other properties

While the light guide coded design produces gaps in the responsivity of the detector of approx. 0.1 mm width between the individual elements, the Anger technology provides for a continuous read-out of the detector area and for an electronically free selection of the measuring point density which can be individually adapted to the resolving power of the detector. The manufacture of large-area scintillator plates still poses technical problems, their maximum dimension being restricted to approx. 15 cm at present. A larger detector surface has to be composed from several parts involving the possibility of discontinuities at the connection points which can be corrected by means of calibration measurements.

The response behaviour across the entire surface of a scintillation detector is uniform within approx.  $\pm 3 \%$  (channel to channel variation) after the detector has been calibrated by means of homogeneous neutron irradiation (e.g. incoherent vanadium scattering).  $^6\text{Li}$  glass has an excellent temperature stability up to  $500^\circ\text{C}$ . A position drift of less than  $0.05 \text{ mm}/^\circ\text{C}$  was measured in Argonne over a period of 20 to 40 hours.

The long-time stability of the physical detector properties is essentially governed by the constancy of sensitivity and the amplifier properties of the photomultipliers. To compensate for amplification variations or drifts (10 % change within 50 days were measured on a multiplier in Jülich /8/), the Jülich detector was equipped with a computer-driven control and stabilization system for amplification of the photomultiplier signals /9/. At regular intervals preselectable by means of the software, all photomultipliers are controlled using reference pulses of a light diode and, if required, readjusted automatically so as to ensure optimum long-time stability. A possible magnetic field dependence of the photomultipliers must be prevented by providing a magnetic shielding which, in the case of the Jülich detector, consists of a  $\mu$ -metal shielding around the entire photomultiplier arrangement.

The mechanical construction of the scintillation detector according to the Anger technology is uncomplicated and robust. Due to its modular design, the system is easy to maintain and repair. The detector needs only a thin Al neutron window of 1 mm thickness in a light-tight casing. There are restrictions with regard to the geometrical flexibility; for example, no bent detector has been developed up to the present. The detector using the light guide technology is geometrically much more flexible, but the detector construction is sophisticated due to the coupling, support and routing of the entire light guide system.

Finally, a short survey of the costs of present-day scintillation detectors. For the linear detector in Jülich of 140 cm effective length (divided into two identical units of 70 cm length with 24 photomultipliers each) the costs of hardware parts are about DM 150,000 including all electronics but without the computer. About 1/3 of this sum is needed for the detector box itself (scintillator and photomultipliers) and 2/3 for the electronics. It should be emphasized that costs of labour, development and testing are not included. An estimate by Seeger /17/ for the cost of a two-dimensional detector in Los Alamos with a sensitive area of  $25.5 \times 31.0 \text{ cm}^3$  and 49 hexagonal multipliers amounts to US\$ 45,000 (not including development).

## 5. Conclusions and Future Prospects

A comparative summary of the potentialities of scintillation and gas detectors with respect to the SNQ requirements is given in the diagram in Fig. 7. Considering the essential physical properties like detection efficiency, spatial resolution dead time and count rate capability, the intrinsic limitations of the scintillation detector are more favourable. Due to its thin detector zone of only a few mm depth the solid-state scintillation detector is especially suitable for TOF experiments. The physical specifications for a detector system defined in Chapter 2. can be realized at reasonable technical expense.

Additional efforts are necessary with regard to investigation and production of proper scintillation material. The poor light output of the  $^6\text{Li}$ -glass obtainable to date is the main obstacle to a more effective gamma discrimination below a value of  $10^{-4}$ . Spowart /19/ proposed a multilayer sandwich structure of the glass with alternating layers containing a high amount of  $^6\text{Li}$  and Ce and layers containing  $^7\text{Li}$  and gamma absorbing atoms (Eu, Tb) but no cerium. This construction pays regard to the different path lengths of the ionisation electrons produced by the neutron and gamma absorption and inducing the scintillation process in the  $\text{Ce}^{3+}$ -ions. It is expected to enhance the probability of a neutron-induced scintillation event by a factor of  $10^3$  in comparison to a gamma-induced event. Up to now the  $^6\text{Li}$ -glass has only been commercially available from Levy Hill Labs. Ltd. in England. Industrial interest is limited by the relatively small requirements for neutron sensitive scintillation material.

An important alternative to the  $^6\text{Li}$ -glass is the  $^6\text{LiI}$ -scintillator giving a light output of one order of magnitude higher and simultaneously reducing the gamma sensitivity almost completely. Here the technological difficulties are in preparing single crystals of sufficient size and in safely housing the hygroscopic material. We are going to install an  $\text{LiI}$ -crystal of  $120 \times 30 \times 2.5 \text{ mm}^3$  dimensions into a prototype detector in Jülich



in order to study its performance. Additional efforts are to be made with further materials like  $^6\text{LiF}$  or plastic scintillators.

Special attention has to be paid to the maximum count rate capability. A value of  $2 \cdot 10^6$  counts/s (Fig. 7) which should be managed by present-day fast electronics with the Anger technology may probably turn out to be insufficient for large area detectors at Laue-TOF experiments for single crystal structure analysis. Further developments for a more sophisticated processing method and read-out procedure of the detector events are necessary.

First attempts to develop a quite different technology for position-sensitive scintillation detectors have been started at our Institute, registering the scintillation event in an electro-optical semiconducting device (e.g. CCD) instead of a photomultiplier tube. Similar developments are in progress for X-ray area detectors envisaged for a count rate capability of  $10^9$  counts/s. The future prospects for highly efficient position-sensitive scintillation detectors at SNQ instruments appear rather encouraging.

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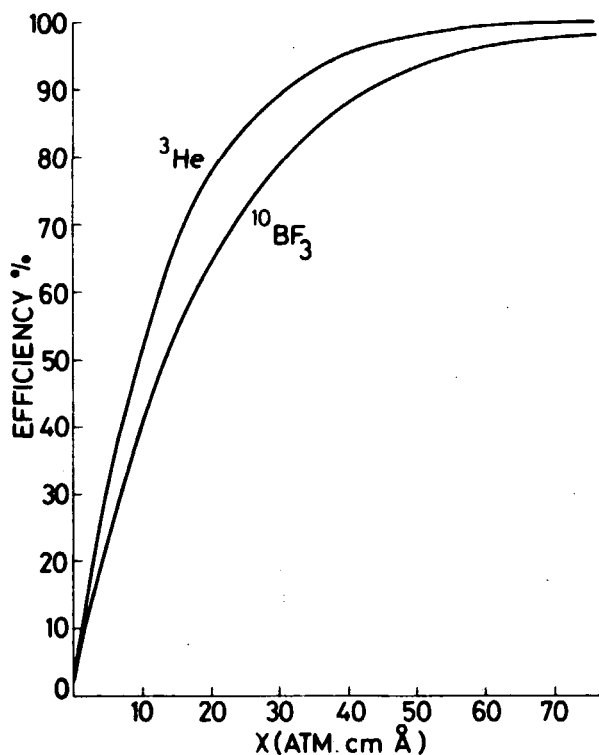


Fig. 1a: Neutron detection probability of  $^3\text{He}$  and  $^{10}\text{BF}_3$  gas as a function of the product of gas pressure, gas depth and neutron wavelength

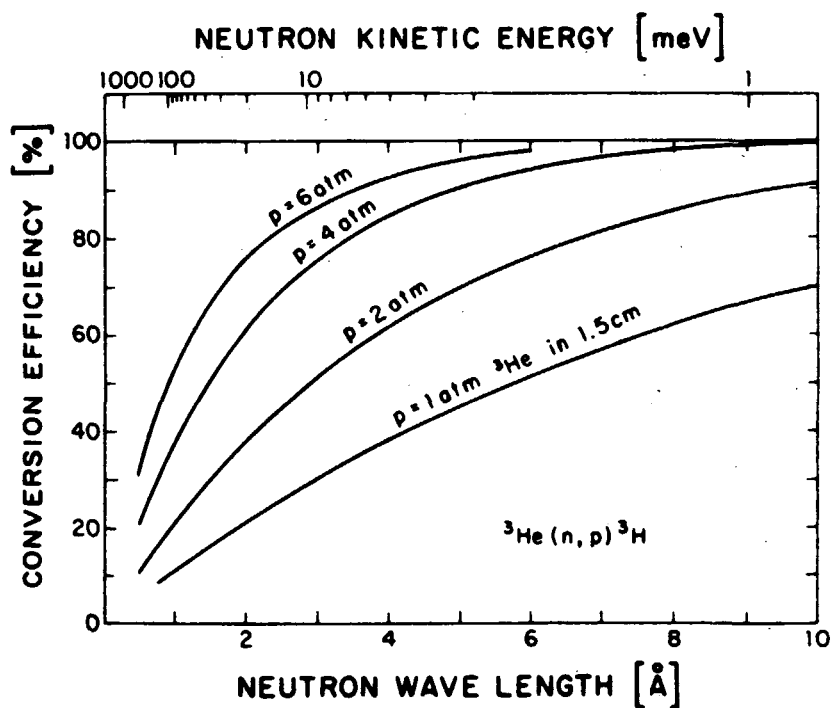


Fig. 1b: Neutron efficiency of  $^3\text{He}$  gas as a function of wavelength and gas pressure for a gas depth of 15 mm according to /4/

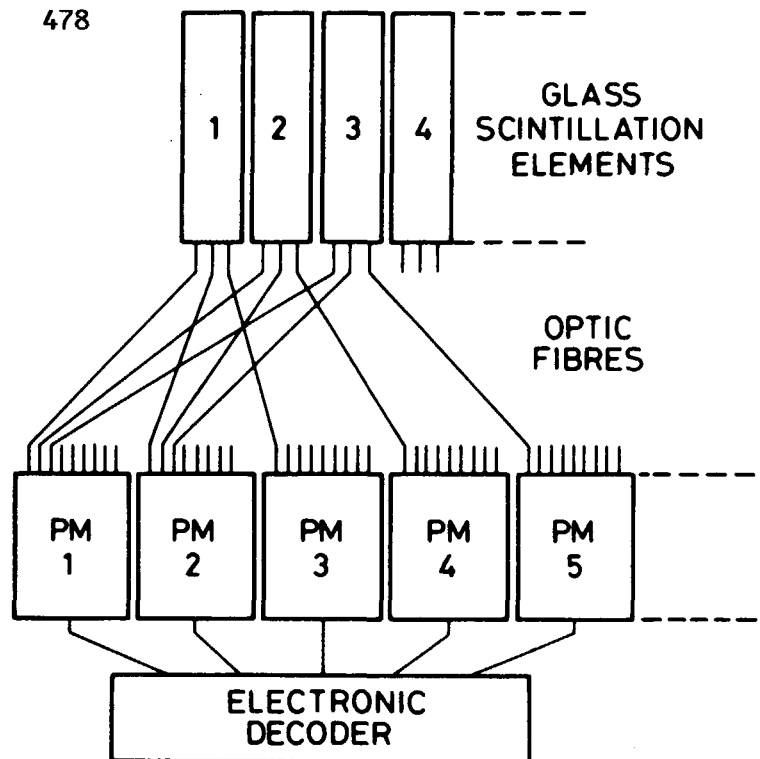
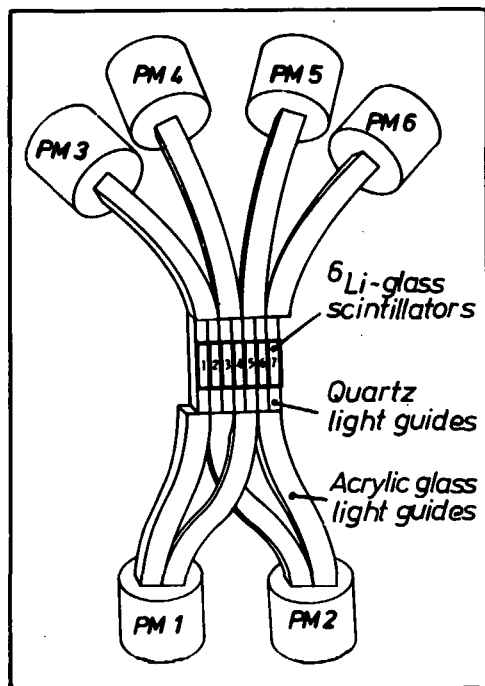


Fig. 2a: Position resolution using digital multi-element techniques with light guide coding

left: dual coding with plexiglass light guides  
(according to Kurz et al. /7/)

right: triple coding with glass-fibre light guides  
(according to Davidson and Wroe /1/)

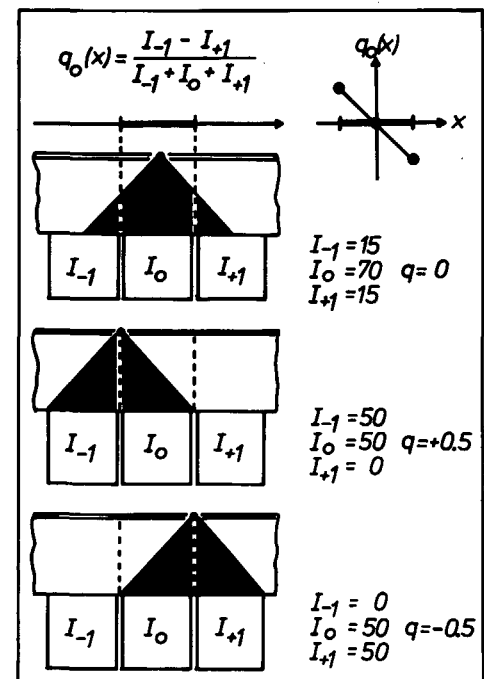
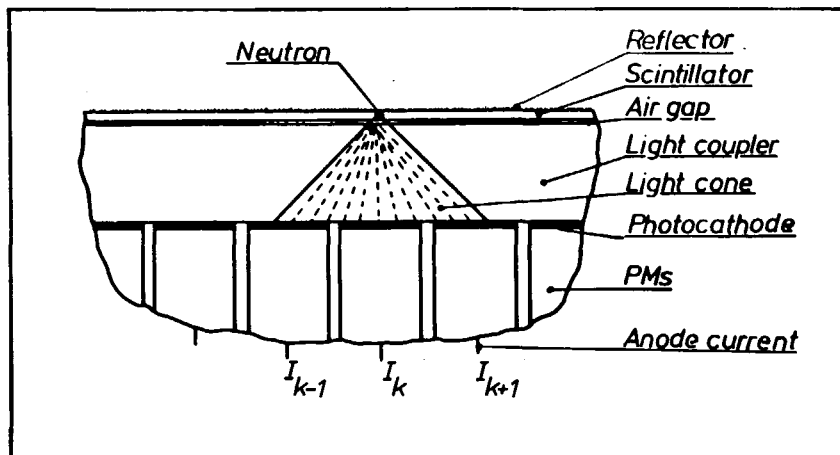


Fig. 2b: Position resolution of continuous design using the Anger technology:

left: schematic design principle (cf. text)  
right: simplified position-dependent signal function from the currents of three adjacent photomultipliers

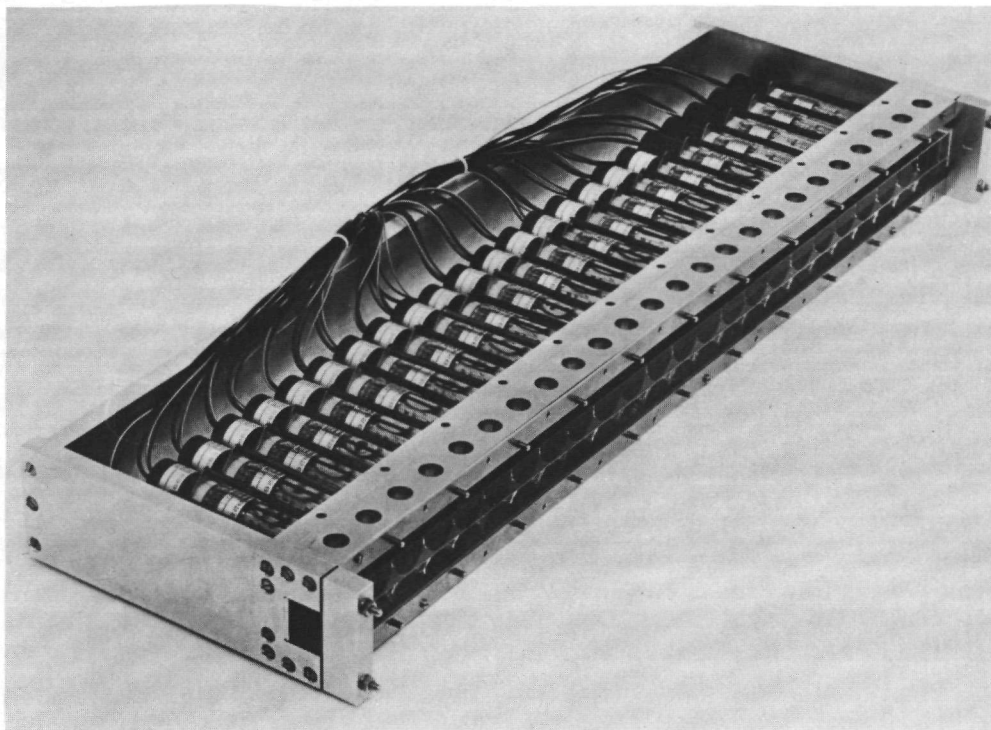


Fig. 3a: Linear position-sensitive scintillation detector  
(one unit of 682 mm sensitive length with 24  
photomultipliers)

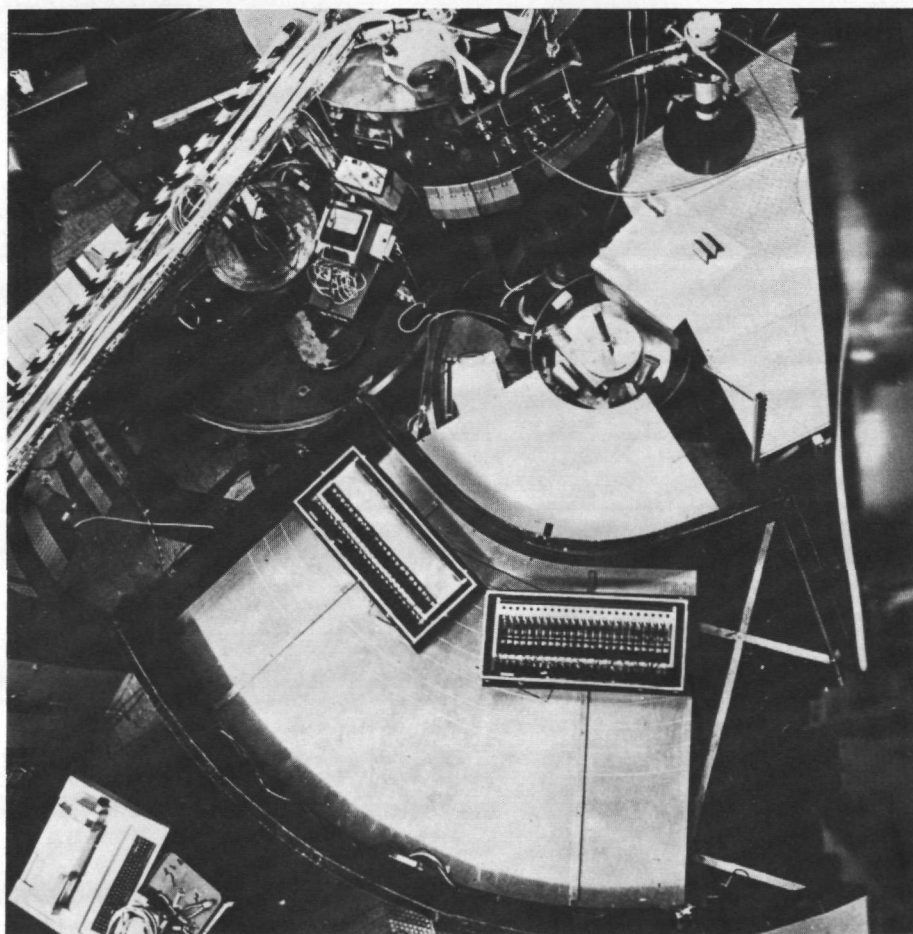


Fig. 3b: Installation of the scintillation detector  
(consisting of two identical units) at the  
two-circle diffractometer in Jülich

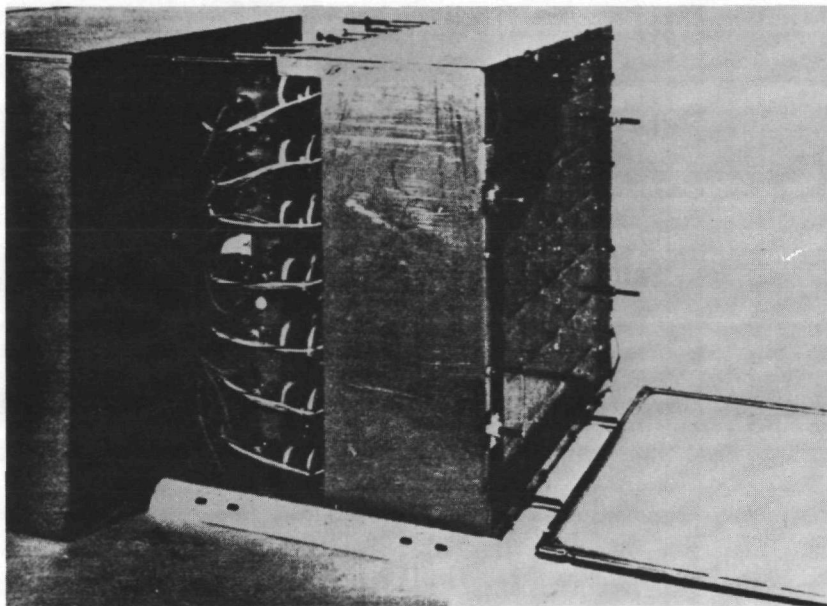


Fig. 4a: Two-dimensional position-sensitive scintillation detector ( $30 \times 30 \text{ cm}^2$  with  $7 \times 7$  square photomultipliers) according to Strauss et al. /15/, installed on a Laue single crystal diffractometer in Argonne

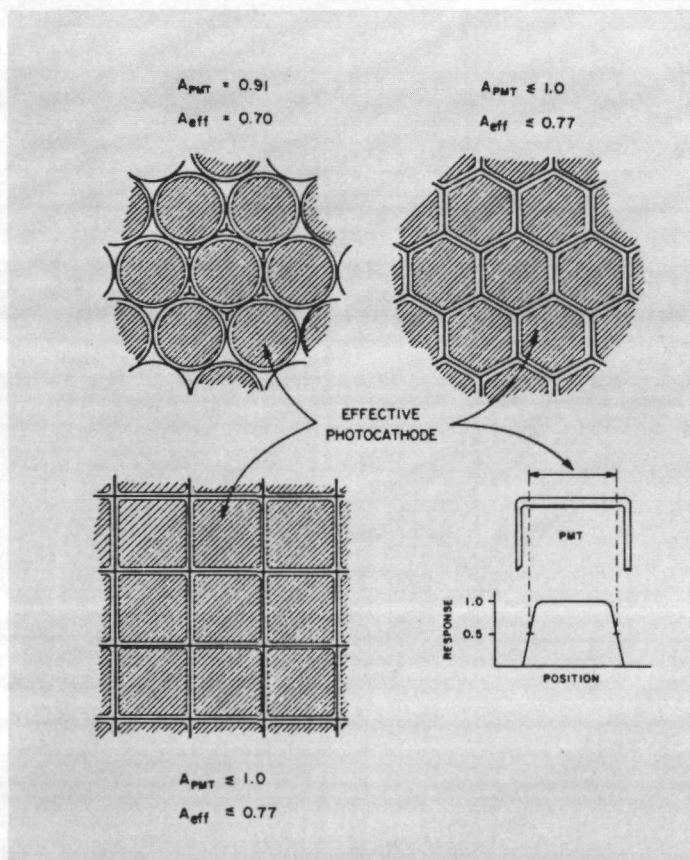


Fig. 4b: Area coverage of a scintillator surface with round, hexagonal and square photomultiplier cathodes /15/

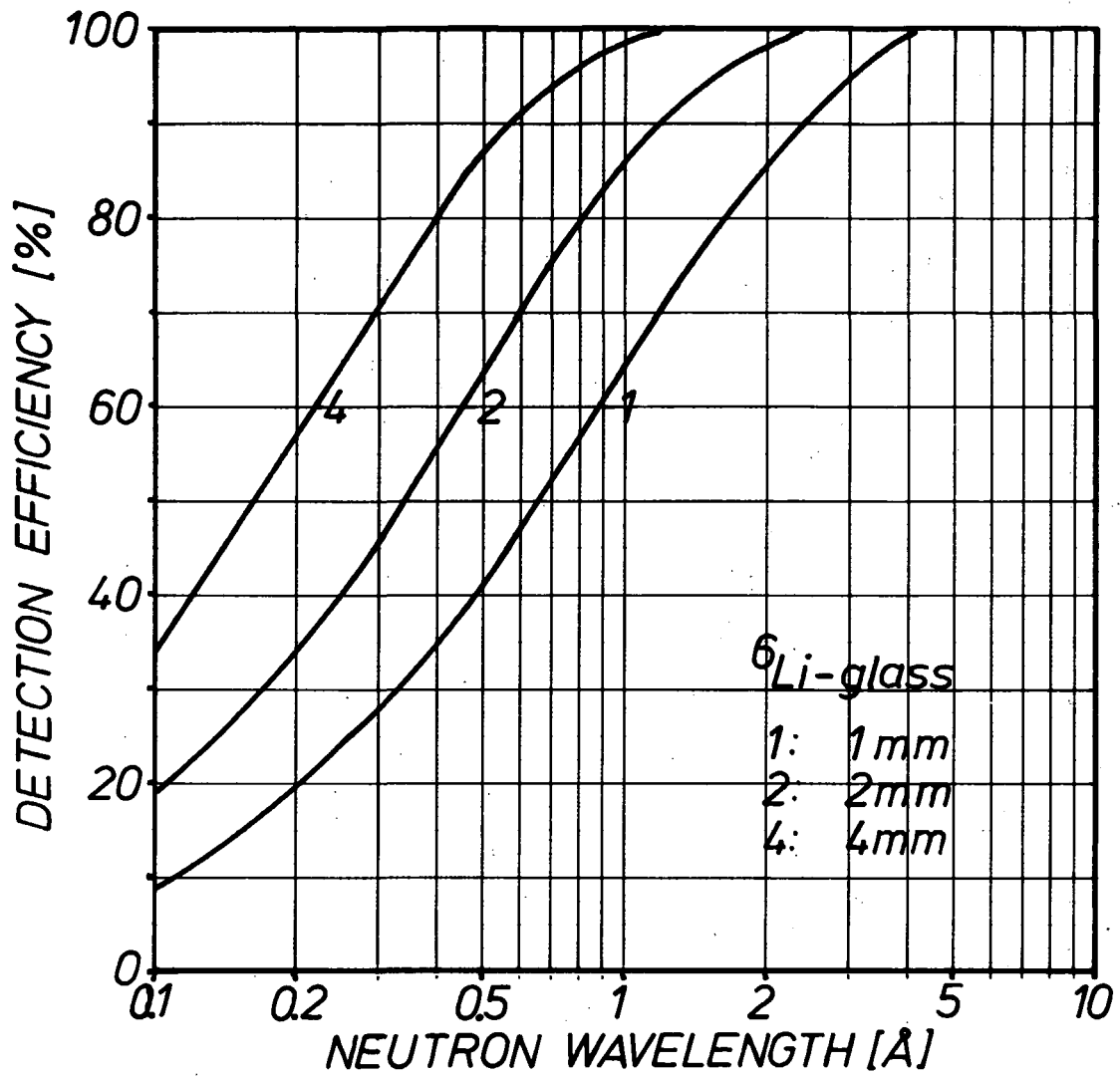


Fig. 5: Neutron detection probability of  ${}^6\text{Li}$  scintillator glass as a function of neutron wavelength and glass thickness



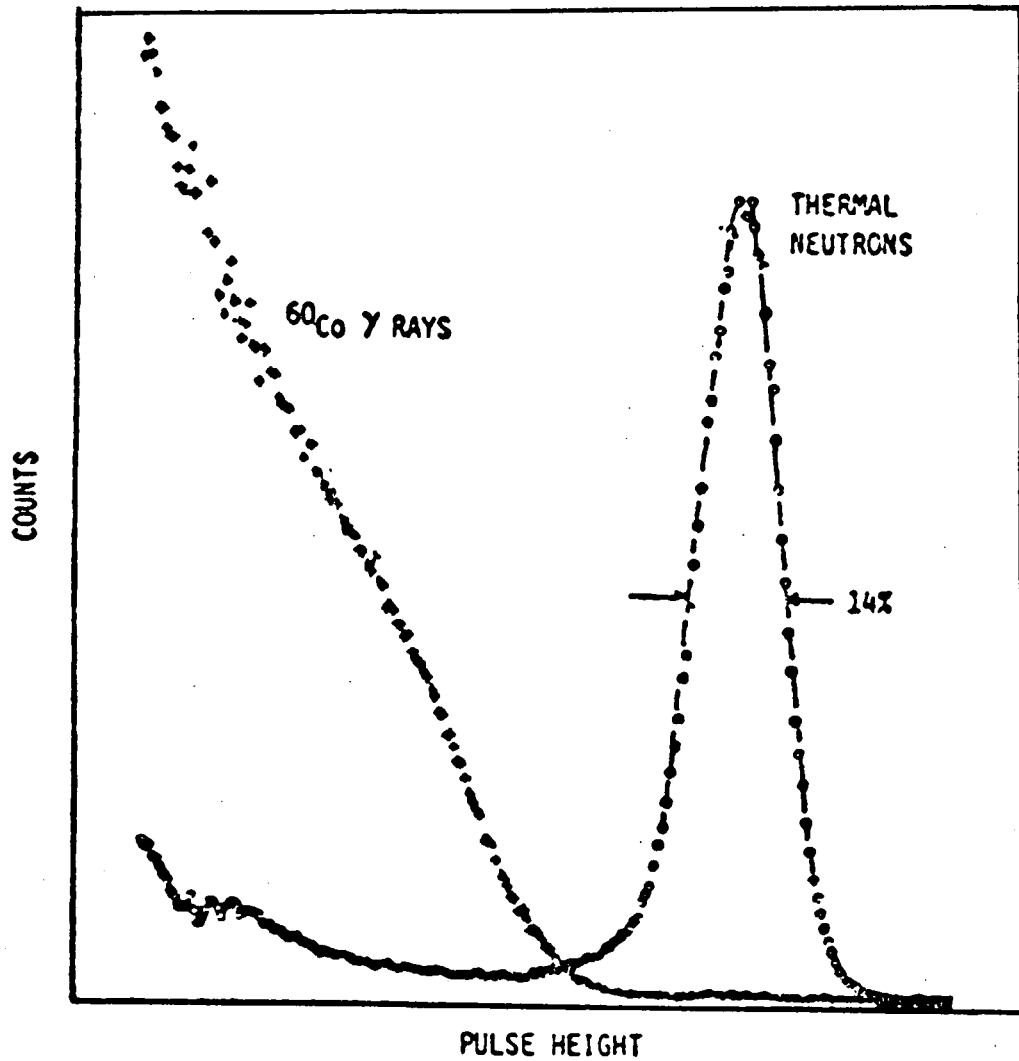


Fig. 6: Pulse height spectrum of  $^6\text{Li}$  scintillator glass with a minor overlap of 1.2 MeV gamma pulses in the thermal neutron peak /15/

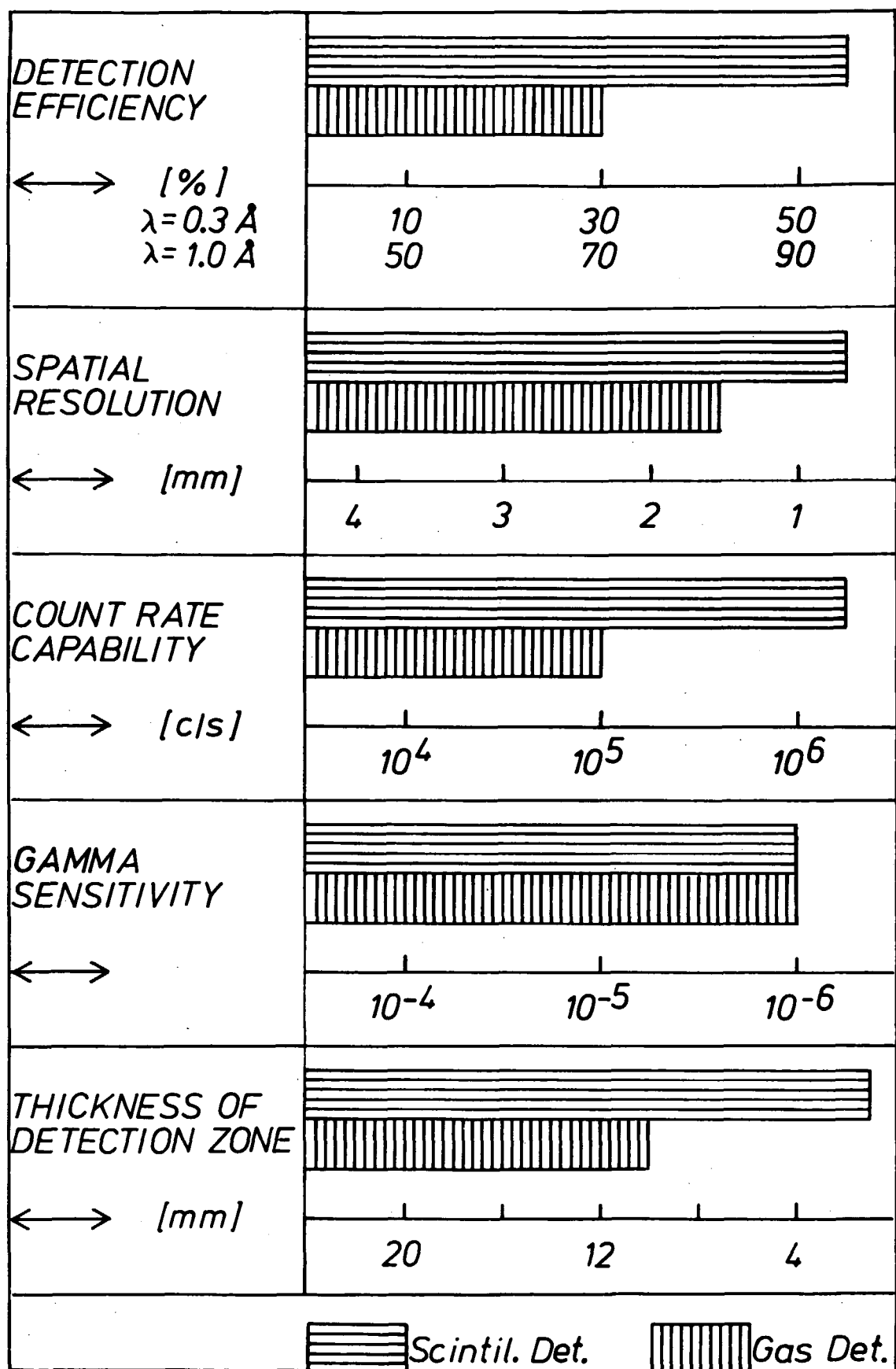


Fig. 7: Comparison of the performance spectra of gas and scintillation detectors

1. We can anticipate the background situation on SNQ by extrapolating from experience gained at the operating spallation sources. Background from the source shield itself, and cross-bulk between beamlines may be made negligible by an appropriate quantity of shielding. The dominant background source in any spectrometer remains that spectrometers own beam.
2. Fast neutrons only become a problem when they moderate down to thermal energies. Even then, if they are moderated quickly, the "power pulse" remains tight and occurs at times substantially earlier than the times of interest. However, since the parent neutrons that give rise to this pulse are extremely penetrating, it may prove difficult to suppress the power pulse on those spectrometers that intend to take data over a timescale longer than a single frame. Careful design of the collimation and get lost tube is essential.
3. A second beam induced background occurs if the time-energy relationship which we assume in a time-of-flight measurement is no longer valid. These "off-energy" neutrons result from scattering and thermalization in the collimator, in any "in-beam" device such as a chopper, and from delayed fission neutrons in the target. Experimental measurements at Los Alamos (on a non-fissile target) showed the contamination to be 1:2500. By careful design and choice of materials, this may be further reduced. In-beam devices should be such that they do not convert fast neutrons to thermal neutrons and the same careful design must be made of the subsequent beamline. The delayed neutron fraction for spallation on  $^{238}\text{U}$  has been measured at Argonne to be 0.65 %. On SNQ the rotating target reduces this by a factor 20 (?) and a further reduction results, in the case of chopped beams, from the duty factor.

4. A third source of beam born background is due to elastic scattering of the white beam by the sample. Again careful design of the local environment with use of neutron absorbing materials ( $B^{16}$ ,  $Li^6$ , not necessarily Cd!) is required.

# Ultracold Neutrons at SNQ

A. Steyerl

TU München

All known UCN applications require a continuous or quasi-continuous beam, where the term "quasi-continuous" means "pulses" of, say, 10 sec duration in intervals of 100 sec (for EDM experiments or other storage experiments). Therefore, a direct use of the time structure of SNQ in time-resolved experiments appears to be impracticable. However, certain methods of UCN production permit us to produce a beam intensity - or density - of UCN as determined by the peak intensity (rather than the mean flux) - at least in principle. In the following I shall discuss two techniques - one proposed and the other realized - aiming at utilization of the peak flux in a pulsed source. (A comparison between various methods of UCN production of SNQ has previously been given by Golub et al. /1/.)

The first method /2/ to be discussed has been proposed for the pulsed reactor at Dubna, but to my knowledge, it has not been put into practice. It is based on direct extraction of UCN from the primary source, using a converter. A fast shutter provided in front of the converter admits UCN to a "reservoir" bottle only during reactor pulses. Thus UCN can, in principle, be accumulated up to the phase space density in the peak. However, the problem of the shutter (magnetic?) has not been solved yet, and the necessary filling times would be forbiddingly long, with the consequence of significant neutron losses during filling.

The second technique, the "crystal turbine" constructed at Argonne /3/, seems to be more adequate to a pulsed source. Receding thermica crystals mounted on a rotating arm of 1.2 m in length were used as reflectors. In this arrangement neutrons of initial velocity  $v_1 = 400$  m/s are Doppler shifted to the UCN regime. An attractive feature of this method is as follows:

Liouville's theorem implies that on deceleration of an ensemble of neutrons by collectively acting forces from a velocity  $v_1$  to a velocity  $v_2$ ,

an enhancement factor  $v_1/v_2$  ( $\sim 100$  for the Argonne case) arises. This factor may appear in several ways, depending on the deceleration technique used:

- a) For inelastic spin flip /4/ (constant energy transfer by  $2\hbar\omega$ ) the longitudinal extension of the considered element in momentum space is broadened. (This seems unattractive for UCN production).
- b) In the "total-reflection turbine" (Garching) the beam cross section is enlarged. (This is quite useful since often wide UCN beams are desirable.)
- c) Finally, for the different geometry of the "crystal turbine" the factor  $v_1/v_2$  appears, in general, partly in the form of a temporal pulse dilatation. (Viewing the zigzag-shaped cloud of UCN left behind the receding crystal from a direction along the zigzag axis, the enhancement is purely temporal; if viewed from a lateral direction, a part of the enhancement factor will be spatial, in general.)

The aspect of pulse width dilatation makes the crystal turbine attractive for use at a pulsed source. For the duty cycle at SNQ (1 ms/10 ms = 0.1 for the Cold Source) we can easily ensure pulse dilatation up to the repetition period of 10 ms, thus obtaining a continuous UCN beam intensity corresponding to the peak flux. This requires a crystal length of  $\sim 5$  cm, and a width of, say, 10 cm. (The feeding guide cross section, however, would have to be larger, say 15 cm x 10 cm, since the crystal traverses the incident beam at an angle of  $\sim 15^\circ$  to the axis). The UCN beam cross section would then be about 10 cm x 20 cm.

We must add a few cautionary remarks in respect to efficiency. The overall conversion efficiency observed in /5/ (corrected for UCN guidance and detection efficiency) is 1.5 %, as referred to the peak flux. This is to be compared to the 10% efficiency which we expect for the vertical UCN source (with total-reflection turbine) to be installed at ILL in 1985. The main source of loss for the crystal turbine seems to be the lack of an adequate crystal. In order to properly fill the spherical UCN region in momentum space, a very large mosaic spread of  $\sim 10^\circ$ , and reflectivity  $\sim 1$  over this full angular range, are required. At Argonne, a stack of thermica crystals with aluminium wedges used as spacers was employed in order to artificially increase the mosaic spread. This results in significant reflection losses, and the UCN sphere was not filled adequately.

Thus, work on an improved version of a "crystal turbine" for SNQ would have to start from a study of an appropriate reflector crystal. If the reflector properties cannot be improved considerably, the ILL solution might be at least as good a choice although it would use only the mean flux, not the peak flux. Its neutron economy is higher in the sense that it uses all neutrons of suitable initial velocity (while the "crystal turbine" can use only about 10 %). Hence the primary neutron guide can be smaller (7 cm x 7 cm at ILL).

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Ultra-Cold Neutrons and Spallation Neutron Sources

R. Golub and K. Böning  
Physics Department  
Technische Universität München  
8046 Garching

Abstract

We present a brief introduction to the field of Ultra-Cold neutron (UCN) research including a review of the properties of UCN and some current and future applications.

We then discuss two types of UCN source suitable for use in connection with spallation neutron sources - "Doppler shifter" and "Superthermal" UCN sources.



## 1) Properties of UCN

Ultra-cold neutrons (UCN) are neutrons with energies so low (a few times  $10^{-7}$  eV) that they are totally reflected from many materials. While reflecting from a surface the UCN penetrate 100 or more Angstroms into the material. The loss probability per reflection depends on the material and the condition of the surface and ranges from  $10^{-3}$  to  $10^{-5}$  for selected materials. These properties mean that UCN can be stored in bottles made of selected materials for periods of time exceeding 100 seconds and perhaps approaching 1,000 seconds (the neutron  $\beta$  decay lifetime) and open up a wide range of interesting applications.

## 2) Applications of UCN

### a. The search for a neutron electric dipole moment (edm)

At the present time this is perhaps the UCN application which is being most energetically pursued. Groups at the ILL, Argonne National Laboratory, Leningrad and Dubna have all put many man-years of effort into this experiment. The search for the neutron electric dipole moment is interesting because the existence of an edm would be direct evidence for time reversal violation outside the  $K^0$ -meson system. The present best experimental limit corresponding to an edm  $\sim 4 \times 10^{-25}$  e-cm is the result of a UCN experiment at Leningrad.<sup>(1)</sup>

It is expected that the observation of a neutron edm will make a clear distinction between the superweak theory on the one hand, and the other theories (e.g. spontaneous T violation, unified theories of the weak and electromagnetic interactions) which have been proposed to account for the T violation.

The UCN densities expected from the sources discussed below are so large compared to currently available densities that the UCN density should no longer be a limiting factor on the experimental sensitivity which will undoubtedly be limited by magnetic field fluctuations. With such high UCN densities it will certainly be advantageous to push the UCN technique to its limit using superconducting magnetic shields and other aspects of low temperature technology. However the presence of large iron radiation

shields at spallation sources may cause some perturbations to the magnetic environment.

#### b. Neutron $\beta$ decay

At present the neutron lifetime is only known to an accuracy of about 1 %. A more accurate determination of the neutron lifetime would improve our knowledge of the weak interaction constants appearing in the theory of  $\beta$  decay, which are currently determined by a fit to a large number of measurements of  $\beta$  decays of nuclei and elementary particles. There would also be an improvement in our knowledge of the radiative corrections to the  $\beta$  decays. The knowledge of several of these theoretical parameters is currently limited by the 1 % accuracy of the neutron  $\beta$  decay lifetime. <sup>(2,3)</sup>

Classical methods of measuring the neutron lifetime involve separate detectors a) to measure the neutron density in the apparatus and b) either electron or proton detectors to measure the decay rate. Thus the measurement requires knowledge of the absolute efficiencies of a neutron and a charged particle detector. By using stored UCN the measurement can be carried out with a single detector whose absolute efficiency is not required.

This experiment has been actively pursued for several years by the group at Bonn <sup>(4,5)</sup> who are hoping to measure the lifetime of UCN stored in a magnetic bottle. The UCN are produced by downscattering in liquid Helium similar to the UCN source discussed below.

UCN can also be used to study asymmetries in the decay of polarized neutrons and may prove useful for studying parity violation in  $(n, \gamma)$  reactions.

#### c. Material Studies with UCN

UCN have the unique property that they penetrate surfaces to depths of a few hundred Angstroms. The depth of penetration is a function of neutron energy as well as the density and composition of the material surface. The loss rate of UCN on a surface is also a function of these parameters. Thus

the loss rate of a surface will be altered by the presence of a layer of oxide or film of other material on the surface and UCN will provide a unique way of studying such films.

UCN can be used for inelastic scattering studies of materials in several ways. For energy transfers large compared to the UCN energy, UCN can be upscattered from the surface region (or from the bulk of those samples which are penetrated by UCN) and the energy spectrum of the upscattered neutrons can be measured. Although this will have the disadvantage that only the region of the  $\omega$ - $Q$  plane that lies along the  $\omega \propto Q^2$  curve of the free neutron is accessible, the technique may allow measurements which are impossible by other methods. For example UCN stored in liquid Helium will be upscattered as a result of interactions with the phonons and rotons at a rate which is temperature dependent. A measurement of the spectrum of the upscattered neutrons would provide information on  $S(Q, \omega)$  in a region far from the phonon resonance at unprecedented levels of sensitivity - mean free paths of kilometers for 5 m/sec UCN. Such experiments seem to be impossible by any other technique and have become more interesting in the light of recent experimental and theoretical results.<sup>(6)</sup> In addition one can study excitations in thin Helium films by a similar technique.

There is also the possibility of accelerating the UCN gas to  $\lambda \sim 100-200 \text{ \AA}$  in order to do scattering experiments. One method to accomplish this is to gradually accelerate a container filled with UCN. When the container is suddenly stopped the neutrons will continue (through the walls of the container) with an average velocity given by the velocity of the container before deceleration. The advantage of such a scheme derives from the very high phase space density available in the UCN source.

Somewhat related to this is the possibility of accelerating the UCN stored in liquid Helium by means of a strong monochromatic phonon beam or perhaps a shock wave in the Helium. In addition to the possibility of producing an intense burst of approximately monoenergetic neutrons the latter approach may provide an interesting method for studying shock waves in the superfluid Helium.

Another method of using UCN to study condensed matter is by means of a UCN spectrometer such as the gravity spectrometer constructed by Steyerl<sup>(7)</sup> (fig. 1b).

This offers the possibility of very high energy resolution ( $\sim 10$  neV) in the Q range of  $10^{-3} - 3 \times 10^{-2} \text{ \AA}^{-1}$ . Such a spectrometer could be used to study slow dynamics in biological systems, hyperfine splittings, magneto-crystalline anisotropies, low lying rotational states and hindered rotations in very large molecules and the slow critical fluctuations close to a phase transition, in particular magnetic phase transitions.

Elastic scattering of UCN can be used to study inhomogeneities in materials and transmission of UCN to study thin films.

#### d. UCN optics

Steyerl and his co-workers have already demonstrated the operation of an achromatic Fresnel lens for UCN.<sup>(8)</sup> Such work can be expected to lead to the development of a UCN microscope with a resolution of perhaps  $1 \mu$ . A prototype with a resolution of a few hundred microns is currently under construction (fig. 2). The advantage of such an instrument would be based on the different scattering properties of materials for neutrons as compared to other radiations, e.g. a UCN microscope is expected to be useful for studying the distribution of Hydrogen in a given sample.

Recent experiments at Garching<sup>(9)</sup> have studied quasi-bound states in one-dimensional potentials produced by multi-layer structures. One could extend this work by constructing a UCN Fabry-Perot interferometer and studying transitions between different bound levels induced by an oscillating magnetic field as proposed by Steyerl. This would demonstrate the operation of the time dependent Schroedinger equation in an almost macroscopic system ( $\sim 10^3 \text{ \AA}$ ).

### 3) New Generation UCN Sources

While existing UCN sources provide densities of at most  $1 \text{ UCN/cm}^3$ , new neutrons sources (e.g. spallation neutron sources) now being planned or constructed open up several possibilities for producing UCN densities on the order of  $10^3 \text{ UCN/cm}^3$  or perhaps even significantly greater.

One type of "new generation" UCN source ("Doppler shifter" UCN source) is based on the slowing down of faster neutrons as a result of reflection from a reflector which is moving in the direction of the neutrons. Such a device would be placed at the end of a guide tube looking at a "cold" Hydrogen or Deuterium source and would produce a UCN density which is limited by Liouville's theorem to correspond to the phase space density in the peak of the pulse in the cold source since the UCN are admitted to a storage vessel by means of a shutter which opens in phase with the neutron pulse.

Two methods of realizing such a UCN source have been proposed. One makes use of the total reflection phenomena from a moving curved surface and produces UCN by slowing down 50 m/sec neutrons (Steyerl turbine). The second uses Bragg reflection from a moving crystal and requires 400 m/sec neutrons as an input; such a device has been constructed at Argonne. Both devices are expected to provide similar UCN densities which are estimated to be about  $3 \times 10^3$  UCN/cm<sup>3</sup> at SNQ,<sup>(10)</sup> produced outside the radiation shield at a position suitable for experiments.

The second general type of UCN source ("superthermal source") is based on the slowing down of 400 m/sec neutrons as a result of the emission of phonons by the neutrons (inelastic down-scattering) in superfluid liquid He<sup>4</sup>. The helium is contained in a vessel suitable for storing UCN so that the UCN, once produced, remain in the source until they are removed as the result of interactions with the Helium or the walls. The steady state UCN density in the source then depends on the production rate and the loss rate. After a time on the order of the UCN storage time the UCN density approaches its steady state value and this high density UCN "gas" can be extracted by means of a valve which would be opened to release the UCN. This device is not limited by Liouville's theorem which applies to the complete system - neutrons and phonons - and not to the neutrons alone.

Such a source, installed 70 cm from the bottom of the SNQ D<sub>2</sub>O tank is expected to produce densities  $\sim 4 \times 10^5$  UCN/cm<sup>3</sup> in the source. In order to be useful, for most experiments the UCN will have to be transported from the source to an apparatus outside the spallation source radiation shield, a process which will introduce some losses.

Steady state currents can be extracted from the UCN storage bottles in both of the sources under consideration (the Helium source and the Doppler shifter source) by fitting the bottle with an outlet port. In both cases the currents available would be proportional to the saturation densities in the sources so the comparisons we give here are also applicable to the case of scattering experiments.

The gain in available density expected from these "new generation" sources in comparison with existing sources is so great that one can expect that an entirely new range of research applications with UCN will be opened.

#### 4) Doppler Shifter UCN Source (12)

A sketch of the apparatus as realized at the Argonne Spallation neutron source is shown in fig. 3. Neutrons (400 m/sec) are transported from a liquid Hydrogen moderator through a 5 m long vertical guide tube. After leaving the guide the neutrons are reflected from a moving crystal which leaves them with a very small velocity after the reflection. The result is a cloud of UCN some of which move into the UCN guide tube which is kept closed by a slotted rotating disc except during the time when the neutron pulse is approaching the UCN guide.

This synchronous opening and closing of the guide allows the UCN density in the guide to build up (in principle) to that corresponding to the phase space density in the peak of the pulse.

Since the phase space density for a Maxwellian spectrum corresponding to total flux  $\phi_0$  is given by:

$$n(v_0) = \phi_0 e^{-(v_0/v_T)^2} (2\pi \cdot 100 v_T)^{-1} n \cdot cm^{-3} (m/s)^{-3} \quad (1)$$

$v_T$  = thermal velocity - m/sec

$v_0$  = velocity of incident neutrons

the maximum UCN density available with this technique is:

$$\rho_{ucn} = \hat{n}(v_0) \left[ \frac{4}{3} \pi v_0^3 \right] \quad (2)$$

where  $v_c$  is the maximum UCN velocity that can be stored and  $\hat{n}(v_0)$  is the phase space density corresponding to the peak of the neutron pulse, because the reflection from the moving crystal ideally shifts the velocity without changing the phase space density.

However because of inefficiencies due to crystal reflectivity, pulse broadening, etc. the UCN density produced is expected to be only 10 % of (2).

In addition the Bragg scattering process distorts the sphere in velocity space so that only a disc around the equator of the sphere is populated. This reduces the expected UCN density by a further factor of 6 as confirmed by experiment and Monte-Carlo calculations, but perhaps some of this can be recovered by a suitable angular collimator at the input of the UCN guide.

#### 5) The "superthermal" UCN source using superfluid He<sup>4</sup>

##### a. Production of UCN by downscattering in liquid Helium

If a volume of Helium is exposed to a flux of neutrons there will be a production of UCN given by (6)

$$P_{UCN} = \frac{d\phi(E^*)}{dE} E_{lim} \frac{4}{3} \sqrt{E_{lim}/E^*} \cdot S(k^*) N \sigma_0 \quad \text{UCN/sec/cm}^3 \quad (3)$$

where  $E_{lim}$  is the limiting energy for UCN storage

$$E_{lim} = 2,5 \times 10^{-7} \text{ eV} \\ = 2,9 \times 10^{-3} \text{ K} \quad \text{for Beryllium}$$

$N$  is the density of Helium atoms in the liquid =  $2,18 \times 10^{22}/\text{cm}^3$

$\sigma_0$  is the free atom coherent scattering length = 1.1. barns

$E^*$  is the critical energy for UCN production by phonon emission

$$E^* = \hbar \omega_f(k^*) = \hbar^2 k^{*2} / 2m \quad (4)$$

$$E^* = 12 \text{ K}$$

where  $\omega_0(k)$  is the phonon dispersion curve for liquid Helium

$S(k^*) = 0.1$  is the one phonon structure factor and

$d\phi/dE(E^*)$  is the energy spectrum of the incident neutrons evaluated at  $E^*$

Putting in the numbers we find from (3)

$$P_{UCN} = \frac{d\phi(E^*)}{dE} \cdot 1,44 \times 10^{-7} \text{ UCN/sec/cm}^3 \quad (5)$$

For a Maxwell Spectrum at temperature T

$$\frac{d\phi}{dE} = \phi_{th} \frac{E}{T^2} e^{-E/T} \quad (6)$$

where  $\phi_{th}$  is the total thermal flux. Thus

$$P_{UCN} = 1,92 \times 10^{-11} \phi_{th} \text{ for } T = 300^\circ K \quad (7)$$

#### b. Saturation steady state UCN density in the Source

Given the production rate  $P_{UCN}$ , the saturation density of UCN is given by

$$\rho_{UCN} = P_{UCN} \tau \quad (8)$$

where  $\tau$  is the storage time of UCN in the Helium containing vessel

$$\frac{1}{\tau} = \frac{1}{\tau_{He}} + \frac{1}{\tau_{abs}} + \frac{1}{\tau_{wall}} + \frac{1}{\tau_B} \quad (9)$$

$\tau_{He}$ , the relaxation time for upscattering of UCN in the Helium.

$\tau_{abs}$  is the absorption time ( $\rightarrow \infty$ ) in the  $He^4$

$\tau_{wall}$  is the relaxation time for wall losses

and  $\tau_B = 10^3$  sec is the B decay lifetime.

According to calculations  $\tau_{He} = 4000$  sec at 0.6 K  
and = 500 sec at 0.8 K

#### 6) Comparison of UCN Sources

From equations (1) and (2) we can write the UCN density from the Doppler shifter source as

$$\rho_{Dop} = \frac{\bar{\phi}_0(E_0)}{\eta} \cdot \frac{(\Delta E)}{v_0} \left( \frac{\Delta \mathcal{L}_{Dop}}{4\pi} \right) \quad (10)$$



where  $\eta$  is the duty cycle of the neutron pulses ( $\bar{\phi}/\eta$  is the peak flux  $\hat{\phi}$ )

$$\Delta E = (2/3) m v_0 v_c \quad (11)$$

$$\text{and } (\Delta \Omega_{\text{Dop}} / 4\pi) = (1/4) (v_c / v_0)^2 = 7.6 \times 10^{-5} \quad (12)$$

for  $v_c = 7 \text{ m/sec}$ ,  $v_0 = 400 \text{ m/sec}$ .

From equations (3) and (8) we can write

$$\mathcal{S}_{\text{He}} = \sum_0 (\Delta E) \bar{\phi}_0(E_0) \tau (\Delta \Omega_{\text{He}}) / 4 \quad (13)$$

with  $\Delta E$  defined by (11) and

$$\sum_0 = N \sigma_b S(k^*) (h_c/h^*)^2 = 4 \times 10^{-7} \text{ cm}^{-1} \quad (14)$$

Thus if both sources were placed at the end of the same guide tube the ratio of UCN densities produced would be

$$\mathcal{S}_{\text{He}} / \mathcal{S}_{\text{Dop}} = \sum_0 v_0 \tau \eta (\Delta \Omega_{\text{He}} / \Delta \Omega_{\text{Dop}}) \quad (15)$$

In the case when the solid angle for the Helium source is limited by the guide  $\Delta \Omega_{\text{He}} = \Delta \Omega_{\text{Dop}}$

$$\text{and } \mathcal{S}_{\text{He}} / \mathcal{S}_{\text{Dop}} = \sum_0 v_0 \tau \eta = 0.8 \times 10^{-2} \tau \eta = 3.2 \eta \quad (16)$$

(if we take  $\tau = 400 \text{ sec}$ )

for the case when the Doppler shifter source gives the maximum output allowable by Liouville's theorem.

However we have seen above that the density produced by the Doppler shifter source must be reduced by an unavoidable factor of 10 so that (16) becomes

$$\mathcal{S}_{\text{He}} / \mathcal{S}_{\text{Dop}} = 32 \eta \quad (17)$$

Now at Argonne  $\eta \sim 1/165$  so the Doppler shifter source would appear to be slightly better while at SNQ  $\eta \sim 1/12$ .

However the real advantage of the Helium source (at a spallation neutron source) becomes apparent when we consider that while  $(\Delta \Omega)_{\text{Dop}}$  is limited

by the mechanism of total reflection to be given by (12),  $(\Delta \mathcal{N})_{\text{He}}$  is only determined by the geometric arrangement of the UCN source relative to the primary neutron source.

For a practical case on the SNQ (fig. 4) we have  $(\Delta \mathcal{N})_{\text{He}} / 4\pi = 0.27$   
 or  $(\Delta \mathcal{N})_{\text{He}} / (\Delta \mathcal{N})_{\text{Dop}} = 350$   
 in a position where there is an acceptable heat input to the Helium container. (10)

Because the cold source supplying the guide tube for the Doppler shifter source can be in a higher flux, and can be larger and hence more efficient than the cold source supplying the "Superthermal" Helium source we have  $\eta_{\text{eff}} = 10^{-2}$  for SNQ so (8) yields  $\mathcal{J}_{\text{UCN}} (\text{He source, SNQ}) = 4 \times 10^5 \text{UCN/cm}^3$  and (17) becomes

$$\mathcal{J}_{\text{He}} / \mathcal{J}_{\text{Dop}} = 32 \times 10^{-2} \times 350 = 110$$

This figure applies only to the UCN produced inside the Helium, so it must be reduced by the losses associated with extraction of the UCN from the Helium to a position where they can be used for experiments. It seems reasonable to suppose that these losses can be kept below a factor of ten so that the "Superthermal" Helium source can offer significantly increased UCN densities if the engineering problems associated with its construction can be overcome.

In addition it might prove possible to do some experiments directly inside the liquid Helium source. (11)

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Fig.1 a)  $\omega, Q$  space showing region accessible to UCN scattering  
 b) UCN inelastic scattering spectrometer (7)

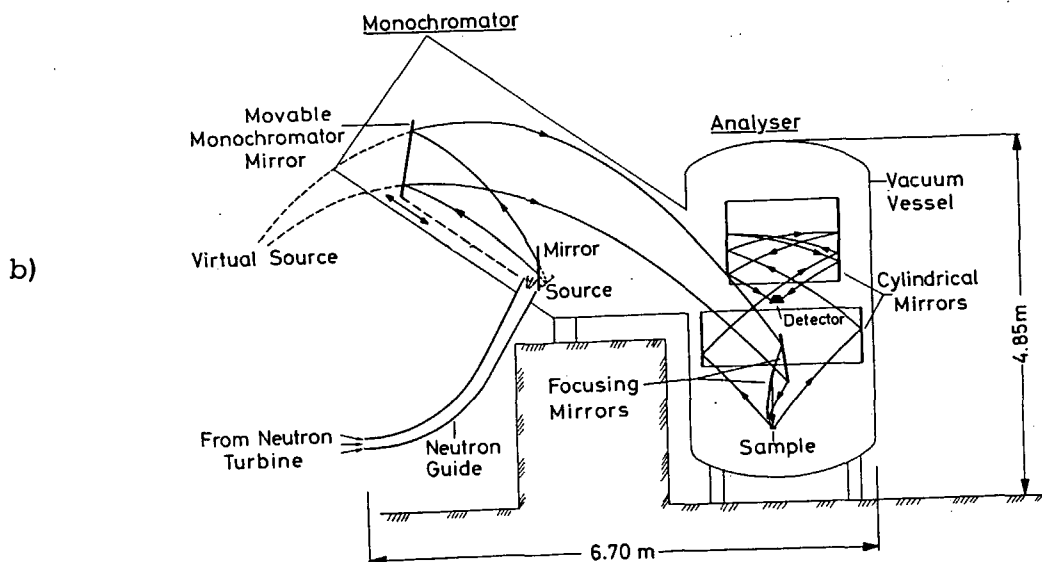
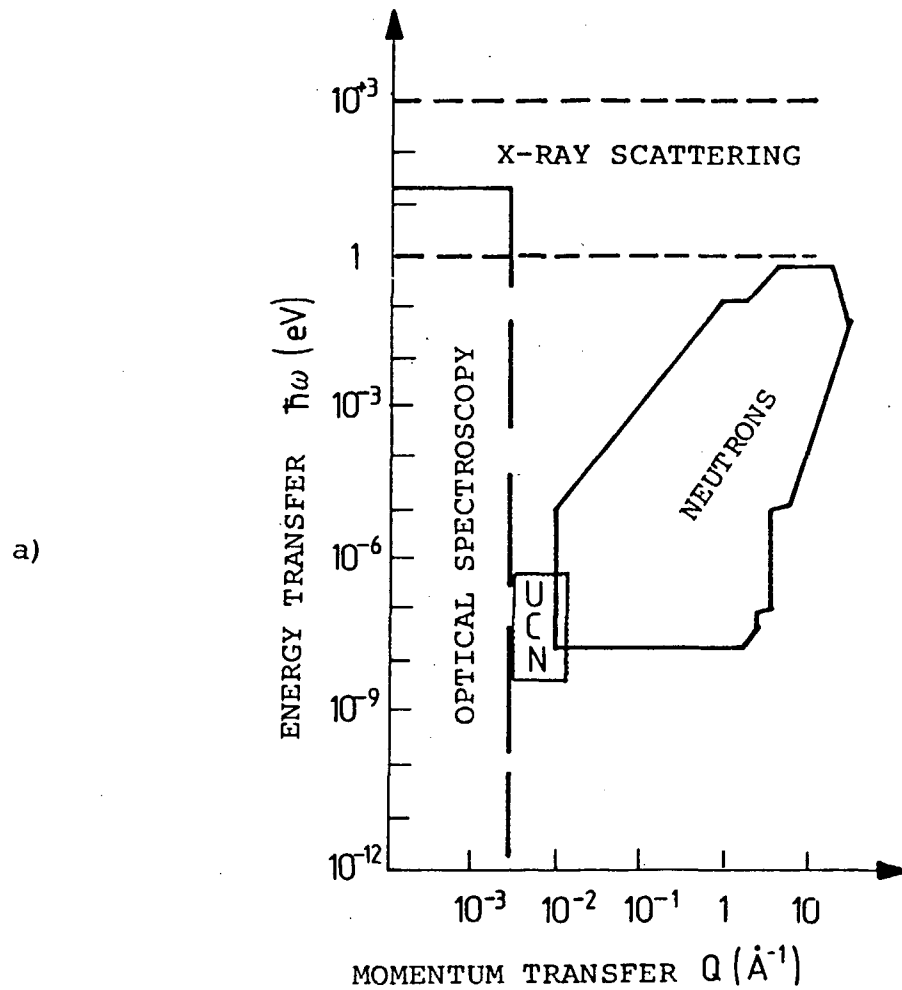
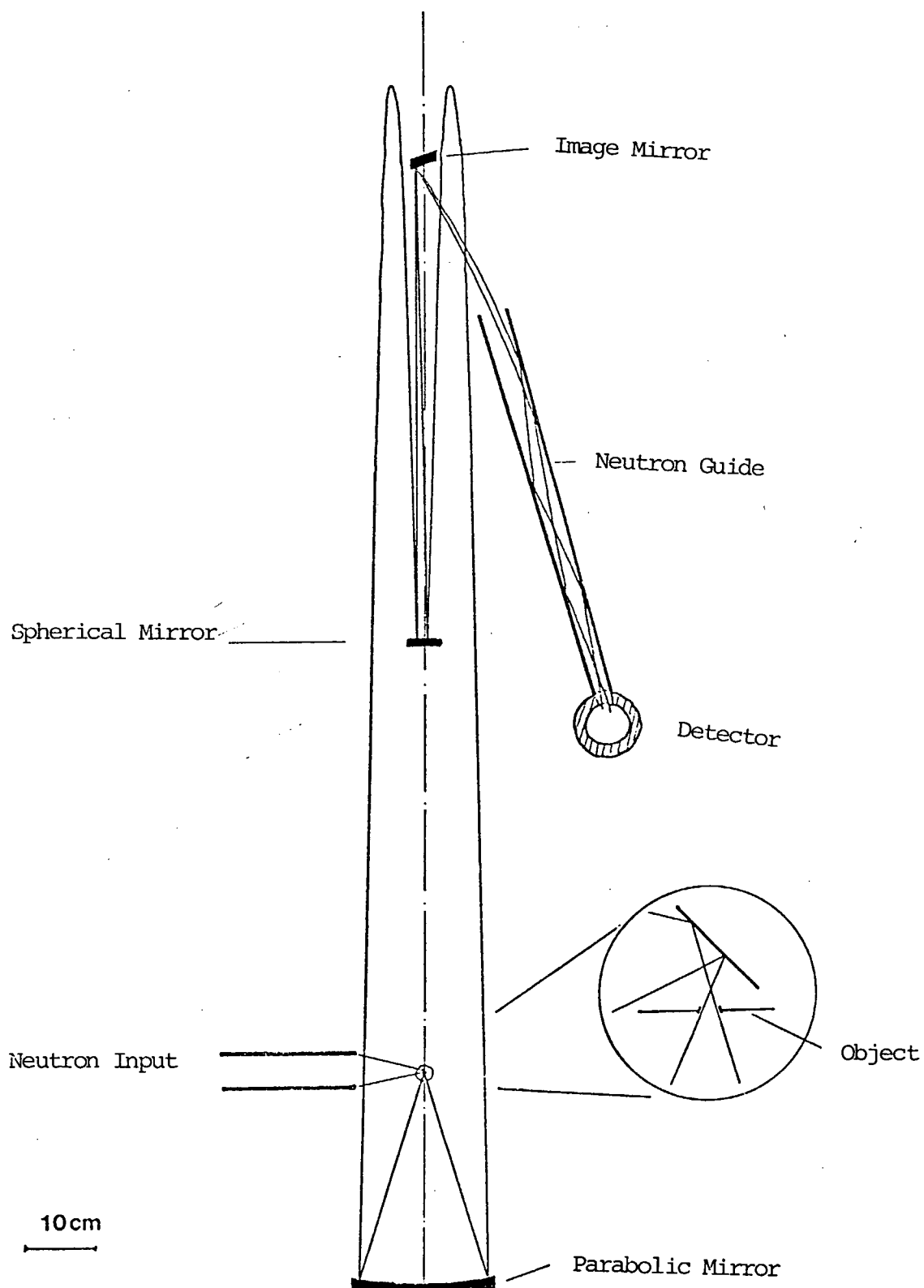


Fig. 2 Neutron microscope using UCN <sup>(13)</sup>



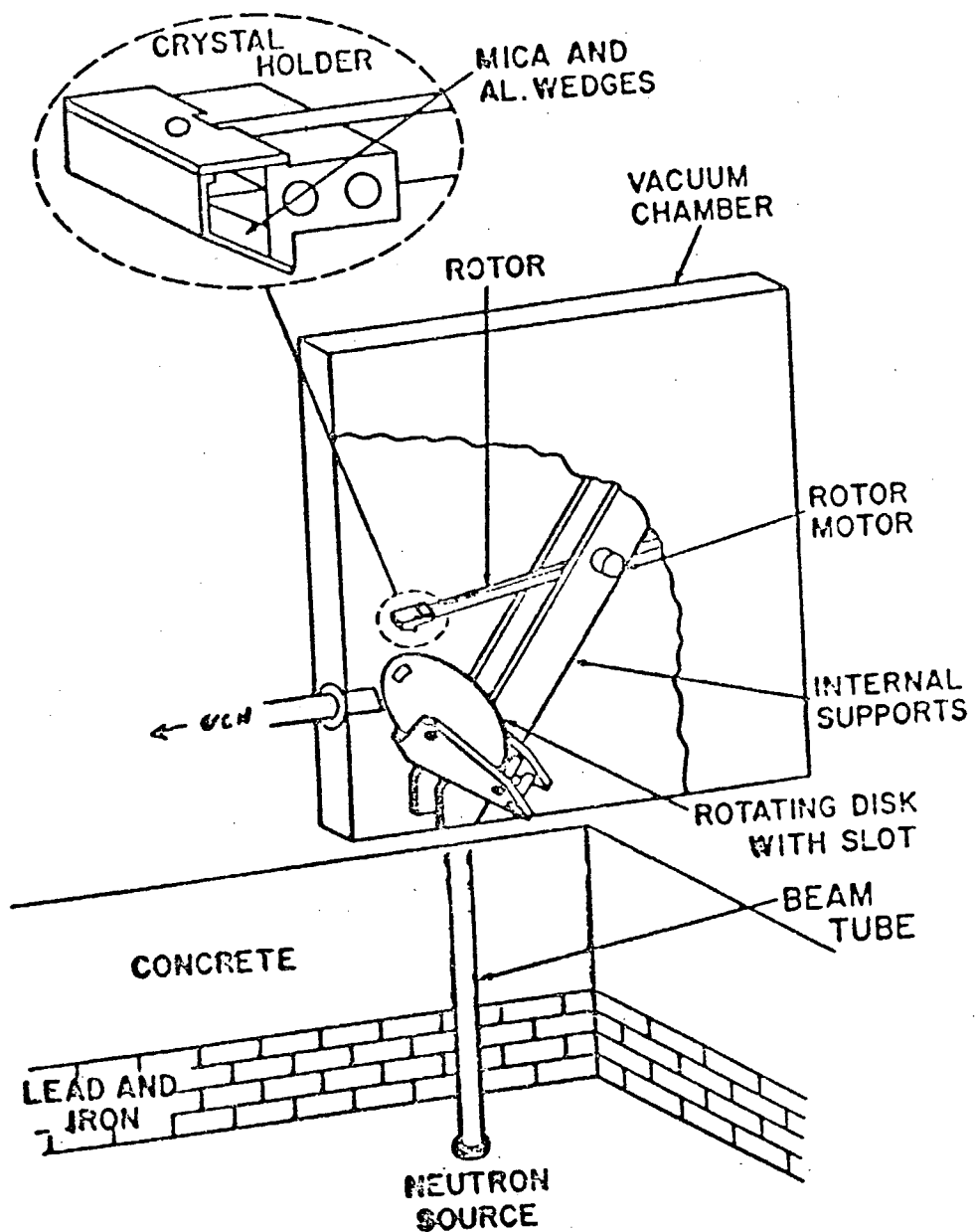
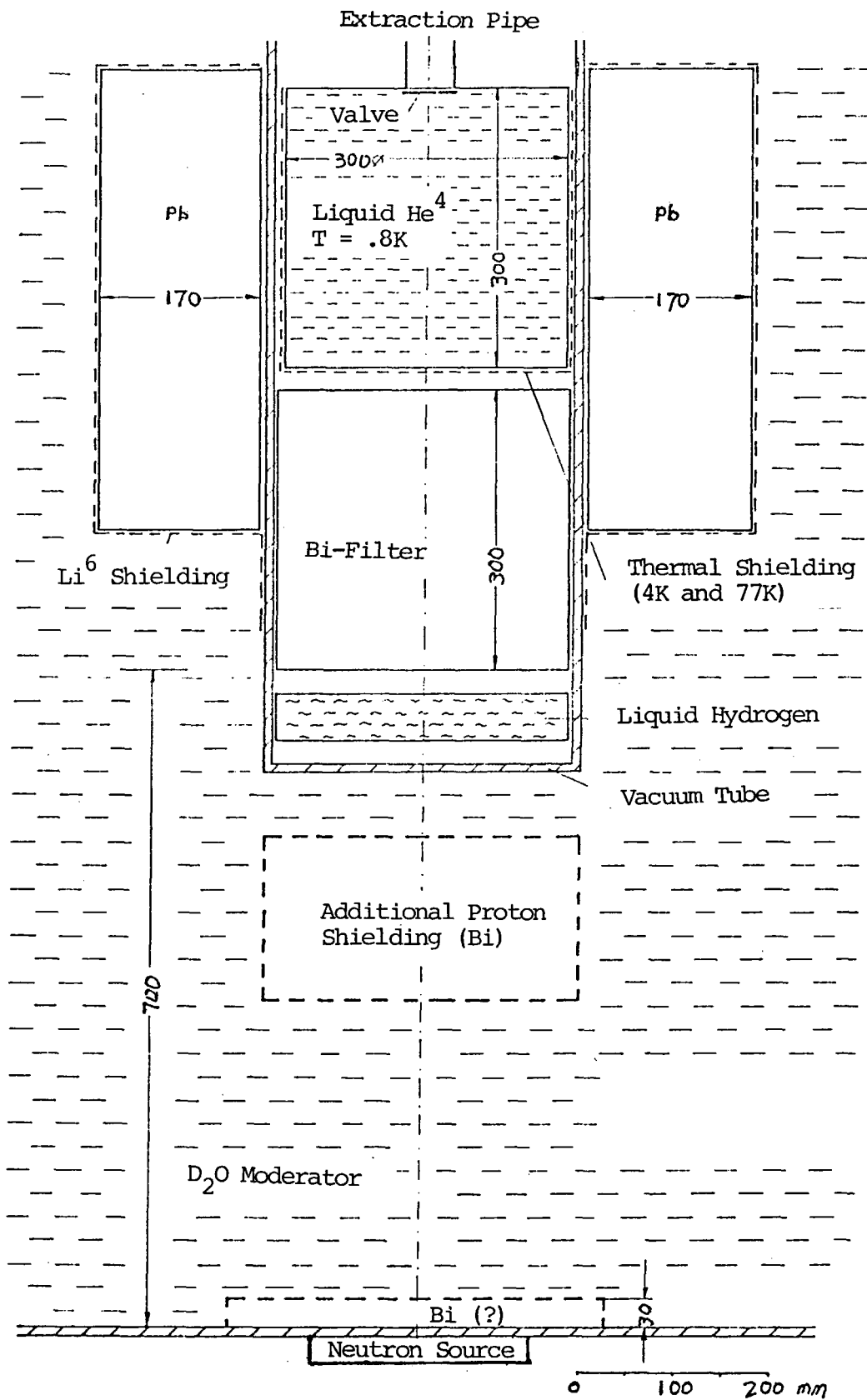


Fig. 3 Doppler shifter UCN source using rotating crystal as realized at Argonne<sup>(12)</sup>

Fig. 4 Superthermal UCN source proposed for SNQ



## Considerations to "Real Time" Experiments

### on a pulsed Neutron Source

by Bruno Dorner (Ill-Grenoble)

Real time experiments (RTE) can be grouped in three different categories.

1. Irreversible processes such as many phase transformations and chemical reactions challenge studies during the time in which the process takes place. One would like to register a film of many pictures, where one picture stands for a whole spectrum like, for example, a powder-diagram, a small angle scattering profile, a topography, etc.. Such processes have their characteristic time scales. The shorter these times are the higher is the required intensity to produce a picture and finally a film. Such investigations are limited by the exposure time per picture. Pioneering work in the field of chemical reactions has been performed by Riekel (1980).
2. Reversible processes are easier to study, because the stroboscopic technique can be applied. Intensity is not crucial for the possibility to perform such experiments. The sample comes cyclicly into the same condition and is then illuminated by neutrons for a short time. These illuminations are repeated until the statistical accuracy of the spectrum is sufficient. Then the phase between the cyclic variation and the neutron puls is shifted to register another spectrum. Such a technique has been developed by Oberthuer (1984).
3. Pseudo RTE are those in which the sample can be brought into a particular condition only for a short time. Here one might think about very high magnetic fields produced by the discharge of a battery of capacitors (Badurek, 1984). It is not the development in time which is investigated (eventually it could become a very interesting study of category 2.) but a status which can be maintained only for a short time.



A pulsed neutron source like the SNQ might offer advantages for RTE. The performance of a pulsed neutron source is characterized by three properties: pulslength, peakflux and averaged flux.

The expected peak flux of SNQ seems still low to obtain a whole spectrum with one pulse. Therefore the SNQ has no particular advantage for experiments of category 1.. In the following we concentrate on category 2..

In a first consideration, the time resolution for RTE is given by the pulslength. But the additional use of a multichannelanalyser may improve the situation considerably.

To discuss the field of RTE in more detail in the following, we will distinguish three different time scales: a) ca. 1 sec, b) ca. 1 msec, c) ca. 10  $\mu$ sec.

For simplicity we start the discussion with experiments which need a monochromatic beam. This means we neglect first the "white" spectrum of the pulse for SNQ.

With this further restriction it is immediately clear that for time scales of about 1 sec the SNQ has no advantage (besides suppression of higher orders) in comparison with a continuous source of a flux comparable to the averaged flux.

As far as b) (ca. 1 msec) is concerned the natural pulse length of SNQ will be sufficient for the resolution in time. With a repetition rate of 10 msec it will be reasonable to use several pulses, before bringing the sample back into the starting condition.

If a stroboscopic phase shift to produce data in shorter time intervals than 10 msec is not necessary, then the SNQ has the advantage that each data point at 10 msec interval contains an average over only 150  $\mu$ sec. With a continuous source, comparable intensity can be obtained in comparable time if time channels of 10 msec are chosen. But this long averaging time reduces the significance of the data.

If several data points are wanted within the interval of 10 msec, then, with SNQ, corresponding phase shifted runs have to be performed. The measuring time will increase as much as with a continuous source and smaller channel widths. With increasing number of data points the advantage of SNQ (averaging

over short time) decreases until for 150  $\mu$ sec channelwidth the quality of the data becomes comparable.

In case c) (ca. 10  $\mu$ sec) a multichannelanalyser is necessary in any case. The SNQ has the highest advantage if the whole process covers a time shorter than the pulse length. The maximum gain for SNQ is peakflux/averaged flux and appears if the necessary time to "reload" the sample is 10 msec or longer.

The out-time for reloading the sample was neglected in case a) and b) because it was assumed that anyway several pulses will be used after the start of the process.

Now we lift the restriction to a monochromatic beam because many experiments for cases a) and b) will benefit from the pulsed white beam. For example, powder diffraction or small angle scattering (see the attached report of Ishikawa et al., 1984) using a multichannelanalyser will register the spectra for different wavelengths separately. This provides at first view an important gain, but one has to consider instrumental restrictions such as varying resolution for different wavelengths and the accessible momentum transfer. In small angle scattering for example, the long wavelengths provide information on small momentum transfer and correspondingly the short wavelengths are limited to large momentum transfer. Note, the neutrons with different wavelengths examine the sample at different times. A factor of true gain cannot be given in general, it depends on the particular investigation.

Experiments of class c) do not benefit from the white beam.

So far we discussed essentially experiments with elastic scattering. In inelastic scattering we study the Fourier transform of correlations in time for time scales orders of magnitude smaller than RTE in neutron scattering. In a RTE one might wish to study the intensity, position and width of a particular phonon or of a quasielastic line.

Inelastic scattering on a pulsed source is in most cases optimally performed in time of flight (TOF). The time of flight analysis of the energy may be used before scattering (white beam) or after scattering (monochromatic beam) in general. But in the particular case of RTE the monochromatic beam has the advantage to examine the crystal and to produce the inelastic signal at a short instance. In such a RTE the approximate position of the inelastic

signal in Q-w-space is known beforehand. Therefore a TOF instrument can be lined up in the most effective position.

On a continuous source one would use a three-axes-spectrometer (TAS) which has the loss-factor 6 (see Reichardt, this workshop) to start with. The time to reload the sample is a loss, if the characteristic time of the process is short and on the other hand if this time is long the significance of TAS data will suffer from long averaging times, as already discussed above.

In conclusion the SNQ has clear advantages over continuous sources for RTE in elastic and inelastic scattering. It appears particularly challenging to perform RTE on processes with a characteristic time comparable or shorter to the pulse length.

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Real-Time Experiments (RTE) in Small-Angle Neutron Scattering (SANS)

K. Ibel \*)

Universität Osnabrück, Fachbereich Biologie/Chemie

- Schwerpunkt Biophysik -

Barbarastr. 11, D 4500 Osnabrück

R.C. Oberthür

Bruchweg 10, D 4454 Bawinkel

The available data flux in optimized small-angle neutron cameras at high-flux reactors allows us to observe transient structures in time intervals of less than seconds. Numerous examples of RTE in SANS from previous and from our current work show the interest in RTE in material science, magnetism, physical chemistry of polymers and micellar systems as well as in biology. One limitation in the use of RTE in SANS stems from the condition, that the scattering event has to be uniquely related to the time of arrival of the scattered neutron at the detector. This condition restricts the use of large wavelength bands in measuring low momentum transfers. A practical limitation may be due to dead time losses in the transfer of counts to the data acquisition system. SANS cameras at pulsed neutron sources can profit from the temporal compression of the neutron beam. They may enable us to scan time-dependent structural changes in less than ms.

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\*) on leave of absence from Institut Laue - Langevin, Grenoble

### 1) Examples of RTE in SANS

Relaxation of magnetic domain orientation. (H. Rauch and J. Schelten, unpublished results). A series of RTE have been done at the Julich camera on the relaxation of domain orientation of Cobalt-precipitates in a Copper matrix for complementing neutron depolarization studies. Due to the Q-dependance of the scattering patterns, information about the size distribution of the precipitates could be obtained. Further experiments are in preparation.

Relaxation of spin glass magnetization. (Y. Ishikawa, M. Arai, M. Furusaka, M. Mera, KENS REPORT IV; KEK Internal (1983) 83-4). The relaxation times of the magnetization process at low temperatures in the cluster type spin glass  $88\text{FeTiO}_3 - 12\text{Fe}_2\text{O}_3$  were shown to be in the range of several ten to several 100 ms. The temporal variation of SANS has been measured using a pulsed neutron source, immediately after having applied or switched off a magnetic field of 5 kOe. The measurements were taken in time intervals of 50 ms, i.e., in the rhythm of the source. The value of the relaxation time has been found to depend on the momentum transfer. The quantitative analysis of the data as well as measurements at different temperatures are in progress.

Periodically deformed polymers. (A. Rennie, R.C. Oberthür, Revue Phys. Appl. 19 (1984) 765 - 768). A stretching apparatus allows to produce large cyclic deformations at frequencies up to 10 Hz which permits to collect spectra corresponding to different states of strain of the sample. Some data on model polydimethylsiloxane networks have been obtained. 20 % of the end-cross-linked chain molecules of relative molecular mass  $10^4$  were deuterium-labelled. A marked delay of the change in their dimensions with respect to the corresponding equilibrium values is observed at higher frequencies. The phase shift corresponds roughly to a relaxation time of 20 ms at the length scale of the overall size of the labelled chain segments between two crosslink points.

Kinetics of H/D-exchange in biological structures. (J. Haas, PhD thesis (1977) University of Mainz). The kinetics of H/D-exchange in proteins has been studied using a specially designed stopped-flow apparatus. It was possible to obtain useful information about zero angle scattering and radius of gyration with single shot experiments at time scales in the order of seconds.

Orientation relaxation of rod-like micelles. (L. Herbst, H. Hoffmann, J. Kalus, U. Schmelzer, H. Thurn, Verbundtreffen Neutronenstreuung, Maria Laach, 1984). RTE in SANS can supply information about the microscopic interaction between rod-like micelles. A viscoelastic solution of Cetylpyridiniumsalicylate was oriented by a shear gradient in a continuously running viscosimeter. The relaxation of the orientation of the rodlike micelles after rapidly stopping the rotation of the instrument has been observed in time intervals of 0.2 sec. The preliminary result of a theoretical interpretation showed that the degree of orientation was initially very high.

Frog muscle in the excited state. (D.L. Worcester, J.M. Gillies, O'Brien, ILL Experimental Report). The classical experiment done by Galvani in 1786 has been repeated using the D 11 camera, for investigating the molecular basis of muscle contraction. The excited state of the muscle was available for 0.2 seconds only. This time limit was set in order to avoid irreversible damages of the muscle. Between each excitation, the steady-state physiological conditions of the resting muscle had to be restored; the time needed to do so was in the order of 20 minutes. A detailed diffraction pattern with statistically relevant information was obtained after cyclic repetition during half a day. The experiments showed the absence of regular cross-bridges between the filaments, in the excited state. The exposure of the muscle to the neutron beam during the experiment did not induce structural changes due to radiation damage.

## 2) Limitations for RTE in SANS

Time dispersion in the secondary flight path. Out of intensity reasons, a broad spectrum in the incoming beam is normally used in SANS. However, with long wavelength neutrons and large distances between sample and detector, we risk at the site of the detector an overlap of slow neutrons with fast ones, which carry information about different structural states.

Rate of data acquisition. The maximum rate of data acquisition at the camera D11 is at about  $2 \text{ to } 3 \times 10^4$  cts/sec. It is set by the maximum speed of data transmission between the multiscaler and the external memory of the data acquisition system. It should be possible with modern electronics to achieve count rates which are by one order of magnitude larger.

Corollary remark. The heavy oversubscription of the Camera D 11 requires a thorough elaboration of the experimental schedule. For static experiments, many different samples can be prepared in advance, and a rapid turnover of an high number of samples in the instrument became a routine procedure. The camera itself can be kept in an universal configuration. Consequently, a high proportion of the data obtained within limited time could be published. With RTE in SANS, however, special equipment is needed. The time required for mechanical installation, for branching the control units in the data acquisition system, as well as for testing the whole system is lost for the scattering experiment. However, the development of sophisticated accessory equipment and procedures for conveniently installing it could be rewarding: RTE supplies unique information whilst economizing the amount sample material and reducing the influence of systematic errors.

3) RTE in SANS at SNQ

Cameras at pulsed neutron sources may in certain cases profit from the time structure of the source. The high peak flux in the neutron pulse will allow to efficiently probe sample structures at the well-defined time intervals between subsequent pulses. Stroboscopic effects can also be used with experiments in which the sample structure is altered in a cyclic manner, with a different period from that of the source. The change of structural state can thus be scanned with a time resolution of less than the pulse duration at the source, i.e., in less than msec. A gain factor up to the ratio of peak flux to mean flux may be achieved as compared to instruments at a continuous reactor of equal mean flux, if the time of arrival of the scattered neutron at the detector is uniquely related to the time of the passage of the neutron through the sample (condition of non-overlap, at the detector, of neutrons emerging from different pulses). The gain factor is reduced towards unity in experiments which require an high resolution in Q-space and a large beam diameter. The gain factor is less reduced if the frequency of the pulsed source is decreased.



Untersuchung von niederdimensionalen Phänomenen  
und Domänenproblemen an der SNQ

H. Boysen und F. Frey

I. Einleitung

Für viele geowissenschaftliche und physikalische Fragestellungen stellt die Analyse der Realstruktur von Kristallen eine wichtige Informationsquelle dar. Mit Realstruktur soll dabei nicht die Existenz einzelner (u.U. sehr vieler) Defekte, sondern das Fehl-ordnungsverhalten ganzer Kristallbereiche oder Teilstrukturen umschrieben werden. Beispiele hierfür sind: Domänen und Zwillinge (Größe, Form, Verteilungsfunktionen), Grenzflächenbereiche (mit oder ohne kohärente Verwachsungen), Ausscheidungen, Kristallwachstumsvorgänge an Störungen, Al-Mg-Si-Ordnungsvorgänge in Silikaten, Strukturen mit schwachgebundenen Teilgittern (Einlagerungsstrukturen, Ionenleiter, ...). Generell treten in Streubildern - oft miteinander kombiniert - Mehrfachreflexaufspaltungen, Satellitenstreuung und diffuse Streuung auf, die anisotrop im rez. Raum verteilt ist. In einfachen Fällen werden diffuse Stäbe oder Ebenen beobachtet, entsprechend langreichweitiger Korrelationen in nur 2 bzw. 1 Dimension (z.B. lamellare Domänen, Ketteneinlagerungen).

Im folgenden werden Überlegungen angestellt, ob sich Untersuchungen der genannten Art an der SNQ vorteilhaft durchführen lassen. Generelle Aussagen über Vor-/Nachteile gegenüber Messungen an einem steady-state-Reaktor sind jedoch nur sehr eingeschränkt möglich, da entsprechend der jeweiligen Problematik nicht ein Instrument an der SNQ, sondern das jeweilige diffuse Streuexperiment zu optimieren<sup>ist</sup>; dies muß von Fall zu Fall diskutiert werden. Benutzt wurden die aus den Arbeitsberichten bekannten technischen Daten der SNQ, sowie Zahlenwerte aus der Studie von Schelten und Bauer (S&B).

## II. Prinzipielle Gesichtspunkte

- (1) Die Variation diffuser Intensitäten erfolgt in der Regel nur langsam in ein oder zwei Dimensionen des rez. Raumes. Für die Unterscheidung verschiedener Fehlordnungsmodelle ist daher die Messung über große  $Q$ -Bereiche nötig:  $\Delta Q \left( \hat{=} \frac{4\pi \sin \vartheta}{\lambda} \right) \simeq 0.5 - 2.0 \text{ \AA}^{-1}$ . Die  $Q$ -Auflösung sollte dabei konstant für alle  $Q$  sein:  $\delta Q \simeq 0.1 \text{ \AA}^{-1}$ . Entsprechend der Anisotropie der diffusen Streuung sind auch anisotrope  $Q$ -Auflösungsbereiche vorteilhaft ( $\delta Q_{\parallel} \neq \delta Q_{\perp}$ ). Optimale scans sind meist senkrecht und parallel zum jeweiligen diffusen Phänomen durchzuführen. Die generelle Fixierung auf radiale scans in TOF-Maschinen ist dieser Problematik i.a. nicht angepaßt.
- (2) Für eine quantitative Analyse "niederdimensionaler Probleme" (Korrelationslängen, Bindungstypen, langreichweitige Domänen) bedarf es der Vermessung begrenzter diffuser Phänomene (in kleinen  $Q$ -Bereichen) mit hoher Auflösung:  $\delta Q \sim 0.01 \text{ \AA}^{-1}$  (auch bei großen  $Q$ ). Hierfür wird sinnvollerweise eine andere Meßanordnung im Vgl. zu (1) einzusetzen sein.
- (3) Die diffuse Streuung kann elast./quasielast./inelast. Natur sein ( $\rightarrow$  Domänenfluktuationen, Grenzflächenverschiebungen,... bzw. "klassische" gitterdynamische Prozesse). Ein wichtiges Kriterium ist somit die prinzipielle Unterscheidung zwischen statischen und dynamischen Problemen. Die Erfassung der rein elastischen Komponente ist somit eine wichtige Meßtechnik. Die Energie-Auflösung sollte rund  $\delta v \sim 0.5 \text{ meV}$  sein.
- (4) Viele Domänenordnungen vollziehen sich zeitabhängig. In real-time-Experimenten könnte die Variation der diffusen Streuung (Intensitäten,  $Q$ -Abhängigkeit) als Funktion der Zeit beobachtet werden. Hierfür scheint die SNQ ideal geeignet zu sein.
- (5) In der Regel ist die diffuse Streuung um Größenordnungen schwächer als die Bragg-Streuung. Ein niedriger Untergrund ist somit "lebensnotwendig". Die Unterdrückung schneller Neutronen und inkoh. inel. Untergrundstreuung an der SNQ ist hierfür vorteilhaft.

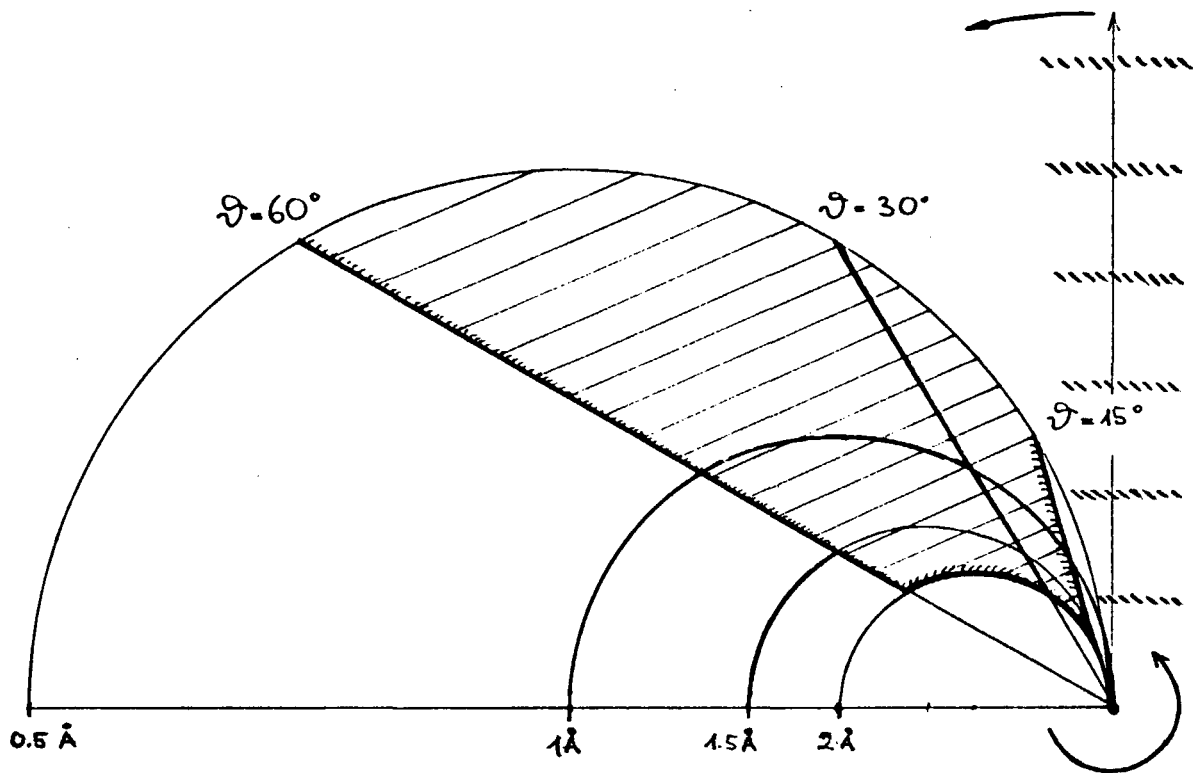


Abb.1 Laue-Methode: Nutzbarer Bereich in einem TOF-Diffraktometer mit Vielfachdetektor im Vergleich zu einem System diffuser Ebenen

### III. Flugzeitdiffraktometer

Ein Flugzeitgerät entsprechend dem Konzept S&B genügt den unter II (1) genannten Anforderungen nur sehr bedingt. Zwar kann - unter Benutzung eines Vielfachdetektors - ein ganzer Sektor des rez.Raumes erfaßt werden (Abb.1), doch ist dieser Sektor bei kleinen Streuwinkeln und großen  $\lambda$  sehr eingeschränkt. Der Kristall muß also nacheinander in verschiedene Positionen gebracht werden. Die radiale scan-Richtung wird zudem in den seltensten Fällen problemangepaßt sein. In der Regel werden rund 10 verschiedene Einstellungen nötig sein, um einen  $90^\circ$ -Sektor abzurastern. Es wird dabei sehr viel redundante Information anfallen. Gravierend ist ferner, daß die Q-Auflösung mit Q stark variiert:

$$\frac{\delta Q}{Q} = \frac{\delta \lambda}{\lambda} + \cot \vartheta \cdot \delta \vartheta; \quad (1) \quad Q = \frac{4\pi \sin \vartheta}{\lambda}$$

$$\begin{aligned} \delta Q &= Q \cdot \frac{\delta \lambda}{\lambda} + Q \cdot \cot \vartheta \cdot \delta \vartheta \\ &= Q^2 \cdot \frac{1}{4\pi \sin \vartheta} \cdot \delta \lambda + Q \cdot \cot \vartheta \cdot \delta \vartheta \\ &= \delta Q' + \delta Q'' \end{aligned}$$

Für die Variation von  $\delta Q$  ist somit der erste Term dominierend.  
Es gilt (s.S&B):

$$\frac{\delta \lambda}{\lambda_M} = \frac{\delta t}{T_M}; \quad \delta \lambda = \frac{\lambda_M}{T_M} \cdot \delta t \quad (2)$$

$\delta t$  = Chopper-Öffnungszeit,  $\lambda_M$  = mittlere Wellenlänge des Bandes,  
 $T_M$  entsprechend eine mittlere Laufzeit dieser Neutronen.

Innerhalb eines Wellenlängenbandes  $\Delta \lambda$  (eine Einstellung des Choppers  $\delta t = \text{const}$ ) variiert  $\delta Q \sim Q^2$ . Um ein zu starkes Anwachsen von  $\delta Q$  zu vermeiden, muß  $\delta t \sim \lambda^2$  gewählt werden (im Gegensatz zu S&B, die  $\delta t \sim \lambda$  wählen um  $\delta Q/Q$  konstant zu halten).

Mit der von S&B gewählten Geometrie (Distanz: Quelle - Detektor:  $L=10$  m) ergibt sich:

$$\delta \lambda [\text{\AA}] \approx \frac{L}{\lambda} \delta t \approx 0.4 \cdot \delta t [\text{ns}]$$

Tabelle 1 zeigt einige Zahlenwerte für  $\vartheta=60^\circ$  (Breite der Einzelbereiche:  $\pm 0.5$  \AA); für  $\delta \vartheta$  wurde angenommen:  $1.9 \times 10^{-2}$  rad (siehe S&B).

Tabelle 1

| $\lambda_m [\text{\AA}]$ | $\Delta Q [\text{\AA}^{-1}]$ | $\delta t [\mu\text{s}]$ | $\delta \lambda$ | $\frac{\delta \lambda}{\lambda} [\%]$ | $\delta Q' [\text{\AA}^{-1}]$ | $\delta Q'' [\text{\AA}^{-1}]$ | $\delta Q [\text{\AA}^{-1}]$ |
|--------------------------|------------------------------|--------------------------|------------------|---------------------------------------|-------------------------------|--------------------------------|------------------------------|
| 1                        | 21.8 - 7.3                   | 10                       | .004             | 0.4                                   | .174- .019                    | .239- .080                     | .413 - .099                  |
| 2                        | 7.3 - 4.3                    | 40                       | .016             | 0.8                                   | .077- .030                    | .080- .048                     | .157 - .078                  |
| 3                        | 4.3 - 3.1                    | 90                       | .036             | 1.2                                   | .062- .032                    | .048- .035                     | .110 - .067                  |
| 4                        | 3.1 - 2.4                    | 160                      | .064             | 1.6                                   | .057- .034                    | .035- .026                     | .092 - .060                  |

Benutzt man einen Flächendetektor (Variation von  $\phi$ ), so sind für jedes Wellenlängenband neue Zahlenwerte einzusetzen. Übrige Überlegungen siehe S&B.

#### Diskussion:

Will man dieses "Laue-Verfahren" nutzen, so sind

- (a) Messungen mit verschiedenen Wellenlängenbändern
- (b) Messungen bei verschiedenen Kristallpositionen

nötig. Die Messungen müssen aneinander angepaßt werden, was wegen unterschiedlicher Auflösungselemente im Überlappbereich mühsam ist. Messungen bei konstantem  $\delta t$  führen zu einer übermäßigen Variation von  $\delta Q$ . Die von S&B abgeschätzten Gewinnfaktoren SNQ - steady-state-reactor werden dadurch reduziert. Hinzukommt, daß die Methode nicht problemangepaßt ist, sowie Fokussierungstechniken, die mit einem konventionellen Diffraktometer möglich sind, nicht bekannt sind (?). Es bleibt somit offen, ob insgesamt ein Gewinn resultiert. Dies wird für das jeweilige Experiment zu überprüfen sein.

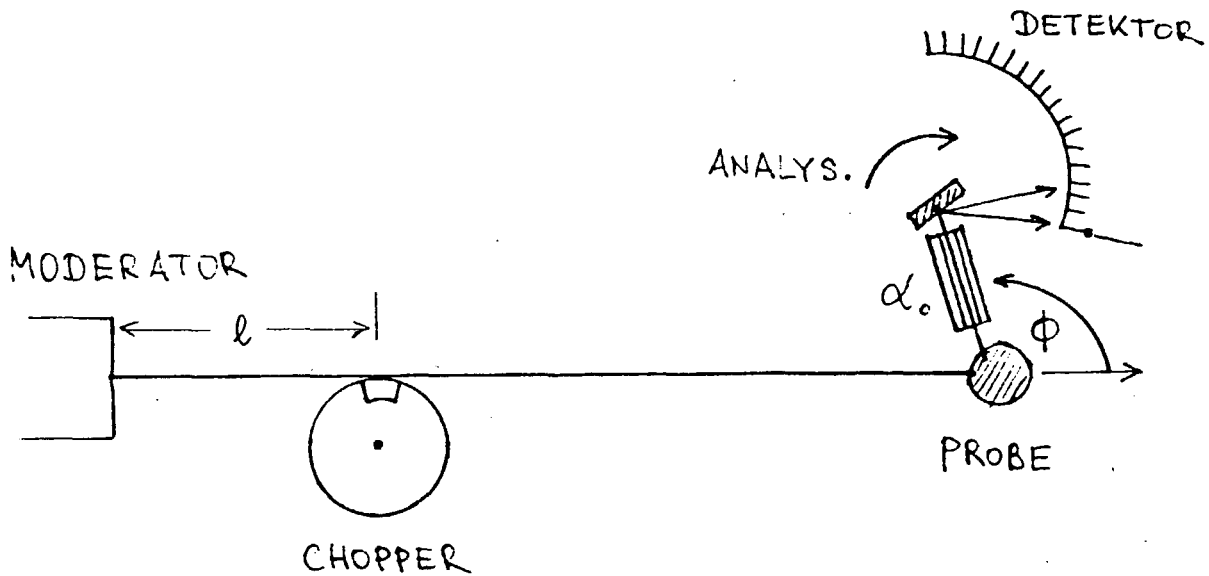


Abb.2 Diffraktometer mit rotierendem Analysator-Kristall, dessen Frequenz auf ein zu messendes Wellenlängenband und die Pulsfrequenz der SNQ abgestimmt ist.

#### IV. Diffraktometer mit rotierendem Analysator-Kristall

Eine Wellenlängeanalyse kann auch über einen rotierenden (Analysator-)Kristall erfolgen (Abb. 2). Über den Streuwinkel  $\phi$  werden die Q-Vektoren festgelegt. Ein Chopper kann zusätzlich für Wellenlängenbandbegrenzung (nicht Auflösung) eingesetzt werden (Abb. 3). Wesentlich ist die Abstimmung der Rotationsfrequenz mit der Flugzeit  $t$  der Neutronen aus dem gewünschten Wellenlängenband:

$$\lambda = \frac{4}{L} \cdot t$$

$$\lambda = 2 \cdot d_A \cdot \sin \vartheta_A(\tau) = 2 d_A \sin \omega \tau \quad (3)$$

Tabelle 2 zeigt einige Zahlenwerte die für folgende Werte berechnet wurden:

Abstand Moderator Analysator: 3.8 m (!) [siehe Anhang S.17]

Analysator: Cu(111)  $\rightarrow d_A \approx 2 \text{ \AA}$

Genutztes Wellenlängenband:  $\Delta\lambda = 0.5 - 3.3 \text{ \AA}$

Winkelbereich:  $\Delta\vartheta_A = 7.2 - 56^\circ \approx 49^\circ$

Reflektiert der rotierende Kristall während eines Umlaufes zweimal, so erhält man für eine der Pulsfrequenz der SNQ von 100 Hz angepaßte Winkelgeschwindigkeit von :

$$\omega = \frac{\Delta\alpha}{\Delta\tau} = \frac{180^\circ}{10} [\%/ms] = 18 [\%/ms] (\hat{=} 50 \text{ Hz})$$

$$\tau_\lambda = \frac{\vartheta_A(\tau)}{\omega}$$

Tabelle 2

| $\lambda [\text{\AA}]$ | $t [\text{ms}]$ | $\tau [\text{ms}]$ | $\Delta t = t - \tau [\text{ms}]$ |
|------------------------|-----------------|--------------------|-----------------------------------|
| 0.5                    | 0.48            | 0.40               | 0.08                              |
| 1                      | 0.95            | 0.80               | 0.15                              |
| 2                      | 1.90            | 1.67               | 0.23                              |
| 3                      | 2.85            | 2.70               | 0.15                              |
| 3.3                    | 3.14            | 3.09               | 0.05                              |

Im Hinblick auf eine Pulslänge von  $250 \mu\text{s}$  muß gelten:

$$\Delta t < 0.25 [\text{ms}];$$

Zur

Bandbegrenzung kann noch ein Chopper nahe am Moderator plaziert werden ( $l = 1\text{m} : t = 700 \mu\text{s}$ ).

Die Q-Auflösung entspricht näherungsweise der eines konventionellen 2-Achsen-Diffraktometers und wird durch die Kollimatoren und Mosaikwinkel von Probe und Analysator bestimmt (s. Cooper & Nathans, Acta Cryst. ). Nutzt man an der Probe große Streuwinkel ( $\phi \sim 120 - 140^\circ$ ), so werden die Bestimmungstücke des Analysatorteils entscheidend sein. Generell wird eine ungefähr lineare Abhängigkeit von Q vorliegen, so daß besonders bei kleinen Q-Werten mit einem langsamer variierenden Auflösungsbereich im Vergleich zu III. gemessen werden kann.

Eine ganz grobe Energieanalyse (praktikabel für Wellenlängen  $\lambda_{\min} \sim 2 \text{ \AA}$ ) ergibt sich in dieser Konfiguration aus Flugstrecke und Pulslänge (vgl. Abb. 4):  $\delta v [\text{THz}] \simeq 40 \cdot \frac{\delta \lambda}{\lambda^2}$ ;  $\delta \lambda = \frac{4}{L} \cdot \Delta t_{\text{puls}} \simeq 0.263 \text{ \AA}$   
 $\delta v: 1.3 \text{ THz } [\lambda = 2 \text{ \AA}]$ ;  $\delta v = 0.4 \text{ THz } [\lambda = 3 \text{ \AA}]$ ;

Diese Zahlen sind nur obere Grenzwerte. Die tatsächliche Energieunschärfe  $\pm \delta v$  ist kleiner und i.a. nicht symm. bez. Verlust-/Gewinnprozessen. Mittels eines Choppers (s.o) lassen sich die Energiefenster wesentlich verkleinern, falls auf die volle Bandbreite  $\Delta \lambda$  verzichtet wird. Günstig wäre ein Abstand Moderator-Chopper  $< 1\text{m}$ , was allerdings technisch nicht realisierbar erscheint. Diese Energieauflösung wird durch den Analysator nicht wesentlich beeinträchtigt.

Da Neutronen innerhalb des gewählten Bandes aus verschiedenen Zeitpunkten des Primärpulses stammen, ist eine diesbezügliche sorgfältige Eichung nötig. Nachteilig ist, daß für einen Winkel  $\phi$  eine Detektorbank (Lineardetektor) benötigt wird, der sehr nahe am Analysator plaziert werden kann. Es lassen sich prinzipiell mehrere  $\phi$ -Winkel nutzen.



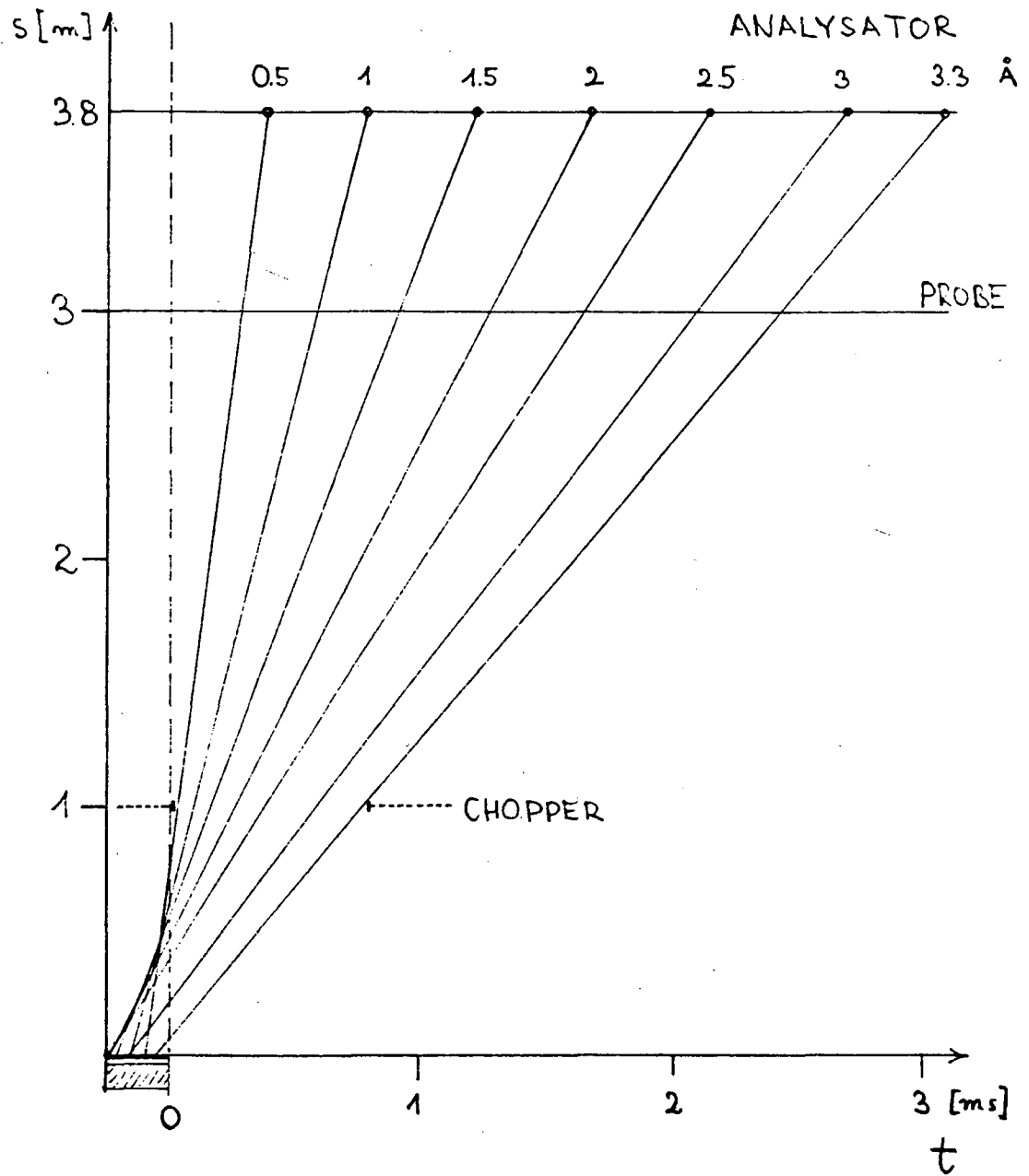


Abb.3 Weg-Zeit-Diagramm für ein Diffraktometer mit rotierendem Analysator-Kristall (Abstände: M-A: 3.8 m; M-P: 3 m)

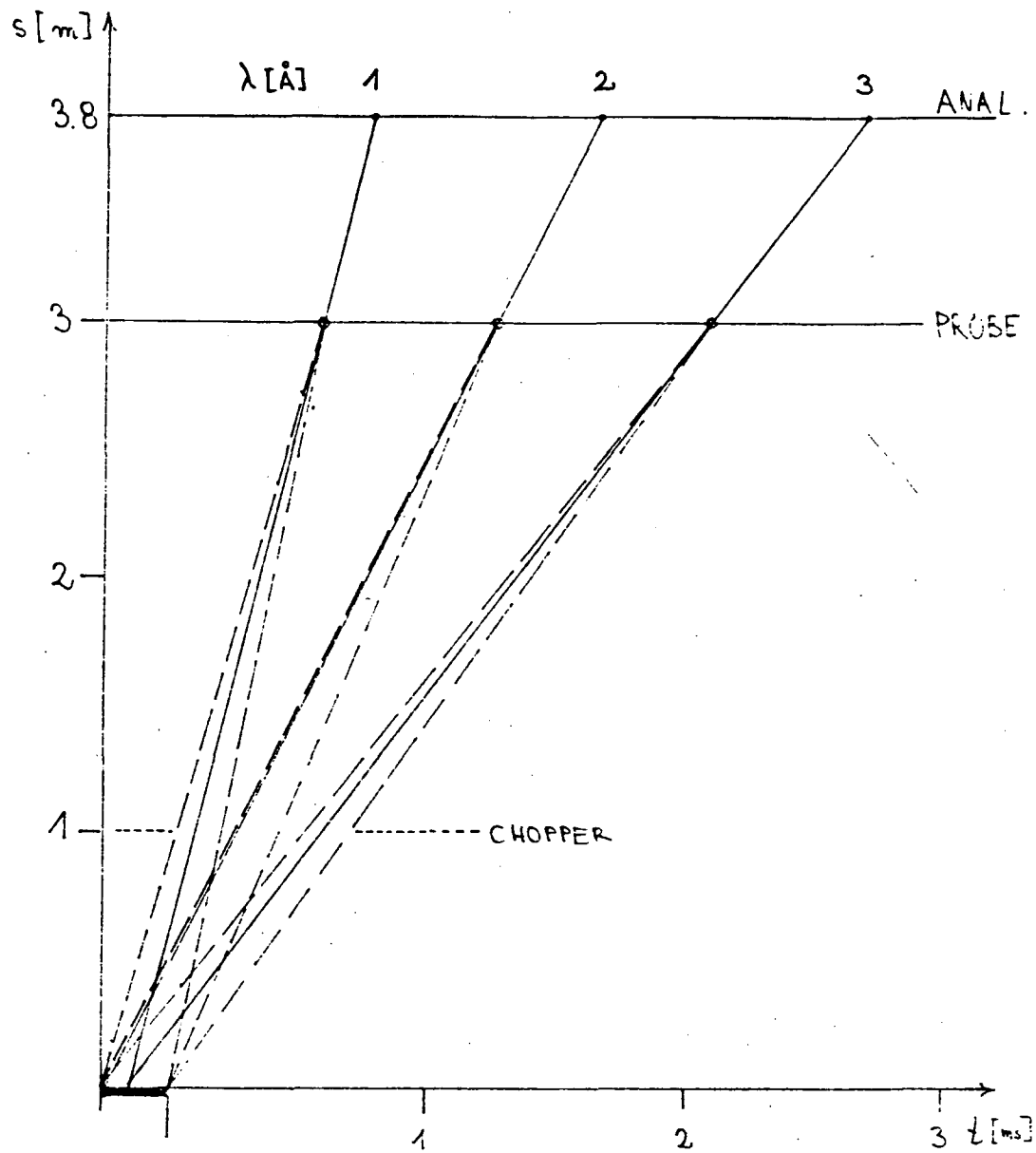


Abb.4 Auflösung eines Diffraktometers mit rotierenden Analysator-Kristall (Chopper nur  $\lambda$ -Band begrenzend)

Luminosität:

Die Luminosität eines solchen Geräts ergibt sich aus:

$$\Lambda \approx \bar{\Phi} \cdot 40 \cdot R_A \cdot \delta\alpha_H \cdot \delta\alpha_V \cdot \frac{2\Delta\vartheta_A}{360} \cdot \frac{\delta\lambda}{\lambda} \cdot R^\Theta \quad (4)$$

Es ist:

$\bar{\Phi}$  = der mittlere Fluß ;  $\hat{\Phi}/\bar{\Phi} = 40$

$R_A$  = die (intrinsic) Reflektivität des Analysators ( $\leq 0.5$ )

$\delta\alpha_H$  = Horizontaldivergenz (bestimmt durch Abstand: M-P: 3m und Quellengröße: 10 cm; Kollimator:  $\alpha_0 \sim 0.3^\circ$ ):  $\delta\alpha_H \approx 0.6 \cdot 10^{-2}$

$\delta\alpha_V$  = Vertikaldivergenz (ebenfalls durch obige Größen bestimmt, falls Analysator und Detektorfenster genügend groß gemacht werden):  $\delta\alpha_V \approx 10/300 \approx 3 \cdot 10^{-2}$  (eher:  $2 \cdot 10^{-2}$ )

$2\Delta\vartheta_A/360$  = Nutzfaktor des rotierenden Analysators (der Pulsfrequenz angepaßte Winkelgeschwindigkeit)

Hier:  $2 \cdot 49/360 \approx 0.3$

$\delta\lambda/\lambda$  = Wellenlängenauflösung ; wird in gröbster Näherung durch  $\text{ctg } \vartheta_A \cdot \delta\vartheta_A$  bestimmt, ist also nicht konstant über den genutzten Winkelbereich

$R^\Theta$  = integrale Reflektivität des rotierenden Kristalls (ohne intrinsic Reflektivität); für einen dicken Mosaikkristall gilt:  $R^\Theta \sim \tilde{Q}/2\mu$  ( $\tilde{Q} = \frac{\lambda^3}{8\pi^2\vartheta_A} \cdot K$  ;  $\mu$  = Absorptionsfaktor )  
Der Faktor K ist aber bereits in  $R_A$  enthalten, so daß in obige Abschätzung ein  $R^{\Theta'} = R^\Theta / K$  eingeht.

Diese Größe  $\Lambda$  soll mit der äquivalenten Luminosität eines Referenzdiffraktometers an einem Reaktor verglichen werden:

$$\Lambda' \approx \bar{\Phi}' \cdot R'_M \cdot \delta\alpha'_H \cdot \delta\alpha'_V \cdot \left(\frac{\delta\lambda}{\lambda_0}\right)' \quad (5)$$

( $R'_M$  = Monochromatorreflektivität; übrige Größen analog)

Unter folgenden Annahmen:  $\bar{\Phi} \approx \bar{\Phi}'$  ;  $\frac{\delta\lambda}{\lambda} \approx \left(\frac{\delta\lambda}{\lambda_0}\right)'$  ;  $\delta\alpha_{H/V} \approx \delta\alpha'_{H/V}$  ;

$R'_M \approx R_A$  (nur zahlenmäßig! versch. Dimension);

läßt sich abschätzen:

$$\frac{\Lambda}{\Lambda'} \approx 0.3 \cdot 40 = 12$$

(Der Absorptionsfaktor wurde vernachlässigt !)

Dieser Gewinn-Faktor stellt allerdings nur eine sehr grobe Näherung dar, da detektierte Neutronen unterschiedlicher Wellenlänge aus verschiedenen Zeitpunkten innerhalb des Pulses stammen. Hinzu kommt natürlich die  $\lambda$ -Abhängigkeit von  $R^0 (\sim \lambda^3)$ . Somit ist also eine sehr detaillierte Analyse der Intensitäten nötig.

Eine Verbesserungsmöglichkeit wäre der Einsatz gekrümmter Analysatorkristalle, wodurch sowohl  $\delta\lambda$  vergrößert als auch die Horizontaldivergenz gesteigert werden könnte. Andererseits wurde nicht berücksichtigt, daß Fokussierungstechniken und spezielle Meßstrategien an einem konventionellen Diffraktometer (steady-state-Reaktor) Gewinne ermöglichen.

Der Vorteil gegenüber III liegt darin, daß mit einer Chopper-Einstellung ein größeres  $\lambda$ -Band erfaßt wird und die Auflösung  $\delta Q$  langsam variiert. Im Vergleich zu III fällt natürlich auf, daß kein Flächendetektor eingesetzt werden kann. Doch muß auch bei III die Kristallorientierung variiert werden ( $\omega$ -scan), falls die Information aus einem vollen  $90^\circ$  Sektor des rez. Raumes (kleine  $Q$  !) benötigt wird.

## V. Spektrometer für rein elastische (diffuse) Streuung

Es werden Überlegungen angestellt, ob sich das Flugzeitgerät III mit einem "Rotierenden-Analysator-Kristall-Diffraktometer" so kombinieren lassen, daß nur elastisch gestreute Neutronen separiert werden können. Das Energiefenster  $\delta E \approx \delta v$  (zentriert um 0) sollte dabei  $\delta \lambda / \lambda$  (siehe III) angepaßt sein. Ferner sollte die Winkelgeschwindigkeit des Analysators der Pulsfrequenz der SNQ angepaßt sein.

Es ergibt sich aus :

$$\frac{\delta \lambda}{\lambda} = c \lg \vartheta \cdot \delta \vartheta \rightarrow \delta \vartheta = \frac{1}{c \lg \vartheta} \cdot \frac{\delta \lambda}{\lambda} \quad \text{mit: } \delta v_{[\text{meV}]} = 40 \frac{\delta \lambda}{\lambda^3} [\text{\AA}]$$

$$\delta v = 40 \cdot \frac{1}{\lambda^2} \cdot c \lg \vartheta \cdot \delta \vartheta$$

$\delta \vartheta$  wird andererseits durch die Kollimationen  $\alpha_0, \alpha_1$  bzw. dem Mosaikwinkel des Analysatorkristalls  $\gamma_A$  bestimmt:

$$\delta \vartheta = \sqrt{\frac{\alpha_0^2 \alpha_1^2 + \alpha_0^2 \gamma_A^2 + \alpha_1^2 \gamma_A^2}{\alpha_1^2 + \alpha_0^2 + 4 \gamma_A^2}}$$

$$\alpha_1 \gg \alpha_0, \gamma_A : \delta \vartheta \approx \sqrt{\alpha_0^2 + \gamma_A^2}$$

Ein typischer Wert für  $\delta \vartheta$  ist:  $0.3^\circ \approx 0.005 \text{ rad}$

Für einen zahlenmäßigen Vergleich wurde wieder das Beispiel Cu(111) gewählt:

Tabelle 3

| $\lambda [\text{\AA}]$ | $\vartheta [^\circ]$ | $\delta \vartheta = \frac{\delta \lambda}{\lambda} \frac{1}{\lg \vartheta} \rightarrow \delta v$ | $\delta v (\approx \delta \vartheta \sim 0.3^\circ)$ | $\tau [\text{ns}]$ |
|------------------------|----------------------|--------------------------------------------------------------------------------------------------|------------------------------------------------------|--------------------|
| 0.5                    | 7.2                  | 0.001                                                                                            | 1.27                                                 | (6.3)              |
| 1                      | 14.5                 | 0.001                                                                                            | 0.15                                                 | (0.8)              |
| 1.5                    | 22.0                 | 0.003                                                                                            | 0.13                                                 | 0.22               |
| 2                      | 30.5                 | 0.005                                                                                            | 0.09                                                 | 0.09               |
| 3                      | 48.6                 | 0.014                                                                                            | 0.02                                                 | 0.02               |
| 3.3                    | 55.6                 | 0.018                                                                                            | 0.014                                                | 0.01               |

Für  $\lambda \gtrsim 1.5 \text{ \AA}$  ist somit eine sinnvolle Energieauflösung und -anpassung möglich. Es gilt andererseits ( $L = 10 \text{ m}$ ):

$$\vartheta_A(\tau) = \arcsin \frac{\lambda(\tau)}{2d} = \arcsin \frac{4}{2 \cdot d \cdot L} \tau = \arcsin(0.1 \cdot \tau)$$

Werte für  $\tau$  sind in der letzten Spalte eingetragen. Man erkennt, daß für einen Nutzbereich von  $\Delta \vartheta \approx 60^\circ$ :  $\tau_{\max} \approx 10 \text{ ms}$  beträgt, d.h.  $180^\circ \leftrightarrow 30 \text{ ms}$ . Entweder läßt sich nur jeder 3. Puls ausnutzen oder es muß ein oszillierender Bewegungsmechanismus für den Analysator entwickelt werden (Realisation?).

#### VI. Spektrometer mit hoher Q-Auflösung

Gemäß II(2) gibt es Probleme, bei denen eine Auflösung von  $\delta Q \sim 0.01 \text{ \AA}^{-1}$  in einem begrenzten Q-Bereich benötigt wird. Nach (1) gilt:

$$\delta Q = \frac{Q^2}{4\pi \sin \vartheta} \cdot \delta \lambda + Q \cdot d_\vartheta \vartheta \cdot \delta \vartheta$$

Kristallspektrometer in Rückstreugeometrie:  $\delta Q \rightarrow 0$

Flugzeitmaschine:

$$\delta Q \sim 0.01 \text{ \AA}^{-1} \rightarrow \delta \lambda = \frac{4\pi \sin \vartheta}{Q^2} \delta Q = \frac{4}{L} \delta t$$

Diese Forderung kann über lange Flugwege/kleine  $\delta t$  erfüllt werden.

Beispiel (Tabelle 4 für  $\vartheta = 60^\circ$  und  $L = 100 \text{ m}$  gerechnet)

Tabelle 4

| $\lambda [\text{\AA}]$ | $t [\text{ms}]$ | $Q [\text{\AA}^{-1}]$ | $\delta \lambda [\text{\AA}]$ | $\delta t [\mu\text{s}]$ | $\delta \nu [\text{THz}]$ |
|------------------------|-----------------|-----------------------|-------------------------------|--------------------------|---------------------------|
| 0.54                   | 13.5            | 20                    | 0.0003                        | 7.5                      | 0.08                      |
| 1.09                   | 27.3            | 10                    | 0.001                         | 25                       | 0.08                      |
| 2.18                   | 54.5            | 5                     | 0.004                         | 100                      | 0.025                     |
| 2.72                   | 68.0            | 4                     | 0.007                         | 175                      | 0.015                     |

Lange Flugstrecken können mittels Neutronenleiter realisiert werden. Ein zweiter Chopper ist zur Impulsunterdrückung nötig. Die volle Pulszahl kann bei Einschränkung des  $\Delta\lambda$ -Bandes auf  $0.4 \text{ \AA}$  genutzt werden. (Öffnungszeit des gephasen 2. Choppers in 1 m Entfernung:  $100 \text{ \mu s}$ )

Wie die letzte Spalte in Tabelle 4 zeigt, ist eine Energieanalyse mit  $\delta v < 0.1 \text{ THz}$  möglich. Das begrenzende Element in einer Anordnung analog V. ist die Analysatoreinheit. Fordert man  $\delta v = 0.1 \text{ THz}$  so ergibt sich mit einem  $\delta\vartheta = 0.005 \text{ rad}$ :

$$\delta\lambda = \frac{0.1}{40} \lambda^3 ; \quad t_{\delta\vartheta} = \frac{\lambda}{\delta\lambda} \delta\vartheta = \frac{40}{0.1} \frac{1}{\lambda^2} \delta\vartheta$$

| $\lambda [\text{\AA}]$ | $\vartheta [^\circ]$ | $d = \lambda / 2 \sin \vartheta [\text{\AA}]$ |
|------------------------|----------------------|-----------------------------------------------|
| 1                      | 63.4                 | 0.56                                          |
| 1.5                    | 41.6                 | 1.15                                          |
| 2                      | 26.5                 | 2.24                                          |

Somit muß für jeden Bereich eine andere Analysatoreinstellung bzw. ein anderer Analysator gewählt werden. Darüber hinaus sind natürlich die zugänglichen Q-Bereiche eingeschränkt. Tabelle 5 gibt einige Zahlenwerte für folgende Annahmen:

$$\lambda = 1.5 \text{ \AA} \quad (d_{\text{A}} = 1.15 \text{ \AA}) ; \quad \rightarrow \quad \delta\lambda = 0.0084$$

Tabelle 5

| $\lambda [\text{\AA}]$ | $\vartheta [^\circ]$ | $t [\mu s]$   | $\tau [\mu s]$   | $\tau + 6.92$ |
|------------------------|----------------------|---------------|------------------|---------------|
| 1.4                    | 38.30                | $t_0 = 35.00$ | $\tau_0 = 28.08$ | 35.00         |
| 1.45                   | 39.95                | 36.25         | 29.28            | 36.20         |
| 1.5                    | 41.63                | 37.50         | 30.52            | 37.44         |
| 1.55                   | 43.35                | 38.75         | 31.78            | 38.77         |
| 1.6                    | 45.12                | 40.00         | 33.08            | 40.00         |

Die Soll-Rotationsgeschwindigkeit ergibt sich somit zu:

$$\omega = \frac{\Delta\vartheta}{\Delta t} = \frac{45.12 - 38.3}{40 - 35} = 1.364 [^\circ/\mu s] \quad (\pm 3.8 \text{ Hz})$$

Die (konst.) Phase beträgt:  $\Delta = t - \tau_0 = 6.92$  ;  $\Delta t = t - \tau \lesssim 60 \text{ \mu s}$ .  
Verglichen mit dem aus  $\delta Q \sim 0.01 \text{ \AA}^{-1}$  errechneten  $\delta t = 51 \text{ \mu s}$  ist dieser Wert nur geringfügig größer. Man kann also in einem begrenzten Q-Bereich mit einem ausgewählten Analysator Messungen mit sehr guter Q- und Energieauflösung durchführen.

## VII. Allgemeine Schlußfolgerungen

Für die Messung anisotroper diffuser Streuung könnten Flugzeit- bzw. Hybrid-Geräte an der SNQ für schnelle Übersichtsmessungen eingesetzt werden. In Verbindung mit Linear- bzw. Flächendetektoren lassen sich ganze Bereiche simultan erfassen, wenn auch dabei ein großer Anteil nutzloser Information gesammelt wird. Günstig sind der relativ geringe Untergrund, sowie die Unterdrückung schneller Neutronen. Die Separation elast./unelast. Streuung ist bis <sup>zu</sup> mittleren  $Q$ -Beträgen möglich, ebenso Messungen mit hoher  $Q$ - und Energieauflösung in schmalen  $Q$ -Bereichen. Die radiale scan-Richtung bei TOF Maschinen ist in der Regel den anisotropen und nicht homogen verteilten diffusen Streuererscheinungen nicht anpaßbar. Es werden Einzelmessungen bei verschiedenen Probenorientierungen nötig sein. Für eine quantitative Auswertung diffuser Daten erscheint die Variation der Auflösungsbereiche im rez. Raum nicht unproblematisch. Diese müssen durch Wechsel der Probenorientierung, Chopperöffnungszeiten oder auch Wechsel des Analysatorkristalls aneinander angepaßt werden. Die Variation der Auflösungselemente tangiert auch die Untergrundsfrage, da der inkohärent, inelastische Untergrund ungleichmäßig erfaßt bzw. unterdrückt wird. Durch diese Aufteilung in Einzelmessungen werden die von S&B abgeschätzten Gewinnfaktoren merklich reduziert. Daher erscheint die Meßmethode mit rotierendem Analysator-Kristall günstiger, da dort die Auflösung stetig variiert. Quantitative Abschätzungen bezüglich Intensitäten wurden nicht vorgenommen : Diese müssen für jedes spezielle Problem einzeln durchgeführt werden (Gitterperioden, Ausdehnung und Variation der diffusen Streuung im rez. Raum, Modulationen innerhalb der diffusen Streuung,...). Ferner muß sehr genau die zeitliche und spektrale Struktur der Pulse bekannt sein. Andererseits kann in günstigen Fällen mit Gewinnfaktoren bis zu 10 gerechnet werden. Die Anordnung mit rotierendem Analysator erscheint auch für die Messung isotroper diffuser Streuung bzw. in einem Pulverdiffraktometer vorteilhaft zu sein.



## Anhang

Da nach neueren Angaben die Abschirmung einen Mindestabstand Moderator - Probe  $\geq 6$  m bedingt, ließe sich das auf S.7 angegebene Konzept nur mit doppelter Pulslänge ( 500  $\mu$ s) und halber Pulsfrequenz (50 Hz) der SNQ realisieren: Es wäre dann zu wählen  $L = 7.6$  m,  $\omega = 25$  Hz. Mit den vorgesehenen Kenndaten (250  $\mu$ s, 100 Hz) wäre jeder 2. Puls zu unterdrücken. Dieser Verlust gegenüber dem Konzept S.7 wird jedoch teilweise wieder kompensiert durch die längere "Öffnungszeit" des - langsamer rotierenden - Analysators. Bei eingeschränkter  $\lambda$ -Bandbreite wird ferner die auf S.8 erwähnte "Energieanalyse" ( $\delta v$ ) verbessert. Tabelle 6 gibt einen Vergleich. Dabei ist:

$\tau_A$  die "Öffnungszeit" des Analysators bei einem nutzbaren Wellenlängenband  $\Delta\lambda$ ,  $f'$  der Verlustfaktor durch Impulsunterdrückung bzw.  $f$  der maximale Gewinnfaktor.

Tabelle 6

| $\omega$ [Hz] | $L$ [m] | $f'$ | $\Delta\lambda=0.5 \dots$ | $\tau_A$ [ms] | $f = 40(?) \times \dots$ | * s.S.8<br>$\lambda_{\min}$ (bei $\delta v=0.5$ THz) |
|---------------|---------|------|---------------------------|---------------|--------------------------|------------------------------------------------------|
| 50            | 3.8     | 1    | ... 3.6                   | 2.6           | $\times 0.26$            | 2.8                                                  |
| 25            | 7.3     | 0.5  | ... 3.0                   | 4.6           | $\times 0.23$            | 2.2                                                  |
| 162/3         | 10.7    | 0.33 | ... 2.8                   | 6.2           | $\times 0.21$            | 2.0                                                  |
| 12.5          | 14.1    | 0.25 | ... 2.7                   | 7.7           | $\times 0.19$            | 1.8                                                  |

Durch Vervielfachung der Analysatorkristalle in einer prismatischen Anordnung könnten Pulsunterdrückungen vermieden werden.