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Lecture Notes of the JCNS Laboratory Course Neutron Scattering (Forschungszentrum Jülich, 2012, all rights reserved)

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2.1 Introduction

Neutrons are an extremely versatile probe to investigate the fundamental properties of matter. The possible applications range from fundamental questions (e.g. electrical dipole moment of the neutron) over condensed matter physics and chemistry to material science and life sciences. The reason for this is threefold:

- The neutron is electrically neutral: hence it can penetrate deeply into matter and prove truly the bulk properties. If you use other massive particles to investigate the properties of matter such as α particles or electrons, you probe usually only the regions close to the surface. Even for x-ray, which is also considered as an bulk technique in general, you penetrate only several hundreds of nm, if you use wavelength delivered by an laboratory x-ray tube.
- The neutron interacts with the sample via nuclear forces: hence the interaction cross section depends on the internal structure of the nuclei in your sample and not on the mass or electric charge of the whole atom. Neutrons are sensitive more or less equally to heavy and light atoms, making them an ideal probe for samples containing hydrogen, carbon or oxygen next to any other heavier atom.
- The neutron has a large magnetic moment: hence it is extremely sensitive to the magnetic properties of your sample. The magnetic field created by the sample scatters the neutron and the analysis of the direction, into which the neutrons are scattered, and the number of scattered neutrons provides the information about the magnetic structure, the size of the magnetic moments and the coupling between different magnetic sites.

Neutrons are in particular useful, because their energy and wavelength corresponds very well with the interatomic distances and the typical excitations in condensed matter problems. We calculate the kinetic energy of a free neutron

$$E_{kin} = \frac{1}{2}m\mathbf{v}^2 \tag{2.1}$$

$$=\frac{\mathbf{p}^2}{2m}\tag{2.2}$$

$$= \frac{\mathbf{p}^2}{2m} \tag{2.2}$$

$$= \frac{h^2}{2m\lambda^2}, \tag{2.3}$$

using the de Broglie relation, that expresses the wavelength of a quantum mechanical particle with momentum p:

$$\lambda = \frac{h}{|\mathbf{p}|} \tag{2.4}$$

If we insert the natural constants, we get

$$E(\lambda) = 81.805 \text{ meV} \text{Å}^2 \times \lambda^{-2}$$
 (2.5)
 $v(\lambda) = 3956 \text{ ms}^{-1} \text{Å} \times \lambda^{-1}$ (2.6)

$$v(\lambda) = 3956 \text{ ms}^{-1} \text{Å} \times \lambda^{-1}$$
 (2.6)

In other words, if we provide neutrons with a wavelength $0.8 < \lambda < 20$ Å suitable for resolving interatomic distances in condensed matter, these neutrons are also ideally suited to study the dynamics in the energy range 0.001 < E < 100 meV.

Apparently the properties of the neutrons make them a attractive probe for a wide variety of applications. In the reminder of the lecture I will try to answer the question, what the providers of neutrons, e.g. JCNS, FRM II, ILL, SNS..., can do to make their users happy. Therefore we first need do understand, what users want. We consider an generic neutron spectrometer, that allows to measure transfer of energy and momentum between neutron and the sample, see Fig. 2.1. How this is done, you will learn in the other lectures of the course and mainly during the practical part. The signal you get finally at the detector of your instrument can be expressed in

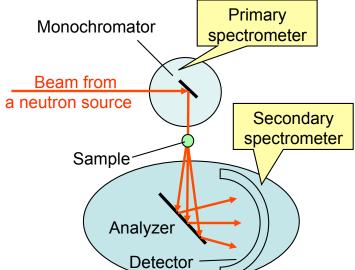


Fig. 2.1: Generic layout of a neutron spectrometer

the following way:

$$I_{det} = I_0 \epsilon_{pr} \epsilon_{sec} \epsilon_{det} \sigma_{sample} V_{sample} + \text{background}$$
 (2.7)

 I_0 is the incident neutron brilliance, i.e. the number of neutrons per second emitted from the source normalized by area, solid angle and energy or wavelength interval, ϵ_x denotes the efficiencies of the primary and the secondary spectrometer and the detector, σ_{sample} , V_{sample} is the cross section and the Volume of the sample, respectively. If you have an interesting scientific question that has not been answered yet, usually the both the cross section and volume are small. Hence to get good data, you need first an efficient instrument with a good signal to noise ratio, which detects ideally all and only the neutrons scattered by the sample. Second you need a low background that allows you to distinguish also tiny signals. And last but not least you need an intense source of neutrons, that brings a lot of useful neutrons to the instrument.

2.2 How do we get free neutrons?

The free neutron has a mean lifetime of about 900 s, hence it is necessary to produce the free neutrons as you run your experiment. While most nuclei are constituted to more then 50 % by neutrons, nuclear forces confine them and hence it is rather difficult to set neutrons free. Nowadays free neutron for scientific applications are released by nuclear reactions mainly in fission reactors or in spallation sources. Both routes require large scale facilities, that operate the source and provide state-of-art instrumentation. One example for the nuclear research reactor

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is the FRM II, where you will perform the practical part of the Laboratory Course. The most powerful spallation source is the SNS installed at the Oak Ridge National Laboratory in the USA. The neutron as a free particle was discovered by James Chadwick in 1932, when he investigated the radiation from Beryllium illuminated with α particles. Finally he described the ongoing reaction as

$${}_{2}^{4}\alpha + {}_{4}^{9}Be \rightarrow {}_{6}^{12}C + {}_{0}^{1}n. \tag{2.8}$$

The uncharged particle in this equation was called neutron. The flux of free neutrons released by the reaction was about 10^0 n cm⁻²s⁻¹. Such a small number would prevent any scattering experiment.

2.2.1 Nuclear fission reactors

With the development of nuclear fission reactors in the 1940ies the situation changed. Using the fission reaction

$$^{235}U + ^{1}_{0}n \rightarrow \text{fission fragments} + 2.52 \times ^{1}_{0}n + 180\text{MeV}$$
 (2.9)

the first experimental reactors released about 10^7 n cm $^{-2}$ s $^{-1}$. Beside the investigation of the nuclear reaction, such a flux enabled the first scattering experiments with neutrons. In the following the thermal neutron flux increased dramatically until it saturated in the mid fifties. The still most powerful research reactor at the ILL became critical in 1974. The modern FRM II reactor has $0.5 \times$ the flux of the ILL, but the thermal reactor power is lower by a factor 0.33 due to special core design. Furthermore, the flux of cold neutrons (see Sec. 2.3) is more or less the same.

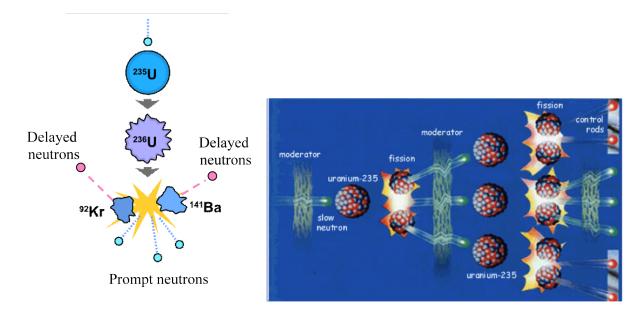


Fig. 2.2: Left) Schematic presentation of the fission process of ²³⁵U. Right) Controlled chain reaction in the nuclear reactor. Control rods reduce the number of slow neutrons to the amount just as necessary for the selfsustaining chain reaction. By the proper adjustment of the control rods position, the reaction may remains critical only with the inclusion of the delayed by a few seconds neutrons. From http://en.wikipedia.org/wiki/Nuclear_fission.

In the nuclear fission reaction eq. (2.9) a slow neutron is captured by an ²³⁵U nucleus, which then splits into two fragments releasing 2 or 3 prompt neutrons, which carry an energy of 1.29 MeV. Each of this instantaneously (within 10 ns) emitted neutrons can fission another nuclei so that each of them will emit another 2 to 3 neutrons. The process is called chain reaction If the mass of the fissile material is larger than the so called critical mass M_C the number of neutron will increase exponentially, leading to an uncontrollable reaction. If the mass of the fissile material is smaller than M_C the number of neutrons will decrease over time and the nuclear chain reaction stops. If you want to sustain the nuclear reaction for a long time it is necessary to control the neutron flux such that the number of neutrons that drive the chain reaction remains constant. The control of the reactor is possible, if the nuclear reaction is not only triggered by the prompt neutrons. The fission fragments are also highly excited nuclei and relax to their ground state by the emission of neutrons among other nuclear reactions. Concerning only the prompt neutrons, the reactor is operated below its critical mass M_C , but the delayed neutrons, which are comprised by the prompt neutrons, which are moderated in the cooling medium and the secondary neutrons from the fission fragments, sustain the chain reaction. The number of delayed neutrons is controlled by rods of neutron absorbing material (usually Boron), which can be inserted in the reactor core. Beside the control rods, which are used to steer the reactor, additional rods exist to fully stop the flux of neutrons and shut down the reactor.

With the development of the nuclear research reactors the thermal neutrons flux increased rapidly until it reached a flux $\Phi=10^{15}$ n/cm²/s at the end of the 1960ties. An increase in neutron flux goes simultaneously with an increase in the thermal power of the reactor. However, the installations for extracting the neutrons suffers strongly by heat and radiation damage. Therefore the development of more powerful research reactors has stopped with the design of ILL reactor. The modern FRM II reactor has a very compact reactor core, which provides half of the thermal neutron flux using only one third of reactor power as compared to the ILL.

2.2.2 Spallation neutron source

As an alternative to nuclear fission reactors neutrons can be released from the nucleus via spallation reactions. Here, high energy protons are accelerated onto a target made of a neutron rich material. Due to the large energy, the de Broglie wavelength

$$\lambda = \sqrt{\frac{h^2}{2mE}} \tag{2.10}$$

is so short, that the protons interacts with the single nucleons instead of the nucleus as a whole. The kicked nucleon may either leave the nucleus leading to an inter-nuclear cascade or may be scattered by other nucleons leading to an intra-nuclear cascade. However, as a result of stage 1 of the spallation process, the nucleus is in a highly excited state. In stage 2 this energy is released by evaporation of a whole particle zoo, including neutrons. The neutron yield per spallation event depends on the target material. For typical materials 20-50 neutrons are released per spallation event. The deposited heat depends on the target material, too, and is on the order of 20 to $50 \text{ MeV}/_0^1 \text{n}$.

Concerning safety, the spallation source can never run out of control as no chain reaction is running. Neutrons are only produced, as long as the protons are accelerated onto the target. Even better, this feature can be used to impose a precise time structure on the neutron spectrum.

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The spallation process happens on a time scale of 10^{-15} s. Therefore the length of the proton pulse determines the length of the neutron pulse. If one measures the time of flight of a neutron from the source to the detector at your instrument, the neutron velocity can be determined, as the flight path is also known. You will learn more about time-of-flight spectroscopy and diffraction in the remaining lectures. Among the spallation source on distinguishes so called long pulse

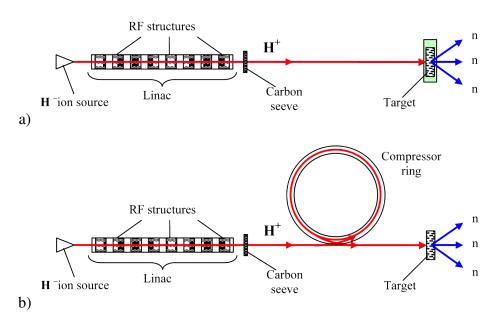


Fig. 2.3: Schematic of a long pulse and a short pulse spallation source.

spallation sources (LPSS) and short pulse spallation sources. Using a linear accelerator a proton bunch with a width of several ms can be tailored. If the neutron pulse should be shorter, the protons have to be compressed. This is done by feeding the protons from the Linear accelerator into a synchrotron. The next bunch is then feed in, when the former one has revolved once, to make a denser proton bunch. Using the compressor, the 1μ s duration pulses. While the latter type provides a higher peak flux, i. e., more neutrons in a short time intervall, the former type yields a significantly higher average neutron flux, in particular in the energy range that is typically used for diffraction experiments. Therefore certain experiments are better of at a SPSS, while the LPSS provides a more versatile spectrum and clearly is superior for 'slow' neutrons. The most powerful existing spallation source, the 1 MW SNS at Oak Ridge is a SPSS, while the planned ESS in Lund, Sweden, will be a LPSS with 5 MW power.

2.2.3 Comparison of reactor and spallation sources

Comparing the different sources, we have to consider a number of features:

Neutron Flux Nowadays reactor source still provide the highest average neutron flux. This flux is still higher as the flux at the 1.4 MW SPSS. The 5 MW spallation source wil

actually reach a similar average flux. However, for most experiments it is necessary, to select only a narrow range in energy or wavelength, respectively. At a pulsed source this can be done natively using time-of-flight monochromatization. Then not the average flux, but the peak flux, i. e., the flux during the proton pulse, counts. In that case, the monochromatic intensity at the spallation source can be higher.

Safety While the fissile material inside the reactor core of a research reactor is only a small fraction of the amount in a nuclear power plant, there is still a nuclear chain reaction ongoing, which in principle can run out of control. The spallation reaction is not possible without the operation of the accelerator and is therefore inherently safe.

As both source use nuclear reactions and create high energy particles, they both produce radioactive waste, which must be treated or stored after the operation of the facility. In case of the spallation source the waste has generally shorter life times.

Stability In fact, the operation of a proton accelerator is quite delicate. As already mentioned this makes the source very safe. On the other hand, sometimes it may also happen, that the proton beam is not available for quite some time during your allocated beam time. The neutron reactor runs usually very stable without interruption. Additionally the neutron flux is more stable at the reactor making it easier to compare individual measurements.

Technical feasibility The source neutron flux at a reactor could be increased only by an increase of the thermal power. There have been attempts to build a more powerful reactor in the US in the nineties, which has been abandoned for economical reasons. The heat removal from the core becomes extremely complex and also the radiation damage to the installations necessary for the extraction of the neutron is a severe issue. Therefore is unlikely, that higher power research reactors will be realized. At a spallation source the deposited heat For the SPSS exist similar arguments. The intense proton beam implants a large amount of heat in a very short time interval. Again the major problem is the removal of this heat. There seem to be a technological limit also for the short pulse spallation sources to increase their power far beyond the present state. For the long pulse spallation sources, the situation seems to be slightly relaxed. Since the heat is implanted during a longer time interval, the heat removal is facilitated. The 5 MW of power for the ESS could possibly increased up to 10 MW. There exist even estimates, that one could design a long pulse spallation target running at 20 MW. However, these are plans for the very far future, as already the ESS will be operational in the 2020ies only.

So far I have not considered the nuclear fusion reaction as a source for neutrons. Technologically this could be a technique at least as far in the future as a 20 MW spallation source. However, as seen from table 2.1 the deposited heat makes this reaction also a candidate for the over next generation of neutron sources.

2.3 How do we make free neutrons useful?

The neutrons as the are released from the nucleus have energies in the MeV range corresponding to a wavelength according to eq. (2.10) $\lambda \approx 10^{-5}$ Å. The energies we are interested in solid state physics, chemistry or biology rather range from the μ eV range for relaxation phenomena

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Reaction	Energy (GeV)	Neutron yield	Deposited heat (MeV)
	per event	per event	per neutron
(T,d) fusion		1	3
²³⁵ U fission		1	180
Pb spallation	1	20	23
²³⁸ U spallation	1	40	50

Table 2.1: *Comparison of neutron producing reactions*

to the eV range for the bonding of the valence electrons in molecules. One my also compare the energy scale to the corresponding temperatures via

$$E_{\text{therm}} = k_B T. (2.11)$$

From here we see that 1 meV is equivalent to a temperature of 11.6 K or vice versa 300 K are equivalent to 25.6 meV.

The distances we want to resolve in a neutron scattering experiment are on a atomic or molecular length scale and range from 1 Å to $1\mu m$ and therefore the neutrons should have comparable wavelength to have an appropriate resolution. If we use again the expression for the de Broglie wavelength eq. (2.10), we find that a neutron with an energy E=25.6 meV has a wavelength $\lambda=1.8$ Å, fulfilling both requirements simultaneously. This is also the reason, why neutron scattering is so versatile for studies of the dynamics of crystalline materials, because all atoms in a crystal show coherent motions due to their arrangement and bonding.

How can the neutrons now slowed down to the energies we are interested in? The best way is, if they collide elastically with other partners of much lower energy and spread this energy in a large volume (don't forget, that $1 \text{ MeV} = 1.6 \times 10^{-13} \text{ J}$). The energy loss per collision depends on the mass of the colliding partners: The highest energy transfer is achieved, if the mass of both partners is equal. Therefore 1H or 2H are the best partners, making water an ideal choice for the moderator. Since protons like to react with neutrons, the moderator often contains heavy water, i.e. D_2O , which has a smaller absorption cross section. For the FRM II the reactor core is surrounded by the heavy water tank. The outer area of the water tank is filled with light water, hence the flux of neutrons hitting the biological shielding outside the tank is already reduced.

Typically it takes several μ s to moderate the neutron to the temperature of the surrounding water. This process is therefore called thermalization. Within this time the neutron travel away from the reactor core, where they are produced. On the other hand, there is a finite probability for the absorption of a neutron, if the flight path inside the water is too long. The maximum of the thermal neutron flux density is displaced from the reactor core with the fuel element by 10 to 15 cm, as shown in Fig. 2.4 a).

For an experiment it is now of main interest to collect as many useful neutrons from the reactor, but not to get the fast neutrons or the Γ radiation that are created in the nuclear reactions into the experimental area. Therefore the beam tubes, as indicated in Fig. 2.4 b) don't face the reactor core, but tangentially look onto the maximum of the thermal flux distribution.

In the end of the thermalization process the neutrons are in thermal equilibrium with the sur-

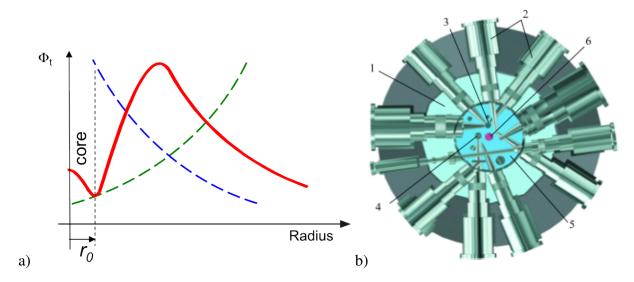


Fig. 2.4: a) Radial distribution of the thermal neutron flux density in the reactor vessel. The green line indicates the distribution, where the full thermalization is reached, the blue line indicates that the absorption decreases the neutron flux. b) Schematic of the reactor vessel of the FRM II showing the reactor core and the beam tubes extracting the neutrons to the experiments. The reactor tank with internal diameter approx. 5m is filled with light water (1). In the centre of the arrangement the reactor core is situated. The experimental installations as horizontal beam tubes (2), a cold (3) and a hot (4) neutron source are arranged in the heavy water tank (5) around the fuel element (6).

rounding medium. The energy distribution takes the form of the Maxwell distribution:

$$\Phi(E) = \frac{2\sqrt{E}}{\sqrt{\pi k_b^3 T_M}} \exp{-\frac{E}{k_b T_M}}$$
(2.12)

The neutrons are commonly classified for certain energy and wavelength ranges according to the position of the maximum of the Maxwell distribution for a given moderator temperature T_M :

	Energy range(meV)	Wavelength range (Å)
Ultra cold	E < 0.0005	$\lambda > 400$
Very cold	0.0005 < E < 0.005	$40 < \lambda < 400$
Cold	0.05 < E < 5	$4 < \lambda < 40$
Thermal	5 < E < 100	$0.9 < \lambda < 4$
Hot	100 < E < 1000	$0.3 < \lambda < 0.9$

To access the respective energy range the moderator should again effectively moderate the neutrons but also be transparent for the neutrons. A liquid hydrogen vessel fulfills the requirements for cold neutrons. A more effective but also more difficult technique employs solid methan as a moderator in a cold source. A carbon block heated to a temperature above 1000 K is used in reactors to provide an intense source of hot neutrons . In Fig. 2.5 the spectra for the different moderator temperatures show clearly, that the maximum is shifted towards shorter wavelength, when the temperature is increased. In a spallation source usually a different route is used to yield an intense beam of hot neutrons: The moderator is made thin enough to not fully moderate the neutrons. Therefore epithermal neutrons still exist in the energy distribution of the

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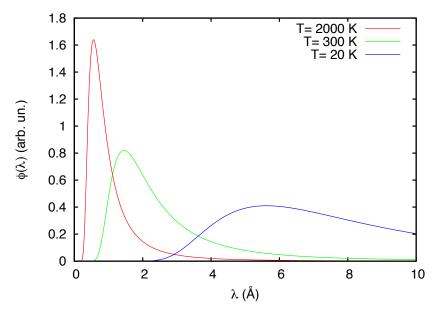


Fig. 2.5: Neutron wavelength distribution for different moderator temperatures. Cold spectrum, $T=20\,$ K, blue line, thermal spectrum, $T=300\,$ K, green line, hot spectrum, $T=2000\,$ K, red line.

source. The time structure of the source might then be used to discriminate the eventually increased background.

2.4 How do we bring the neutrons to the experiment?

The angular distribution of the thermal flux distribution at the end of the thermalization process is fully isotropic. To calculate the flux at the exit of a beam tube approximately one has to divide the thermal flux at the maximum by the surface area of the sphere with the respective radius, in the case of the FRM II 2.5 m, see Fig. 2.4 b). Already at this distance the flux is reduced already by 6 orders of magnitude. If the distance required to build an actual instrument is added, the flux is lowered by 8 orders of magnitude.

To overcome this problem, neutron guides are used. These consist of 4 neutron mirrors, enclosing the flight path of a neutron. The principle of the neutron guide is similar to light wave guides: External total reflections prevents the neutrons from leaving the guide and they are transported to the end of the guide. In the case of the light wave guide, the fibre has a larger index of refraction than the surrounding air, giving rise to typical critical angles $\theta_C \approx 45^\circ$. For the neutron guide, the vacuum inside has a larger index of refraction and the critical angle is given by

$$\theta_C = \lambda \sqrt{\frac{2\rho b_c}{\pi}} \tag{2.13}$$

with the particle density ρ and the coherent scattering length b_c . The element with the largest critical angle is Nickel and for the element the critical angle can simply be approximated $\theta_C = 0.1^{\circ} \text{Å}^{-1}$. If we install such a neutron guide behind a beam tube, all neutrons, that impinge on the Ni surface under a shallower angle than the critical angle, will be guided to the instrument. If we calculate for $\lambda = 5$ Å neutrons we loose only 4 orders of magnitude independent of the distance from the reactor core. Hence such a neutron guide can be used to provide more space for instruments by going further away from the reactor. Nowadays so called supermirrors consisting of thin layers of e.g. Ni and Ti increase the critical angle of Ni by a factor up to

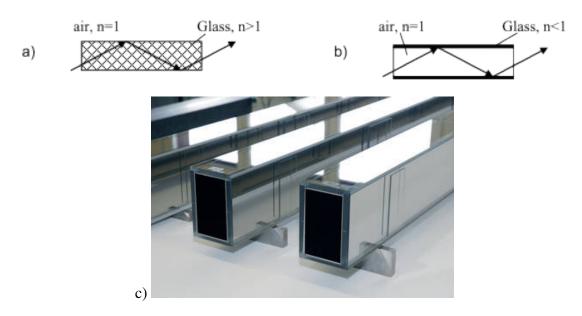


Fig. 2.6: a) Schematic of a light wave guide. External total reflection occurs, because the fibre is optically denser than the air. b) Schematic of a neutron guide. Total reflection occurs, because the index of refraction of the mirror coating is smaller than 1. c) Picture of a super mirror neutron guide, taken from www.swissneutronics.ch.

7. In that case it becomes possible to build neutron guides not only for cold neutrons but also for thermal neutrons. Furthermore complex focusing optics can be realized by neutron guides to increase the number of useful neutrons at the spectrometer and simultaneously keep the background low

At least as important as gaining space is the fact, that the direct sight from the instrument onto the reactor core can be omitted. Fast neutrons and Γ radiation is leaving through the holes for the neutron beamlines. They go mainly in a straight line from where they have been created, because their scattering cross section is very small. These particles contribute mainly to the radiation background around the instruments. They can of course also contribute to the background in your detector. The particles are kept away from users and detectors by massive shielding, containing a lot of concrete (for fast neutrons) or lead (for Γ radiation). If such a neutron guide is bend with a large radius, the direct line of sight hits the wall of the neutron guide and the background of the instrument can be further suppressed. Of course your shielding must then be strongest in the direct line of sight.

2.5 How do we detect neutrons?

On of the strongest advantages of the neutrons is their neutrality. It allows to probe deeply into matter. On the other hand, this makes the detection of the neutrons difficult, as it penetrates large volumes of matter without interaction. Luckily there exists a hand full of isotopes that have a large absorption cross section for thermal or cold neutrons, such as ³He ¹⁰B, Gd or ²³⁵U. The nuclear reactions create charged particles, which can be analyzed by interaction with the electric fields. Since the absorption cross section in the thermal to cold energy range increases more or less linearly with the wavelength, the detection of cold neutrons is more effective than

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the absorption of thermal neutrons

One type of detector is the gas proportional counter filled either with 3 He gas or gaseous 10 BF $_3$. The absorption process releases a certain number of photons, which create secondary electrons by Compton scattering or the photo effect, or high energetic charged particles. The particles are accelerated onto the cathode or anode according to their charge and the resulting current can be related to the neutron absorption event. A refinement of the apparatus allows also the localization of the absorption event yielding a position sensitive detector. Features of the gas proportional counter are a high detection probability, which can be tuned by the filling pressure, and a low sensitivity to Γ radiation. Disadvantages are a limited count rate before the detector saturates and a position sensitivity > 1 cm.

A szintillation detector provides a much higher spatial resolution. Here the neutron absorption at a neutron absorber embedded in the solid szintillation material yields Γ photons that are detected by the photo electric effect. This detectors provides a higher spatial and timing resolution but has also a larger Γ sensitivity.

2.6 The take home messages

Today, intense neutron beams are available a nuclear research reactors and spallation sources. Reactors deliver a very stable continuous beam, while spallation sources provide a very high peak flux that can be effectively used by time-of-flight methods.

Neutrons are extremely useful for condensed matter research, if the wavelength and kinetic energy match the length scale and energy scale of e.g. magnetic compounds, polymers or biological samples. These is realized by moderating the fast neutrons released in the nuclear reaction in a volume containing a lot light elements, e.g. water for thermal neutrons, liquid hydrogen or solid methan for cold neutrons or heated graphite for hot neutrons.

Neutron guides are used to transport neutrons with only small losses quite far away from the actual neutron source. This gives more space for instruments, improves the background conditions and may even be used to tailor the neutron beam properties using complex optics similar to light optics.

Further reading

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Exercises

E2.1 How are neutrons characterized?*

Write down the kinetic energy of a free neutron as a function of its momentum! What is the velocity in ms^{-1} and energy in meV of neutrons with a wavelength $\lambda = 1, 1.8, 5$ Å, respectively?

$$m_n = 1.675 \times 10^{-27} \text{kg}$$

 $h = 6.626 \times 10^{-34} \text{Js}$
 $e = 1.602 \times 10^{-19} \text{As}$

E2.2 How many neutrons are produced?**

Calculate the neutron flux density of a 20 MW reactor, assuming that the flux maximum is displaced 10 cm from a point-like reactor core! What would be the flux density of a hypothetical spallation source with the same thermal power?

E2.3 How do the neutrons come to your experiment?

Why is the neutron flux reduced, when you build the diffractometer/spectrometer at larger distance without a neutron transport system? When is it advantageous to have the instrument close to the neutron source? What reasons can you imagine to separate the instrument from the neutron source?