Determination of effective temperatures through affordable concurrent techniques in CANS

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Abstract. Experimental techniques involving epithermal neutrons can be used to study the kinetic energies of the atoms that make up the sample material. From the point of view of Nuclear Engineering, this is the range of energies that is necessarily traversed in the moderation process. From the point of view of applied physics, it is an essential range for the design of neutron sources associated with compact accelerators. It is also worth mentioning that experimental access to this energy range is a distinctive feature of accelerator-based sources as opposed to reactor-based sources. The atoms that compose the matter perform motions characterised by a kinetic energy, determined not only by the temperature of the medium, but also by interactions resulting in vibrational modes. We can thus define a parameter that we call effective temperature, linked to this effect. The knowledge of the densities of the vibrational states allows its evaluation, so that spectroscopic techniques aimed at its measurement allow an indirect assessment of it. Yet, there are techniques that allow direct experimental access to these quantities which are Deep Inelastic Neutron Scattering (DINS) and Neutron Transmission (NT). The VESUVIO spectrometer (ISIS, UK) allows both techniques to be performed simultaneously in the same experiment. Although ISIS is a large neutron source, such experiments were previously carried out at the defunct LINAC in Bariloche (Argentina), making these techniques affordable for small sources. In this work we show this capability through combined DINS and transmission experiments in a molecular liquid. We also discuss different prospects for future use, extending the possibilities to other techniques.

1 The effective temperature

Theoretical and experimental activity with thermal and epithermal neutrons, stimulate the work and development of models describing the interaction of neutrons with matter, intimately linked to its dynamics and structure. The study of the dynamics is particularly relevant, as it has direct implications for the study of neutron moderators. For the formulation of models involving thermal neutrons, a fairly complete description of the

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dynamics is necessary, whereas in the epithermal regime approximations with few parameters are usually sufficient. This is because in this regime the short collision time approximation is valid [1]. Under these conditions, the system can be described as an effective gas, i.e. a gas in which the temperature is an effective quantity that is a function not only of the thermodynamic temperature, but also of the energies characterising the internal degrees of freedom of the system [2].

We will focus on the incoherent component of the cross section. To illustrate with a monoatomic system, the double differential cross section for a gaseous system is

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k}{k_0}\frac{1}{2\pi\hbar}b^2 S(Q,\omega) \quad , \tag{1}$$

where k_0 and k are respectively the wave numbers of the incident and outgoing neutrons, and b the atom scattering length. The scattering law for an ideal gas is

$$S(Q,\omega) = \sqrt{\frac{M}{2\pi Q^2 k_B T}} \exp\left[-\frac{\left(\hbar\omega - \frac{\hbar^2 Q^2}{2M}\right)^2}{2\hbar^2 Q^2 \frac{k_B T}{M}}\right], \tag{2}$$

where M is the atom mass $\hbar\omega$ is the energy transfer, $\hbar Q$ the momentum transfer, k_B is Boltzmann's constant and T the thermodynamic temperature. It is useful to write also the intermediate scattering function [3]

$$\chi(Q,t) = \exp\left[\frac{\hbar Q^2}{2M}\left(it - \frac{k_B T}{\hbar}t^2\right)\right]. \tag{3}$$

For a general system characterized by a frequency spectrum $Z(\omega)$, Eq. (3) acquires the form

$$\chi(Q,t) = \exp\left[\frac{\hbar Q^2}{2M}\left(\gamma(t) - \gamma(0)\right)\right]. \tag{4}$$

The connection between Eqs. (3) and (4) as well as an exhaustive treatment of correlation functions, can be found in Ref. [1].

Although a rigorous treatment can be found in Ref. [1], to conceptualise, we will concentrate on a system whose dynamics is described by an Einstein oscillator of frequency ω_0. In this case

$$\gamma(t) = \frac{1}{\omega_0}\left[\left(n(\omega_0)+1\right)\right) e^{i\omega_0 t} + \left(n(\omega_0)\right) e^{-i\omega_0 t}\right], \tag{5}$$

where $n(\omega_0) = (\exp(\frac{\hbar\omega_0}{k_B T})-1)^{-1}$ is the Bose occupation number. For epithermal neutrons, the short collision time approximation is applicable [1]. Keeping up to the second order time expansion in (5)

$$\gamma(t) \underset{t\to 0}{\longrightarrow} \gamma(0) + it - (n(\omega_0) + \tfrac{1}{2})\omega_0 t^2, \tag{6}$$

and substituting in (4) we obtain

$$\chi(Q,t) = \exp\left[\frac{\hbar Q^2}{2M}\left(it - \left(n(\omega_0) + \frac{1}{2}\right)\omega_0 t^2\right)\right] . \tag{7}$$

This expression can be assimilated to Eq. (3) for the ideal gas, where the thermodynamic temperature T, is replaced by an effective temperature, such that

$$k_B T_{\text{eff}} = \hbar\omega_0 \left(n(\omega_0) + \frac{1}{2}\right) . \tag{8}$$

This serves to introduce the concept of effective temperature. In the limit $k_B T \gg \hbar\omega_0$, it is satisfied that $n(\omega_0) + \frac{1}{2} \simeq \frac{k_B T}{\hbar\omega_0}$, so $T_{\text{eff}} = T$.

For the general case of a system characterized by a frequency spectrum $Z(\omega)$, the expression is [1]

$$k_B T_{\text{eff}} = \int \hbar\omega Z(\omega)\left(n(\omega)+\tfrac{1}{2}\right)d\omega \,. \tag{9}$$

With these definitions we see, in general, that the effective temperature differs and has larger values than the thermodynamic temperature. The ideal gas model with an effective temperature is a good description of the dynamics of a system at short collision times, approximation valid with epithermal neutrons.

The knowledge of effective temperatures is relevant in models used in numerical simulations employed for example in Nuclear Engineering and accelerator target-moderator-reflector design for the production of neutron beam. In such cases, when dealing with epithermal neutrons, ideal gas approximations are usually used for the dynamics of the atoms involved in the materials, which is a fast and effective approach, provided that the temperatures of these gases are the effective rather than the thermodynamic ones. That is why the theoretical and experimental knowledge of the effective temperature is so relevant, as will be stressed in this paper.

2 Experimental techniques

We will refer to the experimental techniques that have direct access to the effective temperature and to the basic expressions that link the experimental magnitudes to this parameter. The techniques are Deep Inelastic Neutron Scattering (DINS) and Neutron Transmission (NT). Figure 1 shows the layout of the VESUVIO spectrometer (ISIS Neutron and Muon Source, Rutherford Appleton Laboratory, UK). In this facility it is possible to perform measurements with both techniques simultaneously. The detectors located in backward and forward scattering positions are used for DINS, while the S2 monitor is used for transmission.

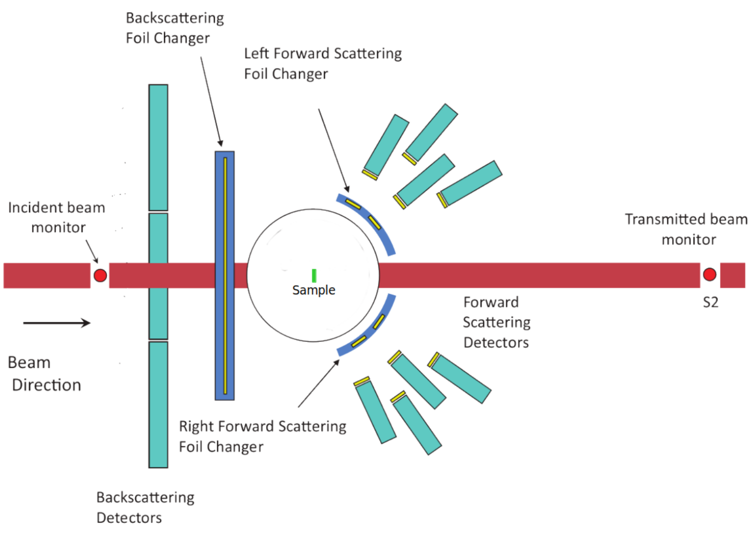

Fig. 1. Schematic setup of the VESUVIO spectrometer (ISIS Neutron and Muon Source, UK).

For the application of the DINS technique, gold analyser filters foils are used and the scattered neutron spectra (as a function of the time of flight) are measured alternately with and without filter. The difference between the two spectra determines the so-called Compton neutron profile. A detailed explanation of the technique can be found in Ref. [4].

The basic expression that links the neutron Compton profile with the experimental basic parameters is described in Ref. [2], and is as follows:

$$c(t) = \int_{\substack{\mathrm{E_{inf}} \\ t=\mathrm{const}}}^{\infty} \mathrm{d}E_0\, \Phi(E_0)\, \sigma(E_0, E, \theta_\mathrm{i})\, \varepsilon(E) \left[1 - \mathrm{e}^{-nd\sigma_\mathrm{F}(E)}\right] \left|\frac{\partial E}{\partial t}\right| \Delta\Omega \quad (10)$$

where $\mathrm{E_0}$ and E are the incident and scattered neutron energies respectively, and θ the scattering angle, $\Phi(E_0)$ is the incident neutron flux, $\varepsilon(E)$ is the detector efficiency, and $\left[1 - \mathrm{e}^{-nd\sigma_\mathrm{F}(E)}\right]$ corresponds to the absorption probability of the resonant filter, characterized by a number density n, a thickness d, and a total cross section $\sigma_\mathrm{F}(E)$. $\Delta\Omega$ is the solid angle subtended by the detector. The lower limit of integration $\mathrm{E_{inf}}$ is determined by kinematic conditions. The term $\sigma(E_0, E, \theta)$ is the double-differential cross section of the sample, which contains the effective temperature to study. For a system containing different kind of atoms its expression is

$$\sigma(E_0, E, \theta) = \sqrt{\frac{E}{E_0}} \sum_A x_A \sigma_{b,A}\, S_A(Q, \omega) \quad (11)$$

where the sum extends to all atoms and $S_A(Q, \omega)$ has the gas expression (2) in the short collision time approximation, x_A is the stoichiometric number of the atom A in the molecular composition and $\sigma_{b,A}$ is its bound-atom scattering cross section. Equation (10) does not involve the use of a resolution function, since the complete gold foil absorption cross section as a function of the energy is employed.

Concerning the NT measurements, they are made with a detector placed on the incident beam line. Alternative measurements of the incident ($I_0(E_0)$) and transmitted ($I(E_0)$) neutron spectra are performed. The attenuation law relates these measurement with the total cross section σ_tot

$$\frac{I(E_0)}{I_0(E_0)} = \exp(-nd\sigma_\mathrm{tot}(E_0)) \quad , \quad (12)$$

where n is the number density of scattering units (atoms or molecules) and d is the sample thickness. In the epithermal neutron region the analytic expression for σ_tot is [1]

$$\sigma_\mathrm{tot}(E_0) = \sum_i x_i \sigma_{b,i} \left(\frac{A_i}{A_i + 1}\right)^2 \left[1 + \frac{k_\mathrm{B} T_{\mathrm{eff},i}}{2 A_i E_0}\right], \quad (13)$$

where the sum extends to the different atoms in the scattering unit of atomic mass A_i which vibrate according to an effective temperature $T_\mathrm{eff,i}$, with a bound atom cross section $\sigma_{b,i}$ and with a number of atoms of type i per scattering unit x_i.

3 Results

As a case study we will show results for solid triphenylmethane at 22 K obtained at VESUVIO [5]. Figure 2 shows the neutron Compton profile determined for a detector placed at an angle of θ = 54.7° with respect to the incident beam. The previous data treatment process includes the evaluation of the multiple scattering components and the evaluation of the attenuation coefficient by numerical simulations described in Ref [6]. The spectrum corrected with this procedure is directly comparable with the experimental data.

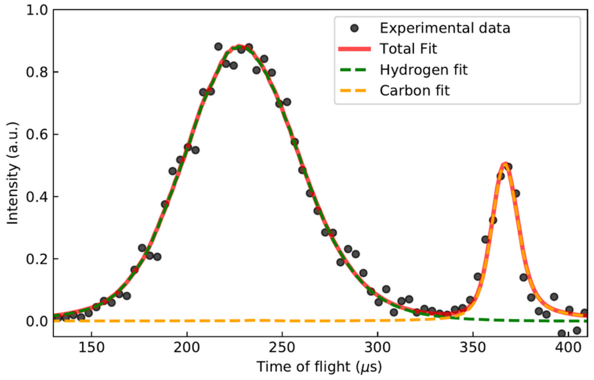

Fig. 2. Neutron Compton Profile of triphenylmethane at 22 K recorded at an angle θ = 54.7° with respect to the incident beam. Fitted values are $T_{eff,H}$ = (92.9 ± 3.3) meV and $T_{eff,C}$ = (28.5 ± 6.9) meV.

Employing Eq. (10) we fitted the effective temperatures which are $T_{eff,H}$ = (92.9 ± 3.3) meV and $T_{eff,C}$ = (28.5 ± 6.9) meV. The determination of the effective temperatures involved the analysis of the individual spectra of the detectors placed at different angles, from which a mean and a confidence interval were obtained. Vibrational frequency spectra calculations were performed with the density functional theory (DFT) method. The effective temperatures evaluated from this method were $T_{eff,H}$ = 98.3 meV and $T_{eff,C}$ = 51.5 meV. At this point it is worth noting that the thermodynamic temperature is 1.9 meV, which confirms the above statement about the differences with the effective temperature. Differences between DFT and the experimental results are attributable to the fact that the calculations were performed on a monocrystalline system, which does not take into account possible distortions due to the polycrystallinity of the real systems. Anharmonic effects were also not taken into account, which may further explain the differences.

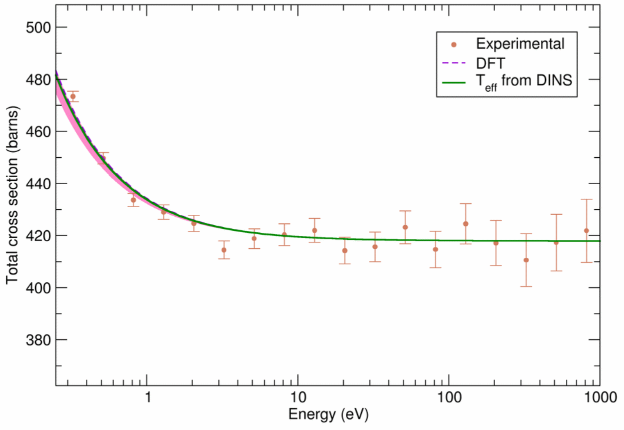

Fig. 3. Total cross section (in barns per molecule) for triphenylmethane at 22 K in the epithermal neutron energy region. Data are shown with orange points with error bars. The green line shows the calculation made with Eq. (13) employing the fitted effective temperatures of hydrogen and carbon with DINS. The dashed line shows the calculation performed with the effective temperatures calculated with DFT. The pink shaded area shows the variation of the calculation performed within limits of the confidence interval of the effective temperatures determined by DINS.

Figure 3 shows the total cross section of triphenylmethane at 22 K as determined from the S2 monitor [5,7] shown in Fig. 1, employing Eq. (12). The total cross sections employing the effective temperatures determined by DINS and calculated by DFT are also shown. The pink shaded area shows the variation within the confidence interval limits of the effective temperatures determined by DINS on different detectors.

4 Conclusions

We have shown and analysed the effective temperature, a useful parameter in modelling the interaction of epithermal neutrons with matter. In the context of CANS, the interest of this work is that these techniques are within the reach of small facilities and can be implemented without difficulty. It is noteworthy that such a facility operated at the LINAC in Bariloche (Argentina) until its decommissioning [8].

It is also worth mentioning other applications of such a facility that have not been analysed in this work, but which are developments proposed by the Bariloche and the VESUVIO groups.

- Total cross sections: They can be measured by transmission at a wide range of energies from subthermal to epithermal, although only the latter was shown in this work [7].
- Diffraction: If we analyse the thermal part of the spectra, Bragg peaks can be observed in crystalline systems as shown by Krzystyniak et al. [9]
- Non-destructive mass spectrometry: It can be performed, since the DINS spectra have separate peaks according to the different atomic masses [10].
- Neutron resonance capture analysis: When the sample has neutron-absorbing nuclei, prompt gamma analysis can be performed as a function of the energy of the incident neutrons determined by time of flight [11,12].

It should be noted that all these techniques can be developed during the same experiment by simply analysing the data from the different detectors at different flight times, which entails developments in analysis methods. Therefore, we consider that the implementation of a detector station such as the one described offers multiple possibilities of development and provides useful information for the knowledge of neutron-matter interaction. Thus it is an interesting application to be taken into account in the development of the use of CANS.

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