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Calculation of neutron cross sections and thermalization parameters for molecular gases using a synthetic scattering function. I

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Based on a synthetic scattering function describing the interaction of thermal neutrons with molecular gases, we developed analytical expressions for cross sections of interest in reactor-physics calculations; these are the zero- and first-order angular moments of the double-differential cross section, $\sigma_0(E_0,E)$ and $\sigma_1(E_0,E)$, respectively, and the total scattering cross section $\sigma(E_0)$. The formulas presented here allow then a very fast and reliable evaluation of thermalization parameters and transport coefficients at any desired temperature of the moderating media. One of the main applications of this study is the generation of (thermal) nuclear data libraries for reactor codes, using a minimum set of input information. In the following paper, calculations based on our formulas are compared with experimental data and other theories for some commonly used moderators under different physical conditions.

I. INTRODUCTION

Reactor-physics calculations involving thermal neutron energies demand the knowledge of reliable cross-section information. In fact, this kind of requirement together with the fundamental interest in the atomic motion in solids and liquids were the main motivations for many slow-neutron inelastic scattering studies. Furthermore, much of that work concentrated particularly on hydrogenous molecular systems due to their importance as reactor moderators.

A central quantity to describe the interaction of thermal neutrons with condensed matter is the Van Hove scattering function[1,2] $S(\mathbf{Q},\omega)$, as it embodies all the structural and dynamical properties of the scattering system. On the experimental side, a great deal of effort was dedicated to double-differential cross-section measurements but, even though a large portion of the relevant features in the ω-Q plane can thus be revealed, there will always be a particular problem for which the experimental data do not cover exactly the required material and physical conditions.

Consequently, many models were developed to describe neutron scattering from molecules as approximations to the essentially exact formalism of Zemach and Glauber,[3] with the aim of making the expressions more amenable for calculations. Those models and their range of applicability have been extensively discussed in the literature.[4–6] As a result of these studies, large computer codes were written to calculate a number of quantities which are relevant to the problem of neutron thermalization.[7,8] It became then possible to perform detailed calculations of scattering kernels and thermal neutron spectra in a moderating media, although they were always computer-time consuming, even for a simple system configuration.

Most present-day lattice codes[9] are supplied with a nuclear data library containing group constants for fast and thermal neutron energies, the data being usually generated from an evaluated library like the Evaluated Nuclear Data File (ENDF/B).[10,11] For practical reasons, those cross-section libraries involve a certain number of materials with the corresponding data at a few temperatures for the thermal groups. Although it is possible to produce an extended version of a library to solve a specific problem,[12] rather long and expensive procedures are normally required.

In this work we present some general results—which are relevant to reactor physics calculations—derived from a synthetic scattering function $T(Q,\omega;E_0)$ which has been developed to describe the interaction of slow neutrons with molecular gases.[13] Unlike the dynamic structure factor $S(\mathbf{Q},\omega)$, the synthetic function does not contain a detailed description of the atomic motions in the molecular units nor correlation between pairs, but rather it is intended to reproduce satisfactorily some integral properties of $S(\mathbf{Q},\omega)$ (the self-component). We have exploited the very simple form of $T(Q,\omega;E_0)$ to derive analytic expressions for scattering kernels and total cross sections. Some preliminary results were already presented with the introduction of the synthetic model, and they have encouraged us to produce a consistent set of formulas which allow a fast and reliable evaluation of quantities of interest in neutron thermalization problems. Of course, our final objective is the generation of cross-section libraries for molecular systems at any desired temperature to feed a reactor computer code. Only the incoherent components of the neutron-scattering process are considered, this approximation being usually a good one for this kind of calculations.

In the following paper[14] we make use of the formulas which are here presented to perform a rather wide range of calculations for some hydrogenous molecules (H_2O, D_2O, C_6H_6), and these results are there compared with experimental data as well as with other theories.

II. THE MODEL

The synthetic model has been fully described previously[13] and therefore only a brief account of its supporting arguments and characteristic features will be given here.

The scattering system is considered to be an ideal molecular gas at temperature T, which means that the translational motion of the molecules' center of mass is taken as that corresponding to a free particle.

Although the motion of the molecular unit may be severely hindered in a real system due to the presence of its neighbors, it is only for very slow neutrons that this collisional regime will become dominant. Consequently, we consider a low-energy limit for the incident neutrons such that the experimental time scale is shorter than that characteristic of diffusive motion. The attainment of this condition clearly depends on the particular system under study, but for most real cases it implies neutron energies (a few meV) which are outside the region of main interest in reactor-physics calculations.

The internal degrees of freedom of the molecule are assumed to be not coupled. This is a first approximation to the real situation which is valid as long as the amplitudes of the atomic oscillations around their equilibrium positions are small compared to the interatomic distances in the molecule. Each of the λ internal modes is represented by an Einstein oscillator with angular frequency ω_λ and effective mass M_λ.

From the requirement that the free-atom cross section be approached at high neutron energies (large compared with $k_B T$ and the largest $\hbar\omega_\lambda$), a normalization condition for these masses is obtained:

$$\frac{1}{M_{\mathrm{mol}}}+\sum_\lambda \frac{1}{M_\lambda}=\frac{1}{M}\ , \tag{1}$$

where M_{mol} is the molecular mass and M is the mass of the nucleus under consideration (in neutron mass units). An additional constraint is imposed on the rotational mass M_R, namely

$$\frac{1}{M_{\mathrm{mol}}}+\frac{1}{M_R}=\frac{1}{\mathcal{M}}\ , \tag{2}$$

where $\mathcal{M}$ is the (spherically averaged) tensorial mass introduced by Sachs and Teller[15] to describe the combined effect of translations and rotations. The remaining vibrational masses are determined by the relative weights of the related amplitude vectors.

Correspondingly, at epithermal neutron energies the scattering nuclei are viewed as possessing a kinetic energy associated with a temperature $\bar{T}$ given by

$$k_B\bar{T}=k_B T+\sum_\lambda \frac{M}{M_\lambda}(E_\lambda-k_B T)\ , \tag{3}$$

where E_λ is the mean energy of the λ oscillator.

From the analysis of the forms that $S(\mathbf{Q},\omega)$ takes for small and large energy transfer $\hbar\omega$ in the scattering process, a function $T(Q,\omega;E_0)$ is proposed which uses the incident neutron energy E_0 as a variation parameter. The main characteristics of the molecular dynamics are then retained through the introduction of an effective mass, temperature, and vibrational factors. This is achieved by the use of the Krieger-Nelkin[16] procedure for orientational averages, and by the introduction of switching functions P_λ defined by

$$P_\lambda(E_0)=\frac{\exp\left[-\left[\dfrac{E_0-\hbar\omega_\lambda}{\hbar\sigma_\lambda}\right]^2\right]+\sqrt{\pi}\dfrac{\hbar\omega_\lambda}{\hbar\sigma_\lambda}\left[1+\mathrm{erf}\left[\dfrac{\hbar\omega_\lambda-E_0}{\hbar\sigma_\lambda}\right]\right]}{\exp\left[-\left[\dfrac{\hbar\omega_\lambda}{\hbar\sigma_\lambda}\right]^2\right]+\sqrt{\pi}\dfrac{\hbar\omega_\lambda}{\hbar\sigma_\lambda}\left[1+\mathrm{erf}\left[\dfrac{\hbar\omega_\lambda}{\hbar\sigma_\lambda}\right]\right]}\ , \tag{4}$$

where the quantities σ_λ are representatives of the width of the frequency spectrum in the vicinity of ω_λ.

Every P_λ tends to zero as the corresponding mode λ becomes fully excited from the point of view of the collision process, that is when a quasiclassical treatment is applicable. At the other end, the value of P_λ is 1 if the neutron cannot excite any of the λ-oscillator energy levels. At intermediate energies, the variation of P_λ should depend on the shape of that part of the molecular frequency spectrum associated with the mode λ. The expression given by Eq. (4) meets the above requirements and its supporting arguments are discussed in Ref. 13; its dependence on the temperature of the system is dictated by the shifting and broadening of the corresponding part of the frequency spectrum.[17]

An effective mass for the nucleus under consideration is defined by

$$\frac{1}{\mu}=\frac{1}{M_{\mathrm{mol}}}+\sum_\lambda \frac{(1-P_\lambda)}{M_\lambda}=\frac{1}{M}-\sum_\lambda \frac{P_\lambda}{M_\lambda}\ . \tag{5}$$

In this way, μ takes values ranging from M_{mol} to M, according to the state of excitation of the different internal modes.

By requiring consistency between the first and second moments of the scattering function, an expression for the effective temperature τ is obtained:

$$\frac{k_B\tau}{\mu}=\frac{k_B T}{M_{\mathrm{mol}}}+\sum_\lambda \frac{(1-P_\lambda)}{M_\lambda}E_\lambda=\frac{k_B\bar{T}}{M}-\sum_\lambda \frac{P_\lambda}{M_\lambda}E_\lambda\ , \tag{6}$$

where $k_B\bar{T}$ is given by Eq. (3). Clearly τ tends to T, the actual system temperature, or $\bar{T}$, the "free-atom temperature," according to whether all P_λ are 1 or zero, respectively.

Finally, an effective Debye-Waller factor Γ is defined by

$$\Gamma=\sum_\lambda \frac{P_\lambda}{M_\lambda}\frac{(2n_\lambda+1)}{\hbar\omega_\lambda}\ , \tag{7}$$

where n_λ is the thermally averaged occupation number of the λ mode.

With the above definitions, the atomic synthetic scattering function which describes a scattering process with energy and momentum exchange $\hbar\omega$ and $\hbar Q$, respectively, is written as

$$T(Q,\omega;E_0)=S_{\mu,\tau}(Q,\omega)\exp\left[-\Gamma\frac{\hbar^2Q^2}{2}\right]+C_{\mu,\tau}(Q,\omega)\ . \tag{8}$$

In this expression $S_{\mu,\tau}(Q,\omega)$ denotes the scattering function for an ideal gas of particles of mass μ at temperature τ:

$$S_{\mu,\tau}(Q,\omega)=(2\pi\hbar^2Q^2\mu^{-1}k_B\tau)^{-1/2}\times\exp\left[-\frac{(\hbar\omega-\hbar^2Q^2/2\mu)^2}{2\hbar^2Q^2\mu^{-1}k_B\tau}\right], \tag{9}$$

whereas $C_{\mu,\tau}(Q,\omega)$ is a correction term given by

$$C_{\mu,\tau}(Q,\omega)=\frac{1}{2}\sum_\lambda p_\lambda\left[n_\lambda\hbar^2Q_+^2S_{\mu,\tau}(Q_+,\omega_+)\exp\left[-\Gamma\frac{\hbar^2Q_+^2}{2}\right]+(1+n_\lambda)\hbar^2Q_-^2S_{\mu,\tau}(Q_-,\omega_-)\exp\left[-\Gamma\frac{\hbar^2Q_-^2}{2}\right]\right], \tag{10}$$

with $p_\lambda\equiv P_\lambda/(M_\lambda\hbar\omega_\lambda)$.

Here, $Q_\pm$ is the modulus of the scattering vector corresponding to a fictitious incident energy $E_{0,\lambda}^{\pm}=E_0\pm\hbar\omega_\lambda$. This correction term accounts for one-phonon processes which may be operative for those cases of thermal- or collision-induced excitations but with a neutron energy not high enough to allow a quasiclassical treatment of the corresponding mode.

With the notation

$$S_{\mu,\tau}^{KN}(Q,\omega)=S_{\mu,\tau}(Q,\omega)\exp\left[-\Gamma\frac{\hbar^2Q^2}{2}\right], \tag{11}$$

one finally obtains

$$T(Q,\omega;E_0)=S_{\mu,\tau}^{KN}(Q,\omega)-\sum_\lambda p_\lambda\left[n_\lambda\frac{\partial}{\partial\Gamma}S_{\mu,\tau}^{KN}(Q_+,\omega_+)+(1+n_\lambda)\frac{\partial}{\partial\Gamma}S_{\mu,\tau}^{KN}(Q_-,\omega_-)\right]. \tag{12}$$

This is the mathematical expression of the synthetic model. It does not pretend to be a real scattering function model for molecules, insofar as the full dynamics of the atomic motion is not accounted for in a detailed manner and also because only the incoherent contribution to the scattering process is considered. In Sec. III, however, we take advantage of its formal simplicity to derive analytical expressions for some magnitudes of interest in reactor physics.

Work is in progress involving applications of the synthetic model to other fields, examples of which are the evaluation of inelasticity corrections in neutron diffraction work on molecular liquids[18] and the optimization of neutron production and time response of moderators used in pulsed neutron sources.[19]

III. EVALUATION OF CROSS SECTIONS

The most basic magnitude which is experimentally accessible is the double-differential cross section. For the scattering of an unpolarized beam of neutrons from a molecular system, it is given by

$$\frac{d^2\sigma}{d\Omega\,dE}=\frac{k}{k_0}\sum_\nu\frac{\sigma_\nu}{4\pi}N_\nu T_\nu(Q,\omega;E_0)\ , \tag{13}$$

where k_0 and k denote the (modulus of) incident and scattered neutron wave vectors, respectively, ν runs over the species of nuclide in the same environment, each with a number N_ν of them and with a bound scattering cross section σ_ν.

This magnitude has been[20] and continues to be[17,21] a valuable source of information on the dynamics of condensed systems. However, from the point of view of neutron thermalization studies, the measurement of double-differential cross sections was mainly aimed at the determination of a continuous frequency spectrum according to the Egelstaff extrapolation method.[2] This quantity or its Fourier transform, the (self-) velocity-velocity correlation function, is related to the width function in the frame of the Gaussian approximation,[22] so that its knowledge permits us to obtain the scattering law by numerical Fourier transformation of the intermediate scattering function.[7] Although well supported on physical grounds, this is usually a rather lengthy and expensive procedure and furthermore, the information thus acquired is unnecessarily rich for most reactor-physics problems.

A. Energy-transfer cross sections

Following in line, the next quantities of interest are the energy-transfer kernels, defined as the coefficients of the double-differential cross section expansion in a base of Legendre polynomials.

1. The P_0 kernel

We start by considering the isotropic scattering kernel, which is

$$\sigma_0(E_0,E)=2\pi\int_{-1}^{1}d(\cos\theta)\frac{d^2\sigma}{d\Omega\,dE}\ , \tag{14}$$

or, in terms of the synthetic model

$$\sigma_0(E_0,E)=\frac{1}{4\pi}\sum_\nu\sigma_\nu N_\nu\sigma_0^\nu(E_0,E)\ , \tag{15}$$

where

$$\sigma_0^{\nu}(E_0,E)=2\pi\frac{k}{k_0}\int_{-1}^{1}d(\cos\theta)T_\nu(Q,\omega;E_0)\ . \tag{16}$$

This is the contribution to the molecular scattering kernel corresponding to each atomic species ν. In what follows we will drop this index when writing expressions for the atomic contributions, keeping in mind that eventually they must be added according to Eq. (15) to give the molecular cross section.

To proceed with the calculation, it is useful to remember that $S_{\mu,\tau}^{KN}(Q,\omega)$ as defined in Eq. (11) can also be written

$$S_{\mu,\tau}^{KN}(Q,\omega)=\exp\left[-\frac{(1-f)\hbar\omega}{2k_B\tau'}\right]S_{\mu',\tau'}(Q,\omega)\ , \tag{17}$$

with

$$f=(1+4\Gamma k_B\tau\mu)^{-1/2},\quad \mu'=f\mu,\quad \tau'=f\tau\ ,$$

and $S_{\mu',\tau'}(Q,\omega)$ denoting a free-gas scattering function, Eq. (9), but for particles of mass μ' at temperature τ'.

Then, from Eq. (16) and the principal term of Eq. (12), we have

$$\begin{aligned}2\pi\frac{k}{k_0}\int_{-1}^{1}&d(\cos\theta)S_{\mu,\tau}^{KN}(Q,\omega)\\&=\exp\left[-\left[\frac{1-f}{2}\right](x_0^2-x^2)\right]\sigma_{\mu',\tau'}(E_0,E)\\&\equiv\sigma_0^0(E_0,E)\ ,\end{aligned} \tag{18}$$

where we are using the notation

$$\sigma_{\mu',\tau'}(E_0,E)=2\pi\frac{k}{k_0}\int_{-1}^{1}d(\cos\theta)S_{\mu',\tau'}(Q,\omega) \tag{19}$$

and $x_\alpha^2=E_\alpha/k_B\tau'$.

Similarly, we obtain

$$2\pi\frac{k}{k_0}\int_{-1}^{1}d(\cos\theta)S_{\mu,\tau}^{KN}(Q_\pm,\omega_\pm)=\left[1\pm\frac{x_\lambda^2}{x_0^2}\right]^{1/2}\exp\left[-\left[\frac{1-f}{2}\right](x_0^2-x^2\pm x_\lambda^2)\right]\sigma_{\mu',\tau'}(E_{0,\lambda}^{\pm},E) \tag{20}$$

for the contributions originated in the correction term of the synthetic function. Collecting the previous results, we formally can write the (isotropic) atomic scattering kernel as

$$\sigma_0(E_0,E)=\sigma_0^0(E_0,E)-\sum_\lambda p_\lambda\left[n_\lambda\left[1+\frac{x_\lambda^2}{x_0^2}\right]^{1/2}\frac{\partial}{\partial\Gamma}\sigma_0^0(E_{0,\lambda}^{+},E)+(1+n_\lambda)\left[1-\frac{x_\lambda^2}{x_0^2}\right]^{1/2}\frac{\partial}{\partial\Gamma}\sigma_0^0(E_{0,\lambda}^{-},E)\right]\ . \tag{21}$$

Clearly, it is the formal simplicity of the synthetic model that allows us to make further progress in this derivation, because in spite of the appearance of derivatives with respect to Γ in the last formula, we know the analytic expression[23] for the free-gas scattering kernel, Eq. (19). With the notation

$$\eta=\frac{\mu'+1}{2\mu'^{1/2}},\quad \rho=\frac{\mu'-1}{2\mu'^{1/2}}\ , \tag{22}$$

and after some lengthy algebra, we finally obtain

$$\begin{aligned}\sigma_0(E_0,E)=&\exp\left[-\left[\frac{1-f}{2}\right](x_0^2-x^2)\right]\sigma_{\mu',\tau'}(E_0,E)\\&+2\mu' k_B\tau'\sum p_\lambda\left[n_\lambda\left[1+\frac{x_\lambda^2}{x_0^2}\right]^{1/2}\chi(E_{0,\lambda}^{+},E)+(1+n_\lambda)\left[1-\frac{x_\lambda^2}{x_0^2}\right]^{1/2}\chi(E_{0,\lambda}^{-},E)\right]\ .\end{aligned} \tag{23}$$

Here we have defined

$$\chi(E_0,E)=\exp\left[-\left[\frac{1-f}{2}\right](x_0^2-x^2)\right]\left[\left[1+\frac{x_0^2-x^2}{2}\right]\sigma_{\mu',\tau'}(E_0,E)+\xi_{\mu',\tau'}(E_0,E)\right]\ , \tag{24}$$

with

$$\sigma_{\mu',\tau'}(E_0,E)=\frac{\pi\mu'}{2k_B\tau' x_0^2}\{\operatorname{erf}(\eta x-\rho x_0)\mp\operatorname{erf}(\eta x+\rho x_0)-\exp(x_0^2-x^2)[\operatorname{erf}(\rho x-\eta x_0)\mp\operatorname{erf}(\rho x+\eta x_0)]\}\ , \tag{25}$$

and

$$\begin{aligned}\xi_{\mu',\tau'}(E_0,E)=\frac{\pi\mu'}{2k_B\tau' x_0^2}&[(x_0^2-x^2)\exp(x_0^2-x^2)[\operatorname{erf}(\rho x-\eta x_0)\mp\operatorname{erf}(\rho x+\eta x_0)]\\&-\frac{1}{(\pi\mu')^{1/2}}((x+x_0)\exp[-(\eta x-\rho x_0)^2]\mp(x-x_0)\exp[-(\eta x+\rho x_0)^2]\\&+\exp(x_0^2-x^2)\{(x+x_0)\exp[-(\rho x-\eta x_0)^2]\mp(x-x_0)\exp[-(\rho x+\eta x_0)^2]\})]\ ,\end{aligned} \tag{26}$$

where the upper (lower) sign holds for upscattering (downscattering) processes.

The formulas given above, Eqs. (23)–(26) with the definitions (22), are the analytic expression of the isotropic energy-transfer kernel derived from the synthetic scattering function. It is convenient to make a few comments on some of its features.

As the model itself, the kernel contains a principal and a correction term as clearly displayed in Eq. (23), the correction part involving the evaluation of quantities at the fictitious neutron energies $E_{0,\lambda}^{\pm}$. The term proportional to n_λ accounts for phonon annihilation processes, while that containing the factor $(1+n_\lambda)$ corresponds to phonon creation. Of course, this latter term only exists when the incident neutron energy is high enough to allow the transfer of a quantum of energy to the λ oscillator.

The assumption that an Einstein oscillator represents the relevant part of the actual frequency spectrum could be unrealistic, specially for rotations where usually a fairly broad band of eigenfrequencies shows up. In the spirit of our prescription,[13] this is accounted for through the widths σ_λ associated with each eigenfrequency ω_λ, such that the phonon contributions $\mathcal{X}_\lambda^{\pm}$ are evaluated at the effective frequencies $\omega_\lambda^{*}=\omega_\lambda-\sigma_\lambda$.

2. *The P_1 kernel*

The first anisotropic scattering kernel is defined by

$$\sigma_1(E_0,E)=2\pi\int_{-1}^{1}d(\cos\theta)\cos\theta\frac{d^2\sigma}{d\Omega\,dE}\ ,$$

and, according to the synthetic model, it will be represented by an expression of the form

$$\sigma_1(E_0,E)=\sigma_1^0(E_0,E)+2\mu' k_B\tau'\sum_\lambda p_\lambda\left[n_\lambda\left[1+\frac{x_\lambda^2}{x_0^2}\right]^{1/2}\kappa(E_{0,\lambda}^{+},E)+(1+n_\lambda)\left[1-\frac{x_\lambda^2}{x_0^2}\right]^{1/2}\kappa(E_{0,\lambda}^{-},E)\right]\ , \tag{27}$$

where the functions σ_1^0 and κ arise from the principal and correction terms, respectively. Their explicit forms are derived in the Appendix, with the result

$$\sigma_1^0(E_0,E)=\frac{(xx_0)^{-1}}{2}[(x_0^2+x^2)\sigma_0^0(E_0,E)-2\mu'\mathcal{X}(E_0,E)]\ , \tag{28}$$

and

$$\begin{aligned}\kappa(E_0,E)=\frac{(x_0x)^{-1}}{2}\Bigg\{&\left[(x_0^2+x^2)\left[1+\frac{x_0^2-x^2}{2}\right]-\frac{\mu'}{2}[12+6(x_0^2-x^2)+(x_0^2-x^2)^2]\right]\sigma_0^0(E_0,E)\\&+\exp\left[-\left[\frac{1-f}{2}\right](x_0^2-x^2)\right]\left[[x_0^2+x^2-2\mu'(4+x_0^2-x^2)]\xi(E_0,E)-2\mu' f\frac{\partial}{\partial f}\xi(E_0,E)\right]\Bigg\}\ ,\end{aligned} \tag{29}$$

where the functions $\mathcal{X}$ and $\xi_{\mu'\tau'}$ are given by Eqs. (24) and (26), respectively, while σ_0^0 denotes the principal term of the isotropic kernel, Eq. (23).

It is worthwhile to emphasize that $\sigma_0^0(E_0,E)$ and $\sigma_1^0(E_0,E)$ are the analytical expressions of the P_0 and P_1 energy-transfer kernels, respectively, corresponding to a nucleus bound to a semirigid molecule. By this we mean a situation in which its complete dynamics can be described in terms of an effective translational motion modulated by a vibrational factor [cf., Eq. (11)], as first proposed by Krieger and Nelkin.[16] Furthermore, by taking the limit $\Gamma\to 0$ in those formulas one regains the P_0 and P_1 kernels corresponding to a monatomic gas of particles of mass μ in equilibrium at temperature τ.

B. Total cross section

The expression of the total cross section derived from the synthetic model has been already presented,[13] but for the sake of completeness we write down the formulas again using the notation of the preceding sections.

The total cross section per molecule for an incident neutron energy E_0 is

$$\sigma^T(E_0)=\sum_\nu\frac{\sigma_\nu}{4\pi}N_\nu\sigma_\nu^T(E_0), \tag{30}$$

where ν, σ_ν, and N_ν have the same meaning as in Eq. (13) and $\sigma_\nu^T(E_0)$ denotes the atomic contribution.

For this quantity we obtain

$$\sigma_\nu^T(E_0)=\sigma^{KN}(E_0)+\frac{1}{\Gamma}\sum_\lambda p_\lambda\left[n_\lambda\left[1+\frac{x_\lambda^2}{x_0^2}\right]^{1/2}\Lambda(E_{0,\lambda}^{+})+(1+n_\lambda)\left[1-\frac{x_\lambda^2}{x_0^2}\right]^{1/2}\Lambda(E_{0,\lambda}^{-})\right]\ , \tag{31}$$

where the first term of the right-hand side is the result derived by Krieger and Nelkin[16] for the total cross section corresponding to the scattering function $S_{\mu,\tau}^{KN}(Q,\omega)$ as defined in Eq. (11):

$$\sigma^{KN}(E_0)=\frac{\pi\mu'}{Z\Gamma k_B\tau'}\{\operatorname{erf}(Z^{1/2})-(1-C)^{1/2}\operatorname{erf}[Z^{1/2}(1-C)^{1/2}]\exp(-ZC)\}\ . \tag{32}$$

The other quantities appearing in Eq. (31) are

$$\Lambda(E_0)=\frac{\pi\mu'}{Zk_B\tau'}\left[\frac{1}{\Gamma}\mathrm{erf}(Z^{1/2})-A\left[\frac{Z}{\pi}\right]^{1/2}e^{-Z}-(1-C)^{1/2}e^{-ZC}\mathrm{erf}[Z^{1/2}(1-C)^{1/2}]\left[\frac{1}{\Gamma}+AZ+\frac{A}{2(1-C)}\right]\right], \quad (33)$$

with

$$Z=\mu' x_0^2 ,$$

$$C=\frac{1-f^2}{[1+\mu'(\mu'+2f)]} ,$$

and

$$A=4\mu' k_B\tau'\left[\frac{1-C}{f+\mu'}\right]^2 .$$

Of course, the value of σ^T thus obtained coincides at each incident neutron energy E_0 with the integral over final energies of the synthetic scattering kernel, Eq. (23).

By an analogous reason to that already discussed in Sec. III A 1, the functions $\Lambda(E_{0,\lambda}^{\pm})$ are evaluated at effective phonon frequencies $\omega_\lambda^*=\omega_\lambda-\sigma_\lambda(x_\lambda^{*2}=\hbar\omega_\lambda^*/k_B\tau')$. The term involving $\Lambda(E_{0,\lambda}^{-})$ in Eq. (31) accounts for the contribution due to phonon creation processes and only exists when $E_0\geq\hbar\omega_\lambda^*$.

We must emphasize again that the phononlike contributions to the cross sections are originated in the correction terms introduced to account for those scattering processes involving small energy transfers. On the other hand, the principal (or λ independent) term collects the inelastic contributions as the neutron energy increases, because all the terms in the phonon expansion are then accounted for through the short collision-time treatment of the molecule internal degrees of freedom.

The forms adopted by the total cross section, Eq. (31), in the limits of low- and high-incident neutron energy (compared with the characteristic excitation energies $\hbar\omega_\lambda$) are easily obtained from the prescribed variation (Sec. II) of the effective quantities μ, τ, and Γ.

For $E_0\to 0$ we find from Eqs. (4) to (7):

$$\mu\to M_{\mathrm{mol}},\quad \tau\to T,\quad \text{and } \Gamma\to\Gamma_{\max}=\sum_\lambda\frac{(2n_\lambda+1)}{M_\lambda\hbar\omega_\lambda} . \quad (34)$$

Also, from Eq. (32)

$$\sigma^{KN}(E_0\to 0)\simeq\alpha\left[\frac{k_BT}{\pi M_{\mathrm{mol}}E_0}\right]^{1/2} , \quad (35)$$

where

$$\alpha=8\pi M_{\mathrm{mol}}^2/[(1+M_{\mathrm{mol}})^2+4\Gamma_{\max}M_{\mathrm{mol}}k_BT] .$$

There is no possibility of phonon creation in this limit of subthermal incident neutrons, and then we finally obtain

$$\sigma_v^T(E_0\to 0)\simeq E_0^{-1/2}\left[\alpha\left[\frac{k_BT}{\pi M_{\mathrm{mol}}}\right]^{1/2}+\frac{(k_BT)^{1/2}}{\Gamma}\sum_\lambda p_\lambda(x_0^2+x_\lambda^2)^{1/2}\Lambda(E_{0,\lambda}^{+})\right] , \quad (36)$$

where the variables entering in the definition of Λ, Eq. (33), are evaluated according to the values of the basic parameters as given in (34). It is clear that in the case of a hypothetical nonvibrating molecule ($\Gamma=0$), the above expressions reduce to the well-known result[4] for a gas of particles of mass M_{mol} at temperature T.

In the opposite limit, when $E_0\gg\hbar\omega_\lambda$ for all λ, we find that $\mu\to M$, $\Gamma\to 0$, and

$$\tau\to\bar{T}=T+\sum_\lambda\frac{M}{M_\lambda}\left[(n_\lambda+\tfrac{1}{2})\frac{\hbar\omega_\lambda}{k_B}-T\right] ,$$

because all P_λ's are then equal to zero. The synthetic model reduces to the form $S_{M,\bar{T}}(Q,\omega)$ and therefore the total cross section in this energy range is given by[4]

$$\sigma_v^T(E_0\gg\hbar\omega_0)\simeq\frac{4\pi M^2}{(1+M)^2}\left[\left[1+\frac{1}{2y}\right]\mathrm{erf}(y^{1/2})+\frac{1}{\sqrt{\pi y}}e^{-y}\right] , \quad (37)$$

with $y=ME_0/k_B\bar{T}$. Moreover, in the limit of very high neutron energy one recovers the asymptotic expression[24]

$$\sigma_v^T(E_0\to\infty)\simeq\frac{4\pi M^2}{(1+M)^2}\left[1+\frac{1}{2}\frac{k_B\bar{T}}{ME_0}\right] , \quad (38)$$

which clearly shows that in this energy limit the atoms are seen as possessing a kinetic energy associated to the temperature $\bar{T}$ rather than T. This is a general result valid for any state of the scattering system.[25]

C. Diffusion parameters

With the magnitudes previously defined in this section, we are in a position to evaluate a number of quantities of interest in neutron- and reactor-physics calculations. These are the transport coefficients whose expressions can be found in standard textbooks,[23] but we write them here for the sake of completeness as they are used extensively in the following paper to test our model predictions.

The average cosine of the scattering angle is given by

$$\langle\cos\theta(E_0)\rangle=\int_0^\infty dE\,\sigma_1(E_0,E)\Big/\int_0^\infty dE\,\sigma_0(E_0,E) , \quad (39)$$

where σ_0 and σ_1 are the zero- and first-order angular moments of the double-differential cross section, respectively, which are expressed by Eqs. (23) and (27) according to the synthetic model.

For a molecular system, the macroscopic transport cross section is

$$\Sigma_{\mathrm{tr}}(E_0)=N_m\sigma^T(E_0)[1-\langle\cos\theta(E_0)\rangle] , \quad (40)$$

where N_m denotes the molecular number density. $\sigma^T(E_0)$ is the total cross section per molecule as defined by Eq. (30) and explicitly given by Eqs. (31)–(33) for the synthetic model. Clearly, $\Sigma_{\mathrm{tr}}(E_0)$ is a strongly temperature-dependent quantity, basically due to the variation of density but also because the energy transfer and total cross sections themselves depend on the temperature.

The diffusion coefficient $\bar{D}$ can be written as[26]

$$\bar{D}(T)=\int_0^\infty dE_0 D(E_0,T) M_T(E_0) , \tag{41}$$

where $M_T(E_0)$ is a normalized Maxwellian flux distribution

$$M_T(E)=\frac{E}{(k_B T)^2}\exp(-E/k_B T) , \tag{42}$$

and $D(E_0,T)$ is an energy-dependent function defined by

$$D(E,T)=\{3[\Sigma_{\mathrm{tr}}(E)+\Sigma_a(E)]\}^{-1} . \tag{43}$$

In this last expression Σ_a represents the macroscopic absorption cross section of the molecular system. We wanted to write down explicitly the form of $D(E,T)$, as there are experimental data available for this quantity at several energies and temperatures; this information has been very rarely used to test theoretical models.

The neutron diffusion constant $D_0(T)$ of a homogeneous medium is related to the diffusion coefficient $\bar{D}(T)$ by

$$D_0(T)=\bar{D}(T)\bar{v}(T) , \tag{44}$$

where $\bar{v}(T)$ is the average velocity for a Maxwellian spectrum at the temperature T of the medium

$$\bar{v}(T)=1.448\,813\times 10^4\sqrt{T}\,(\mathrm{cm/sec}) . \tag{45}$$

Another related quantity is the thermal neutron diffusion length, given by

$$\bar{L}(T)=[\bar{D}(T)/\bar{\Sigma}_a(T)]^{1/2} , \tag{46}$$

where

$$\bar{\Sigma}_a(T)=\int_0^\infty dE\, \Sigma_a(E) M_T(E) .$$

Finally, the diffusion cooling constant $C(T)$ is given by[27]

$$C(T)=\frac{\bar{D}^2\bar{v}}{2N_m M_2}\left[1+2\frac{d\ln\bar{D}}{d\ln T_n}\right]^2 , \tag{47}$$

where

$$M_2(T)=(k_B T)^{-2}\int_0^\infty\int_0^\infty dE_0 dE\, M_T(E_0) \times(E_0-E)^2\sigma_0(E_0,E) \tag{48}$$

is the rate of mean squared energy exchanged by collision between a thermal neutron and the scattering system. In the approximation of evaluating the derivative in Eq. (47) with respect to the medium temperature T rather than the neutron temperature T_n, we obtain the formula

$$C(T)=\frac{2\bar{v}\varepsilon_D^2}{N_m M_2 (k_B T)^2} , \tag{49}$$

with the definitions

$$\varepsilon_D(T)=\int_0^\infty dE\, M_T(E) D(E,T)(E-\bar{E}) ,$$

and

$$\bar{E}=\tfrac{3}{2}k_B T .$$

Values of the diffusion cooling constant obtained from these expressions should only be taken as estimators of the actual ones, because Eq. (49) is based in the neutron temperature concept and diffusion theory to represent the energy and space dependence of the neutron field.[27] Of course, the real value corresponding to the synthetic model is that one obtained by solving the Boltzmann transport equation with this kernel. In spite of the approximations made en route to deriving Eq. (49), we show in the following paper[14] that it provides good results compared with experimental data for the moderators there considered.

IV. SUMMARY

We have used the synthetic scattering function $T(Q,\omega;E_0)$ describing the interaction of thermal neutrons with molecules to derive some expressions for cross sections of interest in neutron and reactor physics. Certainly, there are many other quantities which can be evaluated once the scattering law is known, but in this work we wanted to concentrate ourselves on the formal derivation of those analytical results from the synthetic model. In the following paper we show a range of calculations based on the expressions presented here for some commonly used moderators, namely H_2O, D_2O, and C_6H_6. From the comparison with experimental data and other theories, it becomes clear that our formulas provide a very good representation of many quantities relevant to the problem of neutron thermalization. We hope that the results presented here could serve as a tool to produce in a very fast—yet accurate—way the basic nuclear data to describe the behavior of the neutron field in a moderating media.

In subsequent publications we will present results of moderator optimization for pulsed neutron sources using the synthetic scattering function at the core of a Monte Carlo simulation, as well as a combined treatment of multiple-scattering and inelasticity effects (on the self-component) in neutron diffraction work on molecular systems.

APPENDIX

We present here the derivation of the explicit forms of the functions $\sigma_1^0(E_0,E)$ and $\kappa(E_0,E)$, which enter in the expression of the P_1 energy-transfer kernel, Eq. (27). In order to evaluate them taking advantage of some results already derived in Sec. III, let us first consider an intermediate kernel

$$\sigma_i(E_0,E)=2\pi\frac{k}{k_0}\int_{-1}^{1}d(\cos\theta)\frac{\hbar^2Q^2}{2}T(Q,\omega;E_0)\ , \tag{A1}$$

where

$$\frac{\hbar^2Q^2}{2}=E_0+E-2(EE_0)^{1/2}\cos\theta\ ,$$

and $T(Q,\omega;E_0)$ is the (atomic) synthetic scattering function.

According to Eqs. (8) and (10), we find

$$\sigma_i(E_0,E)=2\mu' k_B\tau'\chi(E_0,E)+\sum_{\lambda}p_\lambda\left[n_\lambda\left(\frac{E_{0,\lambda}^{+}}{E_0}\right)^{1/2}\psi^{+}(E_0,E)+(1+n_\lambda)\left(\frac{E_{0,\lambda}^{-}}{E_0}\right)^{1/2}\psi^{-}(E_0,E)\right]\ , \tag{A2}$$

where $\psi^{\pm}(E_0,E)$ denote the integrals

$$\begin{aligned}\left(\frac{E_{0,\lambda}^{\pm}}{E_0}\right)^{1/2}\psi^{\pm}(E_0,E)&\equiv 2\pi\frac{k}{k_0}\int_{-1}^{1}d(\cos\theta)\frac{\hbar^2Q^2}{2}\frac{\hbar^2Q_{\pm}^2}{2}S_{\mu',\tau'}^{KN}(Q_{\pm},\omega_{\pm})\\ &=2\mu' k_B\tau'\left(\frac{E_{0,\lambda}^{\pm}}{E}\right)^{1/2}[(E_0+E)\chi(E_{0,\lambda}^{\pm},E)-2\sqrt{E_0E}\,\kappa(E_{0,\lambda}^{\pm},E)]\ .\end{aligned} \tag{A3}$$

On the other hand, from Eq. (A1):

$$\sigma_i(E_0,E)=(E_0+E)\sigma_0(E_0,E)-2\sqrt{E_0E}\,\sigma_1(E_0,E)\ , \tag{A4}$$

and therefore

$$\begin{aligned}\sigma_1(E_0,E)=&\frac{1}{2(E_0E)^{1/2}}[(E_0+E)\sigma_0^0(E_0,E)-2\mu' k_B\tau'\chi(E_0,E)]\\ &+2\mu' k_B\tau'\sum_{\lambda}p_\lambda\left[n_\lambda\left(\frac{E_{0,\lambda}^{+}}{E_0}\right)^{1/2}\kappa(E_{0,\lambda}^{+},E)+(1+n_\lambda)\left(\frac{E_{0,\lambda}^{-}}{E_0}\right)^{1/2}\kappa(E_{0,\lambda}^{-},E)\right]\ .\end{aligned} \tag{A5}$$

Here we are denoting by $\sigma_0^0(E_0,E)$ the principal term of the isotropic energy-transfer kernel $\sigma_0(E_0,E)$, Eq. (23), the remaining correction term in it being cancelled out by those proportional to $\chi(E_{0,\lambda}^{\pm},E)$ contained in $\sigma_1(E_0,E)$ through Eq. (A3).

From the comparison of Eqs. (27) and (A5), it follows

$$\sigma_1^0(E_0,E)\equiv 2\pi\frac{k}{k_0}\int_{-1}^{1}d(\cos\theta)\cos\theta S_{\mu,\tau}^{KN}(Q,\omega)=\frac{1}{2(E_0E)^{1/2}}[(E_0+E)\sigma_0^0(E_0,E)-2\mu' k_B\tau'\chi(E_0,E)]\ , \tag{A6}$$

which is the analytical expression of the P_1 kernel for a nucleus in a semirigid molecule. The corresponding monatomic gas formula is immediately obtained by taking the limit $\Gamma\rightarrow 0$ in Eq. (A6).

To complete this calculation we must evaluate the correction term in $\sigma_1(E_0,E)$. Noting that

$$\begin{aligned}2\mu' k_B\tau'\left(\frac{E_{0,\lambda}^{\pm}}{E_0}\right)^{1/2}\kappa(E_{0,\lambda}^{\pm},E)&\equiv 2\pi\frac{k}{k_0}\int_{-1}^{1}d(\cos\theta)\cos\theta\frac{\hbar^2Q_{\pm}^2}{2}S_{\mu,\tau}^{KN}(Q_{\pm},\omega_{\pm})\\ &=-\frac{\partial}{\partial\Gamma}\left[2\pi\frac{k}{k_0}\int_{-1}^{1}d(\cos\theta)\cos\theta S_{\mu,\tau}^{KN}(Q_{\pm},\omega_{\pm})\right]\ ,\end{aligned}$$

from which

$$\kappa(E_{0,\lambda}^{\pm},E)=(-2\mu' k_B\tau')^{-1}\frac{\partial}{\partial\Gamma}\sigma_1^0(E_{0,\lambda}^{\pm},E)\ , \tag{A7}$$

we finally obtain

$$\begin{aligned}\kappa(E_0,E)=\frac{1}{2x_0x}\Bigg\{&\left[(x_0^2+x^2)\left(1+\frac{x_0^2-x^2}{2}\right)-\frac{\mu'}{2}[12+6(x_0^2-x^2)+(x_0^2-x^2)^2]\right]\sigma_0^0(E_0,E)\\ &+\left[[x_0^2+x^2-2\mu'(4+x_0^2-x^2)]\xi(E_0,E)-2\mu' f\frac{\partial}{\partial f}\xi(E_0,E)\right]\exp\left[-\left(\frac{1-f}{2}\right)(x_0^2-x^2)\right]\Bigg\}.\end{aligned} \tag{A8}$$

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