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Slow-neutron scattering by molecular gases: A synthetic scattering function

J. R. Granada
Centro Atómico Bariloche and Instituto Balseiro, Comisión Nacional de Energía Atómica
and Universidad Nacional de Cuyo, 8400 Bariloche, Argentina

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Motivated by the practical requirements of reactor-physics calculations as well as the necessity of applying inelasticity corrections to the observed spectrum in neutron-diffraction work on molecular gases and liquids, I have developed a synthetic scattering function $T(Q,\omega;E_0)$ which allows a fast and reliable evaluation of cross sections. Unlike the dynamic structure factor (or scattering function) $S(\mathbf{Q},\omega)$ in thermal neutron-scattering theory, $T(Q,\omega;E_0)$ 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). However, the main characteristics of the molecular dynamics are retained through the introduction of an effective mass, and temperature and vibrational factors which depend on the incident neutron energy E_0. This is achieved by the use of the Krieger-Nelkin procedure for orientational averages and by the introduction of "switching functions" $P(E_0)$ which define the variation with E_0 of the above effective quantities. A very simple form is thus obtained for $T(Q,\omega;E_0)$ which yields analytic expressions for the scattering kernel and the total cross section. To gauge the merits and limitations of this prescription I compared its predictions with experiments and other theories in the following cases involving hydrogen-containing molecules: (i) the total cross section of H_2O and C_6H_6; (ii) the scattering cross sections (angular distributions) of H_2O and D_2O at several energies; and (iii) the average of the cosine of the scattering angle in H_2O. It is concluded from the comparisons that the model works in a very satisfactory way. It is anticipated that the present prescription could be a useful tool for the evaluation of departures from elasticity in time-of-flight experiments, where a wide range of neutron wavelengths may contribute at each channel in the observed diffraction spectrum.

I. INTRODUCTION

The interaction of thermal neutrons with molecular gases has been the subject of very extensive investigations for many years from both the theoretical and experimental sides. In going through this process we have learned about the almost embarrassing richness of information which is present in neutron-scattering data, as well as the corresponding difficulties in trying to interpret it.

In fact a large portion of the relevant features in the ω-Q plane are revealed by measurements of the neutron double-differential scattering cross section for a given system. The knowledge of this magnitude makes possible the derivation of a number of quantities of interest in pure and applied research.

This is in general a very expensive procedure and, while unavoidable in certain cases, one can safely assume that experimental data will never cover exactly the material and unique physical conditions of a particular problem. Furthermore, it is not always necessary to have such detailed information, but rather, to have what is needed, usually an integral quantity.

To this end microscopic models capable of yielding differential cross sections have been developed, most of them based on the essentially exact formalism due to Zemach and Glauber[1] which accounts for the quantum nature of the scattering system and the temperature-dependent distribution of its energy states. Although the complete dynamics of the system is built into the Zemach-Glauber formalism, the resultant expressions are too complicated and therefore not quite amenable for calculations. As a consequence, several methods have been proposed to describe the Van Hove scattering function[2,3] $S(\mathbf{Q},\omega)$, notably the phonon expansion, the Gaussian approximation,[4] and the method of steepest descents.[5] Furthermore, different approximations have been developed to simplify the evaluation of $S(\mathbf{Q},\omega)$ for molecular gases involving the classical treatment of some of the internal degrees of freedom[6,7] and the use of average values for the Euler angles to represent the orientational average of the corresponding correlation functions.[8] These necessitate the introduction of an "effective-mass" concept to describe the neutron-molecule interaction.[9]

Even though these early studies on slow-neutron inelastic scattering were mainly motivated by fundamental interest in the atomic motion in solids and liquids, a great deal of effort was dedicated to hydrogenous materials due to their importance as moderators in reactor physics. As a result, large computer codes were written to calculate a number of quantities which are relevant to the problem of neutron thermalization.[11] Detailed calculations of scattering kernels and thermal neutron spectra in a moderating media can then be performed, although they are always computer-time consuming, even for a simple system configuration. Less-detailed calculations involving group constants nevertheless require a good computational basis.

A different kind of study, namely, structure determination of molecular liquids and gases using neutron-diffraction techniques, also requires some model describing the molecular (if not the whole condensed system) dynamics in order to evaluate the unavoidable inelasticity effects[11] which must be subtracted from the observed scattering pattern. In this case the measured spectrum represents a time average over all the configurations defined by the system's atomic motion and therefore the search for purely structural contributions demands a proper accounting of all those processes in which energy is exchanged between the neutron and the scattering system. Here again a detailed knowledge of the shape of the scattering function $S(\mathbf{Q},\omega)$ over a large region of energy and momentum transfer is unnecessary and, in fact, only those interactions which are more operative at a given experimental condition have to be adequately accounted for in order to obtain a reasonably accurate estimation of this correction. From the pioneering papers by Placzek[12] and Wick[13] much work has been done to improve and extend the range of validity of this correction,[14,15] although still retaining in most cases the initial idea of an expansion involving the moments of $S(\mathbf{Q},\omega)$. Only recently, expressions for the scattering law were written down in order to analyze the case of diatomic molecules,[16,17] but again, as in the classic Nelkin model[18] for thermalization in water, they emphasized the existence of different "regimes" as the incident neutron energy becomes comparable to different portions of the system's excitations spectrum.

In this work I introduce a neutron-energy-dependent function $T(Q,\omega;E_0)$ which incorporates the main dynamical characteristics of the molecular unit, but still retains a high degree of simplicity. Under these circumstances this function cannot be expected to give a detailed description of the actual scattering function, but rather, it is intended to produce a good representation of some of the integral properties of $S(\mathbf{Q},\omega)$. One can hope that this synthetic model will be able to produce reliable differential-cross-section predictions which can be used in reactor physics as well as in neutron-diffraction work. I expect this work to be especially applicable to time-of-flight experiments where a wide range of neutron wavelengths characterize the incident beam, thus causing a natural composition of dynamic contributions at each time channel. Only the incoherent components of the neutron-scattering process are considered in this work, the incoherent approximation being usually a good one as far as reactor calculations are concerned. On the other hand, departures from elasticity affecting the interference component in a measured diffraction spectrum is a more delicate question as compared to that related to the self-contribution. In any case, the concepts of effective masses and temperatures are no longer valid in their traditional sense when pairs of atoms are involved in the interaction and, in my opinion, a full calculation based on a proper scattering function is imperative in those cases.

II. FORMULATION OF THE PROBLEM

A. The initial hypothesis

In the next few paragraphs I enumerate the working hypothesis on which the present model is formulated.

1. On the scattering system

I consider the scattering system to be an ideal molecular gas. By this I mean that the translational motion of the molecules' center of mass is that corresponding to a free particle. This hypothesis is *a priori* rather unrealistic for molecular liquids where the translation (and rotation) of a molecular unit may be severely hindered by the presence of its neighbors.[17] However, this collisional regime should become dominant at very low neutron energies where the experimental time scale is comparable to that characteristic of diffusive motion. Certainly this is not the energy region of main interest in reactor-physics calculations, nor is it a common situation in a neutron-diffraction experiment intended to reveal the structure of the system.

2. On the dynamics of the molecular system

I will assume that the rotational and vibrational degrees of freedom are not coupled. This hypothesis represents a first approximation to the real situation and it will be valid as long as the amplitude of atomic oscillations around their equilibrium positions are small compared to the (equilibrium) interatomic distances in the molecule. Under these conditions, the system Hamiltonian can be written as

$$H=H_{\mathrm{tr}}+H_{\mathrm{rot}}+H_{\mathrm{vib}}\ , \tag{1}$$

that is, the sum of the Hamiltonians corresponding to translational, rotational, and vibrational motions, respectively. H_{rot} represents a rigid molecule with atoms at their equilibrium positions and H_{vib} assumes a harmonic potential function. Also, I will accept the common situation, in which vibrational frequencies are much higher than the characteristic (band of) rotational frequencies, to be generally valid.

3. The neutron energy range

As anticipated in Sec. II A 1, I will consider a low-energy limit for the incident neutrons such that the time scale is still sufficiently short as to ensure that departures from a free-particle motion of the molecular mass M_{mol} cannot be detected. This limit is of course dependent on the particular system under investigation as well as on the physical conditions in a given practical problem. Nevertheless, for most real situations this implies neutron energies about a few millielectron volts.

At the other limit, the model should reduce to the free-atom expression of the scattering function involving the atomic mass M and an effective temperature $\overline{T}$. This situation is attained at neutron energies of the order of eV.

The ambitious project stated in the preceding paragraph requires some additional comments in order to make clear from the onset the spirit of the present model. The molecular rotations and vibrations will be represented by Einstein oscillators. Roughly speaking, I shall identify three broad regions which dominate the scattering process, depending on the incident neutron energy as compared to the separation of energy levels of the internal modes: (i) the "molecular" regime, (ii) the "Sachs-Teller" regime, and (iii) the "atomic" regime.

The temperature of the scatterer will affect this picture, shifting and broadening the gap between the regions where each of those regimes are in command. In Sec. III A these topics are discussed in a more formal context.

B. Basic formulas

The microscopic double-differential cross section for the scattering of unpolarized neutrons is[19]

$$\frac{d^2\sigma}{d\Omega\,dE}=\frac{k}{k_0}\sum_{\nu}\frac{\sigma^{\nu}}{4\pi}N^{\nu}S^{\nu}(\mathbf{Q},\omega)\ , \tag{2}$$

where k_0 and k denote the (modulus of) incident and scattered neutron wave vectors, respectively, and 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}. $S^{\nu}(\mathbf{Q},\omega)$ is the corresponding scattering function (or scattering law) which in this case is taken as the self-part only:

$$S^{\nu}(\mathbf{Q},\omega)=\frac{1}{2\pi\hbar}\int_{-\infty}^{\infty}dt\langle\exp[-i\mathbf{Q}\cdot\mathbf{R}_{\nu}(0)]\exp[i\mathbf{Q}\cdot\mathbf{R}_{\nu}(t)]\rangle e^{-i\omega t}\ . \tag{3}$$

Here,

$$\hbar\omega=E_0-E$$

and (4)

$$\hbar\mathbf{Q}=\hbar\mathbf{k}_0-\hbar\mathbf{k}$$

are the energy and momentum transferred from the neutron to the system in the scattering process and $\langle\ \rangle$ denotes both a quantum-mechanical and statistical average. The instantaneous position of any nucleus belonging to the species ν can be written as

$$\mathbf{R}_{\nu}(t)=\mathbf{r}_c(t)+\mathbf{d}_{\nu}(t)+\mathbf{u}_{\nu}(t)\ , \tag{5}$$

where $\mathbf{r}_c$ is the vector to the molecular center of mass, $\mathbf{d}_{\nu}$ is from the center of mass to the equilibrium position, and $\mathbf{u}_{\nu}$ is the displacement from the equilibrium configuration.

The Fourier transform of the scattering function (usually called the "intermediate scattering function") $\chi(\mathbf{Q},t)$ is

$$\begin{aligned}\chi(\mathbf{Q},t)&=\langle\exp[-i\mathbf{Q}\cdot\mathbf{R}_{\nu}(0)]\exp[i\mathbf{Q}\cdot\mathbf{R}_{\nu}(t)]\rangle\\&=\chi^{t}(\mathbf{Q},t)\chi^{rv}(\mathbf{Q},t)\ ,\end{aligned} \tag{6}$$

where

$$\begin{aligned}\chi^{t}(\mathbf{Q},t)&=\langle\exp[-i\mathbf{Q}\cdot\mathbf{r}_c(0)]\exp[i\mathbf{Q}\cdot\mathbf{r}_c(t)]\rangle\\&=\exp\left[\frac{\hbar Q^2}{2M_{\mathrm{mol}}}(it-k_BTt^2/\hbar)\right]\end{aligned} \tag{7}$$

is the intermediate function corresponding to the translational motion of the molecular unit. In writing the result (7), valid for a particle which belongs to an ensemble in statistical equilibrium at temperature T, I have already made use of the assumption expressed by the hypothesis considered in Sec. II A 1.

According to the hypothesis considered in Sec. II A 2, I will assume that rotations and vibrations are not coupled and then

$$\begin{aligned}\chi^{rv}(\mathbf{Q},t)&=\chi^{r}(\mathbf{Q},t)\chi^{v}(\mathbf{Q},t)\\&=\langle\exp[-i\mathbf{Q}\cdot\mathbf{d}_{\nu}(0)]\exp[i\mathbf{Q}\cdot\mathbf{d}_{\nu}(t)]\rangle_{T,R}\langle\exp[-i\mathbf{Q}\cdot\mathbf{u}_{\nu}(0)]\exp[i\mathbf{Q}\cdot\mathbf{u}_{\nu}(t)]\rangle_{T,R}\ .\end{aligned} \tag{8}$$

The subscripts T and R have been made explicit to emphasize that the expectation value of the operators involved must be averaged over both the thermal distribution and molecular orientations.

As was stated previously, we will make the approximation that the internal degrees of freedom of the molecule carry out simple harmonic oscillations. The resultant expression for the intermediate scattering functions is[1]

$$\chi^{0s}(\mathbf{Q},t)=\left\langle\prod_{\lambda}\left[\exp\left[-\frac{\hbar}{2\omega_{\lambda}}(2n_{\lambda}+1)(\mathbf{Q}\cdot\mathbf{C}^{\lambda})^2\right]\exp\left[\frac{\hbar}{2\omega_{\lambda}}[(n_{\lambda}+1)e^{i\omega_{\lambda}t}+n_{\lambda}e^{-i\omega_{\lambda}t}](\mathbf{Q}\cdot\mathbf{C}^{\lambda})^2\right]\right]\right\rangle_R\ , \tag{9}$$

where the superscript $0s$ stands for rotations and vibrations and λ runs over all internal modes, each with angular frequency ω_{λ}. I have omitted the subscript ν that characterizes the set of atoms located at equivalent molecular positions. Also, n_{λ} is the thermally averaged occupation number:

$$n_{\lambda}=(e^{\hbar\omega_{\lambda}/k_BT}-1)^{-1}\ . \tag{10}$$

From the requirement that the free-atom cross section be approached at high neutron energies (large compared with k_BT and the largest $\hbar\omega_{\lambda}$), a normalization condition for the amplitude vectors is obtained:[9]

$$\frac{1}{M_{\mathrm{mol}}}+\left\langle\sum_{\lambda}(\mathbf{Q}\cdot\mathbf{C}^{\lambda})^2\right\rangle_R=\frac{1}{M}\ , \tag{11}$$

where M is the mass of the nucleus under consideration. This result applies if an average in the manner of Krieger and Nelkin is performed over molecular orientations (see Sec. III A).

III. THE MODEL

In order to proceed in the search for a function $T(Q,\omega;E_0)$ capable of yielding an adequate representation of some integral properties of $S(\mathbf{Q},\omega)$, I must introduce further approximations beyond those already made in the preceding section. Historically, a number of models were developed starting from the Zemach-Glauber formalism to describe neutron scattering from molecules, with the aim of making the expressions more tractable for calculations although a limitation of their range of applicability was unavoidable. Those models have been extensively discussed in the literature[9,20,21] and I can only add—using a very simplified scheme—that most of them were devised to operate in one of the regimes mentioned in Sec. II A. While they proved to be adequate aids for studying the many problems that involve atomic motion in molecular systems, there has been increasing interest in the search for ways to cover the gaps between those regimes.[15,17,22] Before entering into the details of the prescription used to achieve this, we need to consider some previous results which form the basis for the present formulation.

A. The starting approximations

So far, I have presented in Eqs. (7) and (9) expressions of the intermediate scattering function for the free-particle translation (of the molecular unit) and for a nucleus in a harmonic-oscillator potential. An approximate procedure to perform the average over molecular orientation was introduced by Krieger and Nelkin,[8] which implies performing that average on the arguments of the exponential functions in Eq. (9). Then we obtain

$$\chi^{0s}(\mathbf{Q},t)\simeq\exp\left[-\frac{\hbar^2Q^2}{2}\gamma\right]\times\exp\left[\frac{\hbar^2Q^2}{2}\sum_{\lambda}\frac{M_\lambda^{-1}}{\hbar\omega_\lambda}\left[(n_\lambda+1)e^{i\omega_\lambda t}+n_\lambda e^{-i\omega_\lambda t}\right]\right],\qquad(12)$$

where

$$M_\lambda^{-1}=\langle(\hat{Q}\cdot\mathbf{C}^\lambda)^2\rangle_R\qquad(13)$$

and

$$\gamma=\sum_\lambda\frac{M_\lambda^{-1}}{\hbar\omega_\lambda}(2n_\lambda+1)\;.\qquad(14)$$

Also, from Eqs. (11) and (13)

$$\frac{1}{M_{\text{mol}}}+\sum_\lambda\frac{1}{M_\lambda}=\frac{1}{M}\,,\qquad(15)$$

and therefore M_λ represents an effective mass for the atomic motion under the mode λ. Equation (12) has been the starting point of several models of bound atoms in molecules, including the pioneering work of Nelkin on water.[18]

By remembering that

$$S(\mathbf{Q},\omega)=\frac{1}{2\pi\hbar}\int_{-\infty}^{\infty}dt\,e^{-i\omega t}\chi^t(\mathbf{Q},t)\chi^{0s}(\mathbf{Q},t)\,,\qquad(16)$$

it is evident from Eqs. (7) and (12) that

$$\int_{-\infty}^{\infty}d\omega\,S(\mathbf{Q},\omega)=\chi(\mathbf{Q},0)=1\,,\qquad(17)$$

whereas the first moment of the scattering function gives

$$\hbar\int_{-\infty}^{\infty}d\omega\,\omega S(\mathbf{Q},\omega)=-i\frac{\partial\chi}{\partial t}(\mathbf{Q},t)\Big|_{t=0}=-i\left[\frac{\partial\chi^t}{\partial t}+\frac{\partial\chi^{0s}}{\partial t}\right]_{t=0}.$$

Now, from Eq. (7),

$$-i\frac{\partial\chi^t}{\partial t}(\mathbf{Q},t)\Big|_{t=0}=\frac{\hbar Q^2}{2M_{\text{mol}}}$$

and from Eq. (12)

$$-i\frac{\partial\chi^{0s}}{\partial t}(\mathbf{Q},t)\Big|_{t=0}=\frac{\hbar Q^2}{2}\sum_\lambda M_\lambda^{-1}\,,$$

so that we finally obtain

$$\hbar\int_{-\infty}^{\infty}d\omega\,\omega S(\mathbf{Q},\omega)=\frac{\hbar Q^2}{2M}\,,\qquad(18)$$

a result that validates the normalization condition (11) through the definition (13).

It is instructive now to remember the asymptotic forms of $S(\mathbf{Q},\omega)$ for small and large energy transfer in the scattering process.

(i) $\omega\ll\omega_\lambda$ *for all* λ. In terms of the time-dependent formalism this situation corresponds to long interaction times, or more precisely, times which satisfy the condition $\omega_\lambda t\gg1$ (for all λ). Then Eq. (12) reduces to the first exponential function and

$$S(\mathbf{Q},\omega)\sim S^t_{M_{\text{mol}},T}(\mathbf{Q},\omega)\exp\left[-\frac{\hbar^2Q^2}{2}\gamma\right],\qquad(19)$$

where

$$S^t_{M_{\text{mol}},T}(\mathbf{Q},\omega)=(2\pi\hbar^2Q^2M_{\text{mol}}^{-1}k_BT)^{-1/2}\times\exp\left[-\frac{(\hbar\omega-\hbar^2Q^2/2M_{\text{mol}})^2}{2\hbar^2Q^2M_{\text{mol}}^{-1}k_BT}\right]\qquad(20)$$

is the scattering function for a free gas of particles of mass M_{mol} in equilibrium at a temperature T. The result (19) for a quasirigid molecule has been used by Krieger and Nelkin[8] in the formulation of their quasiclassical approximation and discussed by several authors.[16,17,21]

(ii) $\omega\gg\omega_\lambda$ *for all* λ. This case corresponds to the situation of short collision times in the time-dependent formalism. Expanding the argument of the second exponential in Eq. (12) up to second order in t we obtain

$$\chi^{0s}(\mathbf{Q},t)\simeq\exp\left[\frac{\hbar Q^2}{2}\sum_\lambda M_\lambda^{-1}\left[it-\frac{\omega_\lambda}{2}(2n_\lambda+1)t^2\right]\right].\qquad(21)$$

Introducing the mean energy associated to the mode λ of oscillation,

$$E_\lambda=\hbar\omega_\lambda(n_\lambda+\tfrac{1}{2})\,,\qquad(22)$$

and combining Eqs. (7) and (21), we obtain

$$\chi^t(\mathbf{Q},t)\chi^{0s}(\mathbf{Q},t) = \exp\left\{\frac{\hbar Q^2}{2M}\left[it - \left[\frac{M}{M_{\mathrm{mol}}}k_B T + \sum_\lambda \frac{M}{M_\lambda}E_\lambda\right]\frac{t^2}{\hbar}\right]\right\}, \tag{23}$$

where the normalization condition, Eq. (15), was used. Defining an "effective temperature" $\overline{T}$ by

$$k_B\overline{T} = k_B T + \sum_\lambda \frac{M}{M_\lambda}(E_\lambda - k_B T), \tag{24}$$

we finally obtain

$$S(\mathbf{Q},\omega) \sim S^t_{M,\overline{T}}(\mathbf{Q},\omega) \tag{25}$$

with $S^t(\mathbf{Q},\omega)$ given by Eq. (20). Thus the scattering function has a Gaussian shape with its maximum (in ω scale) located at $\hbar Q^2/2M$, indicating that in the collision the atom recoils, presenting its real mass. However, the appearance of an effective temperature $\overline{T}$ instead of the system temperature T as in the ideal gas case, reveals how the atoms retain their condition of belonging to a molecular unit with internal degrees of freedom.

For illustrative purposes, let us consider the case of neutron scattering by a molecule in the "Sachs-Teller regime," by which we mean a temperature T and incident neutron energy E_0 satisfying[8] $k_B T \gg B$ (rotational constant) and $E_0 \gg (Bk_BT)^{1/2}$ (spacing between rotational levels around the most probable one). These are the hypotheses required for the validity of the so-called "quasiclassical approximation,"[19,20] according to which the combined effect of translations and rotations is described by the free motion of a particle with a mass equal to the (spherically averaged) tensorial mass $\mathscr{M}$ introduced by Sachs and Teller.[6] Then, Eq. (15) is rewritten as

$$\frac{1}{\mathscr{M}} + \sum_{\lambda_{\mathrm{vibr}}} \frac{1}{M_\lambda} = \frac{1}{M}\,. \tag{26}$$

If both $k_B T$ and E_0 are smaller than the lowest vibrational level there cannot be exchange of vibrational quanta in the collision and the scattering function becomes

$$S(\mathbf{Q},\omega) \sim S^t_{\mathscr{M},T'}(\mathbf{Q},\omega)\exp\left[-\frac{\hbar Q^2}{2}\gamma_v\right], \tag{27}$$

where T' is an effective temperature given by

$$k_B T' = E_r + \frac{\mathscr{M}}{M_{\mathrm{mol}}}(k_B T - E_r) \tag{28}$$

with E_r being the mean rotational energy and γ_v given by (14) but including only vibrations.

B. The present prescription

In the preceding section we reviewed some results of the theory of slow-neutron scattering by molecules with the purpose of setting a reference frame for the development of the present prescription. The different forms that the scattering function takes in the cases considered above are indicative of the main features that I want to retain in this formulation, that is, the appearance of different effective translational masses, temperatures, and vibrational factors according to the energy exchanged in the interaction. Along this line a synthetic function $T(Q,\omega;E_0)$ is proposed, which uses the incident neutron energy E_0 as the variation parameter to determine the values of those effective quantities across the three regimes (of Sec. II A 3). The same idea is borne in the classic Nelkin model for water,[18] where different values of the constants and expressions for the scattering function were introduced to evaluate cross sections in separate energy regions.

The effect of individual quantum excitations of the internal modes is not accounted for in a detailed manner, but rather their combined effect is described by a kind of envelope through the use of effective quantities. I start by considering the mass and then, instead of (15), define

$$\frac{1}{\mu} = \frac{1}{M_{\mathrm{mol}}} + \sum_\lambda \frac{1-P_\lambda}{M_\lambda} = \frac{1}{M} - \sum_\lambda \frac{P_\lambda}{M_\lambda}\,. \tag{29}$$

The P_λ's are "switching" functions which depend on the incident neutron energy and on that part of the molecular frequency spectrum associated to each mode λ. Every P_λ tends to zero as the corresponding mode becomes fully excited from the point of view of the collision process, that is when a quasiclassical treatment is applicable. On the other hand, the value of P_λ is 1, if the neutron does not have sufficient energy to excite any of the λ-oscillator energy levels. At intermediate energies the variation of P_λ should depend on the shape of the frequency spectrum in the vicinity of ω_λ, but I have adopted the expression

$$P_\lambda(E_0) = \frac{\exp\{-\frac{1}{2}[(E_0-\hbar\omega_\lambda)/\hbar\sigma_\lambda]^2\} + (\pi/2)(\hbar\omega_\lambda/\hbar\sigma_\lambda)(1+\mathrm{erf}\{(1/\sqrt{2})[(\hbar\omega_\lambda-E_0)/\hbar\sigma_\lambda]\})}{\exp[-\frac{1}{2}(\hbar\omega_\lambda/\hbar\sigma_\lambda)^2] + (\pi/2)(\hbar\omega_\lambda/\hbar\sigma_\lambda)\{1+\mathrm{erf}[(1/\sqrt{2})(\hbar\omega_\lambda/\hbar\sigma_\lambda)]\}} \tag{30}$$

as a simple way to meet the above requirements. Its derivation and supporting arguments as well as the meaning of the quantities σ_λ are given in Appendix A.

In a similar fashion I define an effective temperature τ by

$$\frac{k_B\tau}{\mu} = \frac{k_B T}{M_{\mathrm{mol}}} + \sum_\lambda \frac{1-P_\lambda}{M_\lambda}E_\lambda = \frac{k_B\overline{T}}{M} - \sum_\lambda \frac{P_\lambda}{M_\lambda}E_\lambda\,, \tag{31}$$

where $k_B\overline{T}$ is given by Eq. (24). Clearly τ tends to T, the system temperature, or $\overline{T}$, the "free-atom temperature," according to whether all P_λ are one or zero, respectively.

Finally, an effective vibrational factor Γ is defined by

$$\Gamma = \sum_\lambda \frac{P_\lambda}{M_\lambda}\frac{2n_\lambda+1}{\hbar\omega_\lambda}\,. \tag{32}$$

With the quantities defined by Eqs. (29), (31), and (32), I am in a position to introduce a synthetic function $T(Q,\omega;E_0)$, which is

$$T(Q,\omega;E_0)=S_{\mu,\tau}(Q,\omega)\exp\left[-\Gamma\frac{\hbar^2Q^2}{2}\right]+C_{\mu,\tau}(Q,\omega)\ . \tag{33}$$

In this expression $S_{\mu,\tau}(Q,\omega)$ denotes the scattering function of an ideal gas of particles of mass μ at temperature τ [see Eq. (20)] and $C_{\mu,\tau}(Q,\omega)$ is a correction term given by

$$C_{\mu,\tau}(Q,\omega)=\tfrac{1}{2}\sum_{\lambda}\frac{P_\lambda}{M_\lambda\hbar\omega_\lambda}\left[n_\lambda\hbar^2Q_+^2S_{\mu,\tau}(Q_+,\omega_+)\exp\left[-\Gamma\frac{\hbar^2Q_+^2}{2}\right]+(1+n_\lambda)\hbar^2Q_-^2S_{\mu,\tau}(Q_-,\omega_-)\exp\left[-\Gamma\frac{\hbar^2Q_-^2}{2}\right]\right]\ , \tag{34}$$

where $Q_\pm$ is the (modulus of) scattering vector corresponding to an energy exchange $\hbar\omega_\pm=\hbar(\omega\pm\omega_\lambda)$. 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. The derivation of Eqs. (33) and (34) is given in Appendix B.

It is useful to rewrite $C_{\mu,\tau}(Q,\omega)$ in a formally different way:

$$C_{\mu,\tau}(Q,\omega)=-\sum_{\lambda}\frac{P_\lambda}{M_\lambda\hbar\omega_\lambda}\frac{\partial}{\partial\Gamma}\left[n_\lambda S_{\mu,\tau}(Q_+,\omega_+)\exp\left[-\Gamma\frac{\hbar^2Q_+^2}{2}\right]+(1+n_\lambda)S_{\mu,\tau}(Q_-,\omega_-)\exp\left[-\Gamma\frac{\hbar^2Q_-^2}{2}\right]\right]\ . \tag{35}$$

Then, with the notation

$$S_{\mu,\tau}^{\mathrm{KN}}(Q,\omega)=S_{\mu,\tau}(Q,\omega)\exp\left[-\Gamma\frac{\hbar^2Q^2}{2}\right]\ , \tag{36}$$

we obtain, instead of Eq. (33),

$$T(Q,\omega;E_0)=S_{\mu,\tau}^{\mathrm{KN}}(Q,\omega)-\sum_{\lambda}\frac{P_\lambda}{M_\lambda\hbar\omega_\lambda}\left[n_\lambda\frac{\partial}{\partial\Gamma}S_{\mu,\tau}^{\mathrm{KN}}(Q_+,\omega_+)+(1+n_\lambda)\frac{\partial}{\partial\Gamma}S_{\mu,\tau}^{\mathrm{KN}}(Q_-,\omega_-)\right]\ . \tag{37}$$

This is the central equation of the present work. The function $S_{\mu,\tau}^{\mathrm{KN}}$ can be integrated over scattering angles and final neutron energies, yielding analytical expressions for the scattering kernel $\sigma(E_0,E)$ and the total cross section $\sigma(E_0)$.

IV. CALCULATION OF CROSS SECTIONS

In this section I present results of calculations using the functions $T(Q,\omega;E_0)$ for the evaluation of cross sections in a few practical cases. As the incoherent approximation is used throughout this formulation, I consider examples of hydrogen containing molecules as a convenient test for its predictions. More extensive calculations involving the evaluation of a number of quantities of interest in reactor physics[23] as well as inelasticity effects in neutron-diffraction work[24] will be presented in subsequent papers.

A. Total cross sections

Krieger and Nelkin[8] derived the formula

$$\sigma_{\mu,\tau}^{\mathrm{KN}}(E_0)=\frac{\sigma_b}{4\Gamma E_0}\{\mathrm{erf}(z^{1/2})-(1-c)^{1/2}\exp(-zc)\mathrm{erf}[z^{1/2}(1-c)^{1/2}]\} \tag{38}$$

for the total cross section corresponding to the scattering function $S_{\mu,\tau}^{\mathrm{KN}}(Q,\omega)$, with

$$z=\mu\frac{E_0}{k_B\tau}\quad\text{and}\quad c=\left[1+\frac{(1+\mu)^2}{4\Gamma\mu k_B\tau}\right]^{-1}\ .$$

From Eq. (37) we obtain, after some algebra, the following expression for the total cross section:

$$\sigma^T(E_0)=\sigma_{\mu,\tau}^{\mathrm{KN}}(E_0)+\sum_{\lambda}\frac{P_\lambda}{M_\lambda\hbar\omega_\lambda}\left[n_\lambda\left[1+\frac{\hbar\omega_\lambda^*}{E_0}\right]^{1/2}\Lambda_{\mu,\tau}(E_0+\hbar\omega_\lambda^*)+(1+n_\lambda)\left[1-\frac{\hbar\omega_\lambda^*}{E_0}\right]^{1/2}\Lambda_{\mu,\tau}(E_0-\hbar\omega_\lambda^*)\right]\ , \tag{39}$$

where

$$\Lambda_{\mu,\tau}(E_0)=\frac{\sigma_b}{E_0\Gamma}\left[\frac{1}{4\Gamma}\mathrm{erf}(z^{1/2})-A(z/\pi)^{1/2}e^{-z}-(1-c)^{1/2}e^{-zc}\mathrm{erf}[z^{1/2}(1-c)^{1/2}]\left[\frac{1}{4\Gamma}+Az+\frac{A}{2(1-c)}\right]\right] \tag{40}$$

and

$$A=\mu k_B\tau[(1-c)/(1+\mu)]^2\ .$$

TABLE I. Values of the parameters for this model used in the calculations of Sec. IV. The eigenfrequencies $\hbar\omega_\lambda$ correspond to commonly accepted values, while the effective masses are deduced from the respective Sachs-Teller masses and the quantities $(\mathbf{C}_\lambda)^2$ according to Eqs. (11) and (13) (Refs. 28 and 29). Approximate values of the widths $\hbar\sigma_\lambda$ were taken from the frequency spectra given in Ref. 28. Energies are given in eV and masses in neutron mass units.

	$\hbar\omega_r$	$\hbar\omega_{v_1}$	$\hbar\omega_{v_2}$	$2\hbar\sigma_r$	$2\hbar\sigma_{v_1}$	$2\hbar\sigma_{v_2}$	M_r	M_{v_1}	M_{v_2}
H_2O (H atom)	0.07	0.202	0.48	0.015	0.013	0.013	2.38	4.77	3.18
(O atom)							342	746.0	373.0
D_2O (D atom)	0.05	0.15	0.31	0.015	0.013	0.013	4.39	13.24	6.82
(O atom)							190.5	427	204.0
C_6H_6 (H atom)	0.02	0.12	0.38	0.006	0.021	0.013	28.64	1.53	3.35
(C atom)							91.9	17.7	329.0

In some circumstances the assumption that an Einstein oscillator represents the relevant part of the frequency spectrum is rather poor and indeed this is usually the case for rotations, where a fairly broad band of eigenfrequencies shows up. This is accounted for in this simple prescription through the width σ_λ (Appendix A) associated to each eigenfrequency ω_λ, such that the phonon excitations are evaluated at the effective frequencies $\omega_\lambda^* = \omega_\lambda - \sigma_\lambda$. Of course, the phonon creation term, containing $\Lambda_{\mu,\tau}(E_0 - \hbar\omega_\lambda^*)$, only exists for $E_0 \geq \hbar\omega_\lambda^*$.

In what follows, I will show some results obtained by using the formula (39) to evaluate total cross sections. The values of the constants used for the calculations involving H_2O, D_2O, and C_6H_6 molecules are summarized in Table I. In these examples I have taken $k_BT = 0.0253$ eV, $\sigma_b^{\mathrm{H}} = 81.325$ b, $\sigma_b^{\mathrm{D}} = 7.6$ b, $\sigma_b^{\mathrm{C}} = 5.57$ b, and $\sigma_b^{\mathrm{O}} = 4.25$ b for the thermal energy and the required bound-atom cross sections.

The calculated $\sigma(E_0)$ for H_2O, together with experimental data[25] over the energy range 10^{-3}–1 eV, is shown in Fig. 1. A $1/v$ absorption contribution has been added to the calculated values, using the constant[26] $\sigma_{\mathrm{abs}}^{\mathrm{H}}$ (2200 m/s) = 0.332 b. The overall agreement is very good, except at the lowest energies where the calculation overestimates the total cross section. This disagreement is not unexpected however, as the actual translational motion in the liquid—which controls this regime—is far from the assumed free-particle behavior. In the vicinity of 0.06 eV another discrepancy is apparent, which is shown in greater detail in Fig. 2, where the full experimental points of Russell *et al.*[25] have been drawn in that range. The observed behavior around the "rotational" eigenfrequency $\hbar\omega_r = 0.07$ eV is characteristic of the model and occurs at all $\hbar\omega_\lambda$, particularly in those cases where the actual frequency spectrum is a broad band. The hypothesis of an isotropic Einstein oscillator is then a crude approximation to the real situation and one can hardly expect that a single phononlike term as expressed by $\Lambda_{\mu,\tau}$ could properly describe a feature which is due to a convolution of that type of effect. In the "free-atom region"—at neutron energies higher than the highest vibrational energy—the model reduces to the ideal gas form (atomic mass) with an effective temperature (corresponding to two-thirds of the average kinetic energy) $k_B\bar{T} = 0.115$ eV.[27] For purposes of comparison, the total cross section evaluated by the GASKET-FLANGE[28] computer codes is also shown in Fig. 2; this evaluation is based on a continuous frequency spectrum derived from measured double-differential cross sections according to the Egelstaff extrapolation method.[3]

In Fig. 3 we see the results of this model for the total cross section of benzene together with the experimental data of Sprevak *et al.*[29] As in the case of water, a small

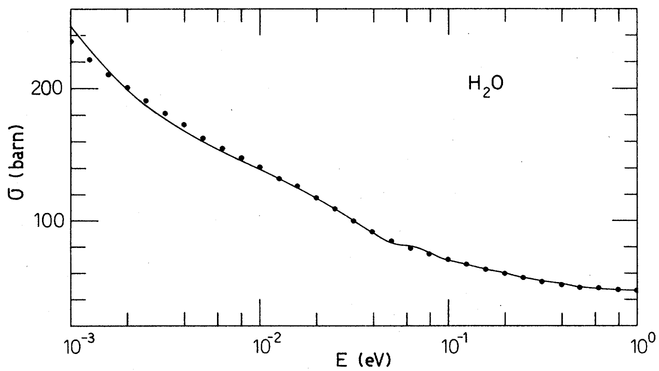

FIG. 1. The total cross section of light water in the thermal energy range. Solid circles are experimental data from Ref. 25. The solid curve is the result of the present model [Eq. (39)].

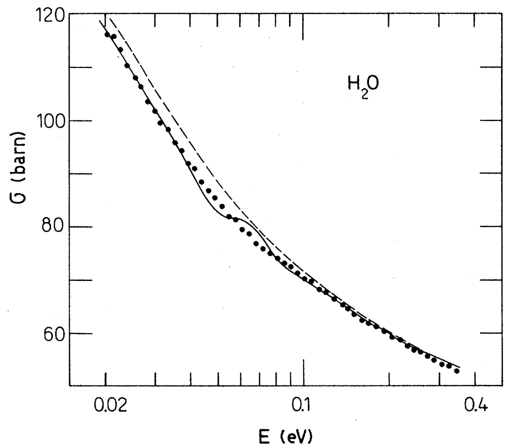

FIG. 2. The total cross section of light water around 0.07 eV, showing in an enlarged scale the behavior of this calculation (solid line) over that region. Solid circles denote the full experimental points of Russell *et al.* (Ref. 25); the dashed curve is the result of a GASKET-FLANGE calculation (see Ref. 28).

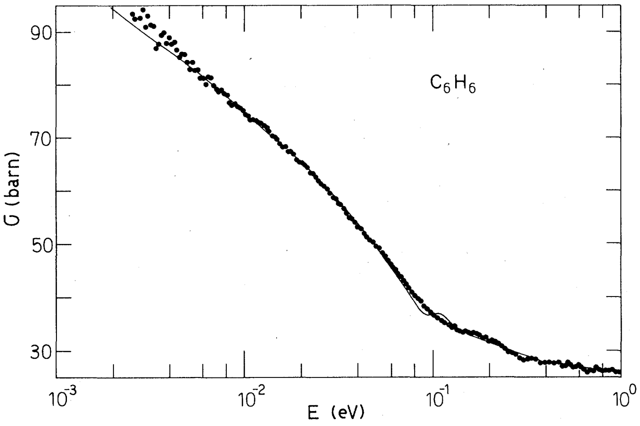

FIG. 3. The total cross section of benzene at thermal energies. The solid line is the result of this model; solid circles are experimental points from Ref. 29.

discrepancy is observed at around 0.10 eV, which originated in an analogous reason already discussed. In spite of this fact, the agreement is very satisfactory.

B. Angular distributions

I present in this section some examples of differential cross sections evaluated according to the present model and compare them with experimental data and other theories. For reactor experiments the observed differential cross section is calculated as

$$\frac{d\sigma}{d\Omega}(\theta;E_0)=\sum_{\text{atoms}}\frac{\sigma_b}{4\pi}\int_{-\infty}^{E_0}d(\hbar\omega)(E/E_0)^{1/2}\times T(Q,\omega;E_0)\frac{f(E)}{f(E_0)}\,, \qquad (41)$$

where E_0 is the energy of the incident neutrons and $f(E)$ denotes the detector efficiency for an outcoming neutron energy E.

In Fig. 4 I show the results for light water corresponding to three well-separated energies. The evaluation at $E_0=0.0205$ eV is compared with Beyster's[30] ($E_0=0.0205$ eV) and Lemmel's[31] ($E_0=0.0225$ eV) data in curve (a), where a calculation based on the Nelkin model is also included. The result of this model for $E_0=0.071$ eV together with Beyster's ($E_0=0.0748$ eV) and Lemmel's ($E_0=0.071$ eV) data is shown in curve (b). Finally, in curve (c) this calculation is shown compared to Beyster's data at $E_0=0.376$ eV. In these examples a black detector (i.e., $f=1$) has been assumed in Eq. (41) as the experiments were done using high-efficiency lithium glass detectors.

Although the present model underestimates the differential cross sections at large scattering angles, one can say that its results are intermediate between those obtained from the Nelkin model and the more elaborate GASKET-FLANGE system. It is also fair to note that Beyster's experiment was done using a linear accelerator, and a more complex integral than that of Eq. (41) must be calculated in the case of a pulsed neutron source; this to-

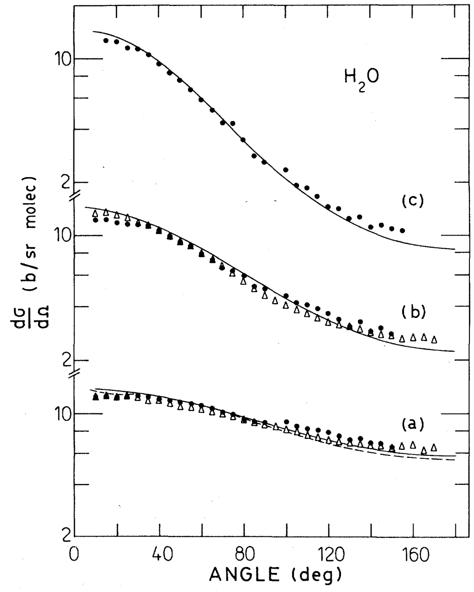

FIG. 4. Angular distribution of neutrons scattered from light water at three different energies: (a) $E_0=0.0205$ eV, (b) $E_0=0.071$ eV, and (c) $E_0=0.376$ eV. The solid curves are the predictions of this model, the dashed curve (a) is the result of the Nelkin model. Experimental points are from Ref. 30 (solid circles) and Ref. 31 (triangles).

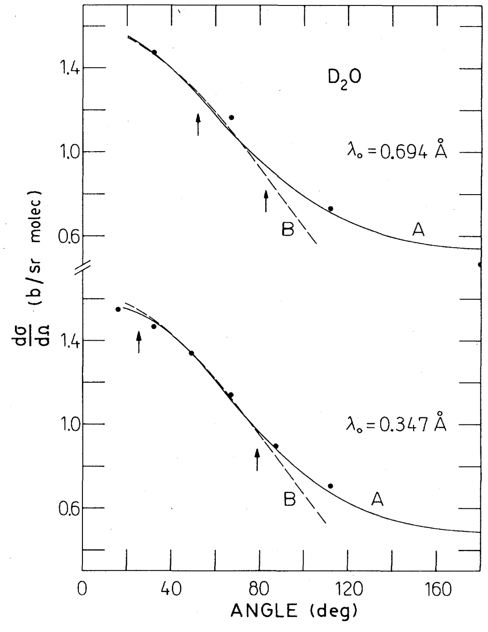

FIG. 5. Angular distribution for neutron scattering from heavy water at two neutron wavelengths available at instrument D4 (ILL, Grenoble). The solid curves are the predictions of this model. The dashed curves are the first-order Placzek expansions used by Walford *et al.* (Ref. 32) to correct the self-component in their observed diffraction spectra; the arrows define the range over which they fit the experimental data. Solid circles correspond to an evaluation made by Powles (taken from Fig. 2 of Ref. 33).

pic will be fully discussed in a forthcoming paper.[24]

Differential cross sections for D_2O (the self-component) are shown in Fig. 5, calculated at energies of 0.1648 eV ($\lambda_0=0.694$ Å) and 0.6794 eV ($\lambda_0=0.347$ Å) which correspond to the experiment performed by Walford *et al.*[32] on the instrument D4 at the High Flux Reactor [Institut Laue-Langevin (ILL) Grenoble]. I used in this case an "exponential" detector law:

$$f(\lambda)=1-\exp(-\lambda/\lambda_d)$$

with a characteristic detector wavelength $\lambda_d=0.27$ Å for the time the measurements were made. In this figure the dashed lines represent the first-order Placzek correction with best-fit parameters used by the experimentalists to account for inelasticity effects, while the arrows indicate the range over which the parameters fit was done. Although the overall normalization constants C_N given by Walford *et al.* have been slightly changed (from 1.06 to 1.03 at $\lambda_0=0.694$ Å and from 1.08 to 1.05 at $\lambda_0=0.347$ Å), good agreement with this evaluation is observed over the region in which they found the first-order correction to be applicable. According to this model—and as far as the "pedestal" is concerned— the C_N values determined by Walford *et al.* imply a 3% error in the absolute normalization of the observed scattering cross sections. I have also included in Fig. 5 some points corresponding to a calculation due to Powles,[33] taken from Fig. 2 of his paper, in which the same data of Ref. 32 are reinterpreted.

C. Average cosine of scattering angle (H_2O)

Another integral property of the scattering kernel is the average cosine of the scattering angle

$$\overline{\cos\theta}(E_0)=\frac{1}{\sigma(E_0)}\int d\Omega\cos\theta\frac{d\sigma}{d\Omega}(\theta;E_0)\ , \quad (42)$$

a quantity of interest in neutron and reactor-physics calculations since it permits the derivation of several transport magnitudes.[34] As a further check of this model, I have calculated this quantity for H_2O and its results compared to other theories are shown in Fig. 6. For the sake of clarity, I did not include experimental points although the best data[30,31] are very well described by the GASKET-FLANGE calculation. Both the Nelkin and McMurry-Russell models[35] give results significantly higher than those from the former one at energies above 0.01 eV, while the present results are closer to the GASKET-FLANGE calculation over most of the range, with a maximum discrepancy of about 10% at the difficult region in the neighborhood of 0.07 eV.

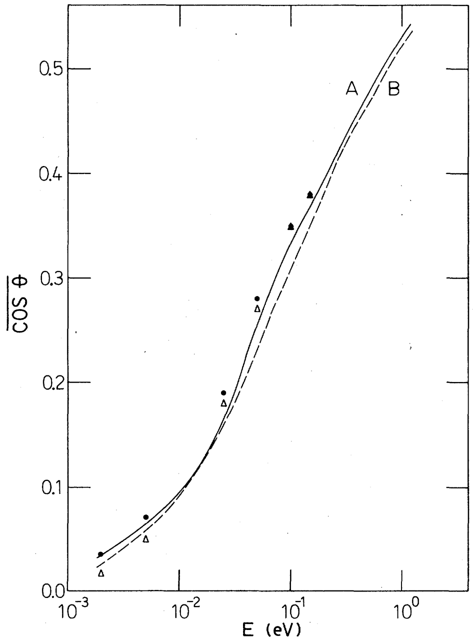

FIG. 6. The average cosine of the scattering angle for light water at thermal energies. The solid line is the result of the present model; the dashed line is a GASKET-FLANGE calculation (Ref. 28). The symbols indicate results from the Nelkin (solid circles) and McMurry-Russell (triangles) models (Ref. 35).

V. SUMMARY AND CONCLUSIONS

The results presented in the preceding section are indicative of what can be expected from this model in the extreme cases of hydrogenous substances. Its merits and limitations are revealed through those few examples which then allow us to discuss its main features on a comparative basis.

The total cross-section calculations (Figs. 1—3) clearly show the difficulty for this molecular-gas model to follow the experimental curve at very low incident neutron energies, certainly an expected result for an energy region in which the assumption of free molecular translation is far from realistic. Still, the discrepancy with data taken on liquid water and benzene is only 5% at 10^{-3} eV. If a special interest exists for a better description at such low energies, this prescription can be modified to include a kernel developed by Egelstaff and Schofield[5] which reduces to the simple diffusion form when collisions dominate. At higher energies, the present model describes very closely the observed behavior of the total cross sections, thus indicating the adequacy of Eq. (39) and the correct variation of the effective quantities μ, τ, and Γ. Admittedly, agreement with experimental $\sigma(E_0)$ data is not sufficient support for a bound-atom model, but nonetheless it is a necessary condition.

The evaluated differential cross sections of H_2O (Fig. 4) shows a slightly more pronounced drop than the experimental data, although the results are still similar or better than those obtained from the Nelkin model. One can therefore expect that in the less unfavorable case of D_2O (Fig. 5) the results should be even more accurate, as the comparison with the first-order Placzek correction found by Walford *et al.*[32] seems to indicate, at least over the limited range where such a correction is applicable.

The average cosine of the scattering angle evaluated for H_2O (Fig. 6) emphasizes the discrepancy already mentioned in relation to the $d\sigma/d\Omega$ results for this molecule, as $\overline{\cos\theta}$ strongly weighs the forward angles contribution. Nevertheless, the present results are better than those from the McMurry-Russell and Nelkin models, if the standard is set by the GASKET-FLANGE calculation.

Finally, I must emphasize once again that the proposed prescription does not pretend to be a scattering function model for molecules, insofar as the full dynamics of atomic motion is not accounted for in a detailed manner and also because only the self- or incoherent contribution to the scattering process is considered. However, I believe that this synthetic function is especially suited for reactor-physics calculations, as it provides analytical expressions for the energy-transfer kernels[23] and the total cross sections. This has been achieved through a simple formulation which still contains the basic dynamical characteristic of the molecular unit. I hope that this model can also offer a useful alternative for the evaluation of self-components in neutron-diffraction work on molecules, especially in time-of-flight experiments where a significant part of the incident spectrum may contribute to each time channel via processes involving a range of energy exchanges with the scattering system.

ACKNOWLEDGMENT

I wish to thank V. Gillette for his assistance during the numerical computations.

APPENDIX A

The functions $P_\lambda(E_0)$ introduced in Sec. III B are intended to represent in a heuristic manner the effect of quantum excitations of the molecule internal modes as the incident neutron energy increases. According to this formulation, those "switching" functions should provide a plausible way of variation for the parameters μ and τ rather than representing a kind of envelope through the total contribution of phononlike terms.

Let $f_\lambda(\omega)$ be that part of the molecular frequency spectrum associated to the mode λ of oscillation. We define a changeover function $g_\lambda(E_0)$ by

$$g_\lambda(E_0)=\int_0^{E_0/\hbar} d\omega \frac{f_\lambda(\omega)}{\omega} \tag{A1}$$

from which the function $P_\lambda(E_0)$ is written as

$$P_\lambda(E_0)=1-g_\lambda(E_0)/g_\lambda(\infty)\ . \tag{A2}$$

Thus the value of P_λ is one for neutron energies much lower than those for which $f_\lambda(\omega)$ is significant, whereas it tends to zero at the other extreme.

The factor ω^{-1} in (A1) represents a first-order approximation to the thermal excitation distribution which is mainly operative for low-frequency modes (as compared with $k_BT/\hbar$). As the shape of $f_\lambda(\omega)$ is chosen in a somewhat arbitrary way I do not feel that the whole expression for $(2n_\lambda+1)$ needs to be included in the definition of $g_\lambda(E_0)$, but only its limiting form as $\omega\to 0$. Within the spirit of this simple prescription and according to the previously stated ideas, I adopted the form[36]

$$f_\lambda(\omega)\sim\omega^2\exp\left[-\frac{(\omega-\omega_\lambda)^2}{2\sigma_\lambda^2}\right] \tag{A3}$$

to represent the frequency distribution of a bound atomic motion. The widths σ_λ are directly taken from the observed or calculated spectrum.

From (A1) and (A3), Eq. (30) is immediately obtained. Clearly its dependence on the temperature of the system is dictated by the shifting and broadening of the corresponding part of the frequency spectrum.

APPENDIX B

In Sec. III B we have introduced some effective quantities (μ,τ,Γ) as a simple way to describe the attainment of a situation in which successive internal degrees of freedom can be treated in a short collision time approximation, as the incident neutron energy increases. Such a procedure leads to a scattering function as that of Eq. (36), i.e., a free-gas expression times a Debye-Waller factor involving the effective mass μ, temperature τ, and vibrational factor Γ. Admittedly, in this process we have associated large energy transfers with high neutron energies (as compared with the energy $\hbar\omega_\lambda$ of each already included mode) and this is certainly a necessary condition. To account—at least partially—for the contribution to the cross section due to processes involving small energy transfers, we make a one-phonon approximation in Eq. (12):

$$\chi^{0s}_{\pm 1}(Q,t)\simeq\exp\left[-\frac{\Gamma\hbar^2Q^2}{2}\right]\left[\frac{\hbar^2Q^2}{2}\sum_\lambda\frac{P_\lambda}{M_\lambda\hbar\omega_\lambda}\times[(n_\lambda+1)e^{i\omega_\lambda t}+n_\lambda e^{-i\omega_\lambda t}]\right]. \tag{B1}$$

We have omitted the elastic term which gives rise to the Debye-Waller factor in the first term on the right-hand side in Eq. (33), once the Fourier transform indicated in Eq. (16) is performed. Also, the functions P_λ appear explicitly to "switch off" the inelastic contributions in (B1) as the neutron energy increases, because all the terms in the phonon expansion are then accounted for through the short collision time approximation.

The next steps are straightforward. The intermediate scattering function (B1) is Fourier transformed, resulting in a scattering function which contains $\delta(\omega\pm\omega_\lambda)$. The expression (34) is finally obtained as a convoluted form:

$$C_{\mu,\tau}(Q,\omega)=\int d\omega' S_{\mu,\tau}(Q,\omega')S^{0s}_{\pm}(Q,\omega-\omega')\ , \tag{B2}$$

where $S_{\mu,\tau}(Q,\omega)$ is the scattering law of a free gas of particles of mass μ at temperature τ and $S^{0s}_{\pm 1}(Q,\omega)$ is the scattering function derived from (B1).

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