

Slow-Neutron Scattering by Hindered Rotators*

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Using a description of a rotator in a strong static electric field the scattering of low-energy neutrons by a condensed system of polar molecules with only rotational degrees of freedom is discussed. Energy and angular differential scattering cross sections for symmetric and linear molecules are derived. It is shown in the case of symmetric molecules that there exist inelastic effects of the same order of magnitude as free-rotation transitions. A simple expression useful for the description of hindered rotation excitations is given. The cross section for water is computed and compared with measurements and existing theory.

I. INTRODUCTION

IN analyzing inelastic scattering of low-energy ($\sim 10^{-2}$ eV) neutrons by macroscopic systems, methods have been given in which the molecules are assumed to be freely rotating.^{1,2} The formalism is therefore not applicable to systems in which appreciable orientation-dependent intermolecular forces are known to exist. For these cases a more realistic description of molecular rotations, in addition to being useful in neutron thermalization studies, should also be of considerable interest in view of recent efforts to investigate liquid dynamics by means of inelastic neutron scattering. Among the various systems studied thus far, water is perhaps the most interesting as well as important. A relatively intense band of transitions have been observed in experiments with water and interpreted to be associated with small-angle or hindered rotational motions of a molecule in the potential field of its neighbors.³ This type of motion has been considered phenomenologically by Nelkin⁴ who assumed that rotations of the water molecule can be described by an oscillator of adjustable mass and frequency.

In this paper we derive the energy and angular differential scattering cross section of a rotator using a description appropriate for polar, symmetric, and linear molecules in a condensed state. An early calculation⁵ of the energies required for complete rotations of a water molecule in an ice-like structure has shown a large potential barrier restricting the rotation of the axis along which the permanent electric dipole moment is directed. The fundamental assumption in the present discussion is that the hindrance can be completely ascribed to a coupling between the dipole moment and a uniform and constant local electric field. This internal field presumably then represents the net effect arising from the presence of the near neighbors, and its order

of magnitude should be essentially that of the corresponding crystalline field.⁶

The assumption that the local electric field is constant in time is not to be taken literally—especially so since our prime concern here is with the liquid state. Instead the assumption should be interpreted in the sense that there exists a time interval corresponding to the duration of local ordering throughout which the local field may be regarded as approximately constant. Furthermore, it is presumed that these “order intervals” are long compared to neutron-nuclear interaction times.

Under the condition of strong coupling, the rotator, to first order in the energy, behaves as a two-dimensional isotropic oscillator. The second-order terms in the energy are independent of the coupling, and must, therefore, be associated with free rotational motions. Cross sections are then derived which include the second-order effects. The presence of energies proportional to the rotational constant gives rise to small energy transitions which conceivably will complicate the interpretation of center-of-mass motions from inelastic neutron scattering data. Moreover, a whole spectrum of frequencies is obtained with each transition involving the oscillator energy. These excitations can therefore be interpreted as giving rise to an effective hindered rotation band which will reduce to only one frequency in a first-order theory. In this sense the present work provides a systematic generalization of Nelkin's treatment.

II. THE HINDERED ROTATOR

The eigenvalue problem of a rigid symmetric molecule with dipole moment \mathbf{u} in a uniform and static electric field \mathbf{E} is well known,⁷

$$\left\{ \frac{1}{\sin\theta} \frac{d}{d\theta} \left(\sin\theta \frac{d}{d\theta} \right) - \frac{(M - K \cos\theta)^2}{\sin^2\theta} - \frac{I}{I_z} K^2 + \frac{(\lambda \cos\theta + E)}{B} \right\} Y(\theta) = 0, \quad (2.1)$$

* Preliminary account of this work has been presented at the Brookhaven Conference on Neutron Thermalization, 1962.

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¹ A. C. Zemach and R. J. Glauber, *Phys. Rev.* **101**, 118 (1956).

² T. J. Krieger and M. S. Nelkin, *Phys. Rev.* **106**, 290 (1957).

³ D. J. Hughes, H. Palevsky, W. Kley, and E. Tunkelo, *Phys. Rev.*, **119**, 872 (1960); K. E. Larsson, S. Holmryd, and K. Otnes, in *Inelastic Scattering of Neutrons in Solids and Liquids* (International Atomic Energy Agency, Vienna, 1961).

⁴ M. Nelkin, *Phys. Rev.* **119**, 747 (1960).

⁵ M. Magat, *Ann. Phys.* **6**, 109 (1936).

⁶ For water a crude estimate gives a coupling of ~ 0.4 eV which may be compared to the height of the hindering potential of about 0.7 eV calculated by Magat.

⁷ The Eulerian angles are defined in the same order as that used by M. E. Rose, *Elementary Theory of Angular Momentum* (John Wiley & Sons, Inc., New York, 1957). Throughout this paper we use a system of units in which $\hbar = 1$.

where the rotational wave function is

$$R(\psi\theta\varphi) = e^{iM\psi} e^{iK\varphi} Y(\theta). \tag{2.2}$$

In this notation \mathbf{u} and $\mathbf{\xi}$ are chosen along the body and space Z axes, respectively, $\lambda = \mu \xi$, and $B = (2I)^{-1}$. The direction of $\mathbf{\xi}$ may be regarded as fixed for the molecular problem; however, in the cross section we must average over all possible field orientations.

Equation (2.1) and the corresponding equation for a linear molecule have been studied quite extensively in the theory of Stark effect in molecular spectra.⁸ The corresponding equation for a plane rotator (Mathieu's equation) has also been used to study the transition from rotation to oscillation as the field increases.⁹ The magnitude of an external field is generally such that λ_{ext}/B is of order unity or less. On the other hand, we can expect strong orientation-dependent intermolecular interactions in crystals and even liquids if sufficient crystalline symmetry still persists, whenever the molecules possess large dipole moment. For these cases $\lambda_{\text{int}} \gg B$. The high-field solutions of (2.1) should, therefore, correspond to a description of hindered rotations.

The energies of rotational states in the limit of strong perturbation have been derived by Maker,¹⁰ and Martin and Strandberg.¹¹ We adopt a somewhat different approach here by observing that under the influence of a strong field, likely values of θ will be confined to a small region about the origin, and so in this sense it is meaningful to examine (2.1) in the small angle approximation. To order θ^2 the resulting equation takes the form of the confluent hypergeometric equation,¹²

$$\left\{ x \frac{d^2}{dx^2} + (|K-M|+1-x) \frac{d}{dx} + \left[\frac{\lambda+E}{B} - \left(\frac{I}{I_z} - \frac{2}{3} \right) K^2 - \frac{M}{3} (M+K) \right] (4\eta)^{-1} - \frac{|K-M|+1}{2} \right\} Y(x) = 0, \tag{2.3}$$

where

$$x = \eta \theta^2,$$

$$\eta^2 = \lambda/2B,$$

$$Y(x) = x^{|K-M|/2} e^{-x/2} L(x).$$

⁸ See, for example, P. Kusch and V. W. Vernon, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1959), Vol. 37, p. 1; and M. W. P. Strandberg, *Microwave Spectroscopy* (Methuen and Company Ltd., New York, 1954).

⁹ L. Pauling, *Phys. Rev.* 36, 430 (1930).

¹⁰ P. D. Maker, thesis, 1961, University of Michigan (unpublished).

¹¹ M. Peter and M. W. P. Strandberg, MIT Research Laboratory of Electronics Technical Report 336 (1957).

¹² P. M. Morse and H. Feshbach, *Methods of Theoretical Physics* (McGraw-Hill Book Company, New York, 1953), Vol. I.

By requiring the wave function to be square integrable in x over the range $(0, \infty)$ we find

$$R = |\zeta KM\rangle = N_{\zeta KM} e^{iM\psi} e^{iK\varphi} x^{|K-M|/2} \times e^{-x/2} L_{\zeta}^{|K-M|}(x), \tag{2.4}$$

$$E_{\zeta KM} = -\lambda + (2\lambda B)^{1/2} (2\zeta + |K-M| + 1) + BK^2 \left(\frac{I}{I_z} - 1 \right) + BMK - \frac{1}{4} B \{ 2\zeta(\zeta+1) + 2\zeta|K-M| + |K-M| - |K-M|^2 + 2 \}, \tag{2.5}$$

$$N_{\zeta KM}^2 = \eta \zeta! / 2\pi^2 [(\zeta + |K-M|)!]^2, \tag{2.6}$$

where $\zeta = 0, 1, 2, \dots$; $M, K = 0, \pm 1, \pm 2, \dots$; and $L_{\zeta}^{|K-M|}$ is the associated Laguerre polynomial. Henceforth, we shall designate the terms proportional to $(2\lambda B)^{1/2}$ and B in $E_{\zeta KM}$ as first- and second-order energies, respectively. By keeping terms to θ^2 , the small-angle approximation gives the energy correctly only to first order. However, Eq. (2.5) is correct to second order because we have included the contributions from terms ignored in (2.3) but which contribute to the energy of order B . Higher order energies can likewise be developed, in fact, the form of $E_{\zeta KM}$ implies an asymptotic series in inverse powers of $(\lambda/B)^{1/2}$.¹¹

The above strong-field solutions should provide a meaningful description of rotations of polar molecules whenever λ is sufficiently large so the small-angle approximation is justified. This condition can be stated as

$$\langle \theta^2 \rangle = (1/\eta) (2\zeta + |K-M| + 1) \ll 1. \tag{2.7}$$

Equation (2.7) implies that only a subset of the totality of available eigenstates corresponds to physical solutions, and in principle only these states should be considered in the cross section calculation.

We have investigated a possible generalization to the asymmetric molecule according to the method used by Wang¹³ and have found, as expected, that the terms arising from the asymmetry couple all states of the symmetric molecule. Thus, numerical methods are necessary to diagonalize the resulting infinite matrix.

III. THE DIFFERENTIAL CROSS SECTION

The energy and angular differential scattering cross section for a system with only rotational degrees of freedom is given by¹

$$\sigma(E_i \rightarrow E_f, \theta) = \frac{k_f}{2\pi N k_i} \sum_{\alpha\alpha'} a_{i\alpha} a_{f\alpha'} \times \int_{-\infty}^{+\infty} dt \langle \chi_{\alpha\alpha'} \rangle_T e^{-iet}, \tag{3.1}$$

¹³ S. C. Wang, *Phys. Rev.* 34, 243 (1929).

where $a_{l\alpha}$ denotes the scattering length for the α th nucleus in the l th molecule; $\hbar k_i$ and $\hbar k_f$ are, respectively, the initial and final neutron momenta; $\epsilon = E_f - E_i$ is the neutron energy exchange; θ is the scattering angle, and N is the total number of nuclei in the system.

The intermediate scattering function is given by

$$\langle \chi_{\alpha\alpha'} \rangle_T = \sum_{\zeta KM} P_{\zeta KM} \langle \zeta KM | \exp(iH) \exp(i\mathbf{k} \cdot \mathbf{b}_{l\alpha}) \times \exp(-iH) \exp(-i\mathbf{k} \cdot \mathbf{b}_{l\alpha'}) | \zeta KM \rangle, \quad (3.2)$$

where H is the Hamiltonian for the l th molecule, b denotes the nuclear position measured with respect to the center of mass of the molecule, $\mathbf{k} = \mathbf{k}_i - \mathbf{k}_f$, $P_{\zeta KM}$ is the probability that the system is initially in the state $|\zeta KM\rangle$:

$$P_{\zeta KM} = (\sum_{\zeta KM} e^{-E_{\zeta KM}/T})^{-1} e^{-E_{\zeta KM}/T},$$

with temperature T in units of Boltzmann's constant.

We consider first the scattering by a single symmetric

molecule,

$$\langle \chi_{\alpha\alpha'} \rangle = \sum_{\zeta KM} \sum_{\zeta' K'M'} P_{\zeta KM} \Lambda e^{i\epsilon t (E_{\zeta KM} - E_{\zeta' K'M'})}, \quad (3.3)$$

$$\Lambda = \langle \zeta KM | \exp(i\mathbf{k} \cdot \mathbf{b}_\alpha) | \zeta' K'M' \rangle \times \langle \zeta' K'M' | \exp(-i\mathbf{k} \cdot \mathbf{b}_{\alpha'}) | \zeta KM \rangle. \quad (3.4)$$

It is advantageous to first perform the average over electric field orientation. Dependence upon this direction is made explicit by the expansion

$$\exp(i\mathbf{k} \cdot \mathbf{b}) = 4\pi \sum_{lkmr} i^l j_l(\kappa b) Y_l^{k*}(\hat{b}') Y_l^r(\hat{\mathbf{k}}') \times D_{-m, -k}^l(\psi\theta\varphi) D_{-r, -m}^l(\psi'\theta'\varphi'), \quad (3.5)$$

where primed and double primed arguments are measured with respect to the body and laboratory axes, and $D_{m, k}^l$ is the familiar rotation matrix.¹⁴ The orientation of the space system (along whose Z axis $\mathbf{\epsilon}$ is directed) with respect to the fixed laboratory axes is specified by Eulerian angles $(\psi'\theta'\varphi')$. We now make the assumption that the direction of $\mathbf{\epsilon}$ is random, and obtain after some manipulation

$$\Lambda = 2(2\pi)^5 N_{\zeta KM}^2 N_{\zeta' K'M'}^2 \sum_{lkm} Q_{lk}^{\alpha\alpha'} F_{\zeta, K-k}^2 \delta_{K', K-k} \delta_{M', M-m}, \quad (3.6)$$

$$Q_{lk}^{\alpha\alpha'} = j_l(\kappa b_\alpha) j_l(\kappa b_{\alpha'}) Y_l^{k*}(\hat{b}_\alpha') Y_l^k(\hat{b}_{\alpha'}), \quad (3.7)$$

$$F = \frac{(-)^{m-k}}{2\eta} G_{mk}^l \sum_s g_{-m, -k}^l(s) \left(\frac{1}{4\eta} \right)^{s+(k-m)/2} S, \quad (3.8)$$

$$S = \int_0^\infty dx x^p e^{-(\xi+1)x} L_\xi^{p-j} L_{\xi'}^{p-q} \\ = (-)^{\xi+\xi'} (\xi+p-j)! (\xi'+p-q)! \sum_{\sigma} \sum_d \frac{(-)^{\sigma+d} \xi^{\sigma-d} (p+\sigma)! (j+\sigma-d)! (q+\sigma-d)!}{d! (\sigma-d)! (\xi'-d)! (\xi-d)! (j+\sigma-d)! (q+\sigma-d)!}, \quad (3.9)$$

where

$$G_{mk}^l = [(l+k)! (l-k)! (l+m)! (l-m)!]^{1/2}, \\ g_{-m, -k}^l(s) = (-)^s [(l+m-s)! (l-k-s)! (s+k-m)! s!]^{-1}, \\ 4\eta\xi = (2l+m-k-2s), \\ 2p = |\alpha| + |\alpha+\Delta| + 2s - \Delta, \\ 2j = |\alpha+\Delta| - |\alpha| + 2s - \Delta, \\ 2q = |\alpha| - |\alpha+\Delta| + 2s - \Delta, \\ \Delta = m - k, \\ \alpha = M - K.$$

In writing an explicit expression for the rotation matrix, use has been made of the small-angle approximation. The integral S is evaluated using generating functions of $L_\xi^{|\alpha|}$ and is valid for j and q positive or zero.

The above equations represent a formal calculation of the cross section in which all possible transitions are taken into account. In view of the fact that the rotator model used is meaningful only if $\eta \gg 1$ it is then appro-

priate to develop a series expression for the cross section in inverse powers of this parameter. The form of Eq. (3.8) is very convenient for this purpose and we find

$$2\eta F = f_0 + (1/4\eta)^{1/2} f_1 + (1/4\eta) f_2 + O[(1/4\eta)^{3/2}], \quad (3.10)$$

where

$$f_0 = N_{\zeta|\alpha|}^{-2} \delta_{mk} \delta_{\xi\xi'},$$

$$f_1 = \pm \delta_{m, k\pm 1} [(l \mp k)(l \pm k + 1)]^{1/2}$$

$$\times \int dx x^{(|\alpha|+|\alpha\pm 1|+1)/2} e^{-x} L_\xi^{|\alpha|} L_{\xi'}^{|\alpha\pm 1|},$$

$$f_2 = -\delta_{mk} (l^2 - k^2 + 2l) \int dx x^{|\alpha|+1} L_\xi^{|\alpha|} L_{\xi'}^{|\alpha|}.$$

The double sign in f_1 denotes a sum of two terms corresponding to upper and lower signs, respectively. The indicated integrals are seen to be special cases of

¹⁴ M. E. Rose, see footnote 7,

S with $\xi=0$ which will provide relative selection rules for ζ and ζ' .

Since the square of F enters into Λ , we find to order $(4\eta)^{-1}$ in the cross section

$$\langle \chi_{\alpha\alpha'} \rangle_T = 4\pi \sum_{lk\zeta KM} Q_{lk}^{\alpha\alpha'} P_{\zeta KM} \times \left[W_0(t) + W_1(t) + O\left(\frac{1}{16\eta^2}\right) \right], \quad (3.11)$$

where

$$W_0(t) = \left[1 - \frac{1}{2\eta} (l^2 + l - k^2) (2\zeta + |K-M| + 1) \right] \times e^{itBk[(IIZ)(2K-k)+M-K]},$$

$$4\eta W_1(t) = \sum_{\zeta'} [(\zeta+1)\delta_{\zeta'\zeta} + \zeta\delta_{\zeta'\zeta-1}] a_{\pm} e^{itE_{\pm}} \quad (K=M)$$

$$= a_{\pm} \sum_{\zeta'} \beta_{\pm} e^{itE_{\pm}} \quad (K \neq M)$$

$$a_{\pm} = (l \mp k)(l \pm k + 1),$$

$$E_{\pm} = E_{\zeta KM} - E_{\zeta' K-kM-k\mp 1},$$

$$\beta_{+} = (\zeta + |K-M| + 1)\delta_{\zeta'\zeta} + \zeta\delta_{\zeta'\zeta-1},$$

$$\beta_{-} = (\zeta + |K-M|)\delta_{\zeta'\zeta} + (\zeta+1)\delta_{\zeta'\zeta+1}.$$

In W_1 the \pm signs again imply the sum of the two terms corresponding to upper and lower signs, respectively. The corresponding expression for the linear molecule is obtained from (3.11) by ignoring the quantum number K and replacing $Q_{lk}^{\alpha\alpha'}$ by

$$[(2l+1)/4\pi] j_l(\kappa b_{\alpha}) j_l(\kappa b_{\alpha'}) (2\delta_{\alpha\alpha'} - 1)^l \delta_{k0}.$$

When $\langle \chi_{\alpha\alpha'} \rangle_T$ is inserted into (3.1) the time integration gives delta functions which are merely statements of energy conservation. In $W_0(t)$ the first term represents the only part of the scattering that is independent of the coupling. All terms with $k=0$ contribute to purely elastic scattering whereas the $k \neq 0$ terms correspond to energy transitions involving second-order energies. These excitations are of the same order of magnitude as what one may expect for the possible hindered translational modes in a liquid. Since only the $k=0$ terms appear in the linear molecule cross section there are thus no small energy transfers associated with rotational motions. This fact suggests that experiments involving polar liquids of linear or diatomic molecules are better suited for the study of molecular center-of-mass motions.

The term $W_1(t)$ represents excitations of first-order as well as second-order energies. It is readily seen that the first-order energy exchange is $(2\lambda B)^{1/2}$. Transitions involving multiples of this amount appear only in higher order terms which have been neglected in (3.11). For example, one can easily show that f_2 leads to an energy transfer of $2(2\lambda B)^{1/2}$, the intensity of which is of order $1/16\eta^2$. We consider a transition involving an

exchange of $(2\lambda B)^{1/2}$ as a hindered rotation excitation. Because of second-order energy transfers, the resulting line shape, even in a reasonably good resolution experiment, will likely appear as a broad distribution rather than a single sharp line.

For scattering from different molecules ($l \neq l'$) the intermediate scattering function is time-independent. If we perform the average over field orientation for the two molecules separately, then

$$\langle \chi_{\alpha\alpha'} \rangle_T = j_0(\kappa b_{\alpha}) j_0(\kappa b_{\alpha'}). \quad (3.12)$$

A consequence of the single-particle model is that the "outer" effects are purely elastic and independent of the initial states of the molecules.

In order to exhibit inelastic effects specific to hindered rotation excitations we shall assume $\lambda \gg B$ so that terms proportional to B in $E_{\zeta KM}$ can be ignored. This simplification leads to

$$\langle \chi_{\alpha\alpha'} \rangle_T = 4\pi \sum Q_{lk}^{\alpha\alpha'} \times \left\{ 1 + \frac{l(l+1) - k^2}{2\eta(1 - e^{-\nu})} (e^{-\nu} e^{it(2\lambda B)^{1/2}} + e^{-it(2\lambda B)^{1/2}}) \right\}, \quad (3.13)$$

where $\nu = (2\lambda B)^{1/2}/T$. In writing (3.13) we have kept only the leading term in $W_0(t)$. Furthermore, although it is not entirely consistent with (2.7) to admit very large values of ζ and $|K-M|$ we have ignored this restriction in carrying out the average over initial states. We anticipate no significant error in doing so since the series is, in general, very rapidly convergent. For water at room temperature $\exp(-2\nu) \sim 10^{-2}$.

The intermediate scattering function (3.13) now describes the scattering process in terms of two components, the time-independent component gives elastic effects while the time-dependent component gives inelastic effects due to hindered rotations. The factor $e^{-\nu}$ associated with neutron energy gain assures that the condition of detailed balance is satisfied,

$$\sigma(E_1 \rightarrow E_2, \theta) = \sigma(E_2 \rightarrow E_1, \theta) (E_2/E_1) e^{(E_1 - E_2)/T}.$$

Equation (3.13) is actually comparable to the cross section used by Nelkin.⁴ The results are similar in that both describe the mechanism underlying hindered rotation excitation as an oscillator transition. Otherwise, the intensity factors and associated parameters in the two models differ significantly. The present work, taking explicit account of molecular symmetry and the polar nature of the molecule, is less empirical. Moreover, by using the rotator description discussed here we obtain a generalization of the first-order result (3.13) to include effects of rotations whose energies are of the same order as those of free rotations.

According to the foregoing discussion the hindered rotation excitation is associated with an energy transfer of about $(2\lambda B)^{1/2}$. From an early discussion given by Pauling⁹ we estimate $\lambda \sim 0.15$ eV for HCl, and thus expect a line at 0.02 eV. Such a transition has recently

been observed for HCl at -130°C in cold-neutron studies of hydrogen halides.¹⁵ A similar transition of ~ 0.066 eV has also been found in both solid and liquid phases of HF. We can then estimate $\lambda \sim 0.83$ eV, a value comparable to that for another highly associated system, water.

IV. NEUTRON SCATTERING BY WATER

As an illustration of the formalism just developed we compute the energy distribution of a beam of 0.065-eV neutrons scattered at 90° by water at 296°K . To simplify the calculation we will assume the water molecule can be treated as a mass 18 diatomic molecule. Moreover, since the experiment¹⁶ under consideration is not sensitive to center-of-mass motions¹⁷ we will use, for convenience, the free gas description for the translational degrees of freedom.¹ The incident neutron energy is well below the first excited internuclear vibrational state, $E_v = 0.2$ eV,¹⁸ so only the effect of zero-point vibration is considered. The cross section thus becomes

$$\begin{aligned} \sigma(E_i \rightarrow E_f, \theta) &= \frac{\sigma_b}{4\pi} \left(\frac{E_f}{4\pi E_R E_i T} \right)^{1/2} e^{-18E_R/E_v} \\ &\times \left\{ e^{-(\epsilon + E_R)^2/4E_R T} + \frac{1}{2\eta(1 - e^{-\nu})} \sum_l l(l+1) \right. \\ &\times (2l+1) j_l^2(\kappa b) \left[e^{-\nu} e^{-(\epsilon + E_R - \sqrt{2}\lambda B)^2/4E_R T} \right. \\ &\left. \left. + e^{-(\epsilon + E_R + \sqrt{2}\lambda B)^2/4E_R T} \right] \right\}, \quad (4.1) \end{aligned}$$

where $E_R = \kappa^2/2M$ is the recoil energy and σ_b is the bound atom cross section of hydrogen. Equation (4.1) is actually the incoherent cross section for hydrogen since the contributions from coherent scattering and the oxygen effects have been estimated to be about 5% and is, therefore, ignored. The following values are

¹⁵ H. Boutin, G. Safford, and V. Brajovic, *Bull. Am. Phys. Soc.* **7**, 500 (1962). We thank Dr. Boutin for sending us the data prior to publication.

¹⁶ B. N. Brockhouse, *Suppl. Nuovo Cimento* **9**, 45 (1958).

¹⁷ S. Yip, thesis, 1962, University of Michigan (unpublished).

¹⁸ G. Herzberg, *Molecular Spectra and Molecular Structure* (D. Van Nostrand, Inc., Princeton, New Jersey, 1950), Vol. II.

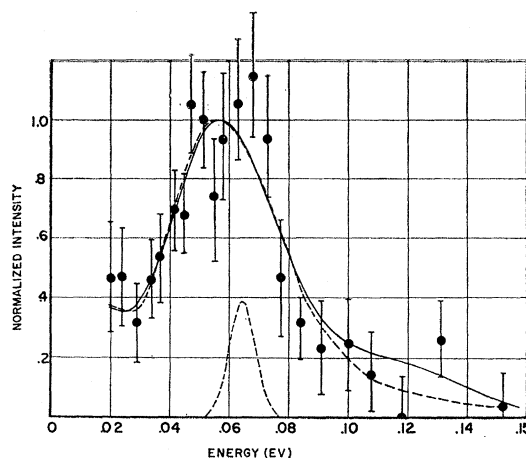


FIG. 1. Energy distributions of neutrons scattered by water, present calculation (solid curve), Nelkin's calculation (reference 4) (dashed curve), and experimental points (reference 16). Also shown is the experimental resolution (reference 16).

used for the parameters, $\lambda = 0.825$ eV, $B = 2.2 \times 10^{-3}$ eV, $b = 9 \times 10^{-9}$ cm.

In Fig. 1 the average of (4.1) over the experimental resolution is shown along with Nelkin's calculation⁴ and the experimental points of Brockhouse.¹⁶ It is observed that within the rather large experimental uncertainties both curves are in qualitative agreement with the measurements. The two predicted intensities, however, differ significantly in the region 0.10–0.14 eV, a region where prominent hindered rotation effects can be expected. Before the accuracies of the two models can be assessed on any quantitative basis, it appears that additional calculations and more sensitive comparisons should be made.

Work is now underway to study the effects of small energy transitions. This investigation along with a discussion of the center-of-mass motions in liquids will be reported later.

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