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THE ROLE OF ROTATIONAL STATES IN THE SCATTERING OF SLOW NEUTRONS BY GAS MOLECULES

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The Zemach-Glauber formalism is used to calculate the differential cross section for scattering of slow neutrons by gas molecules. The limits of applicability of the Krieger-Nelkin formula are examined. A calculation is made of the differential cross section at low momentum transfers, when the quasiclassical approximation is not applicable.

THE question of the role of rotational states of molecules in the scattering of slow neutrons has been treated by many authors.[1-8] In the work of Krieger, Nelkin[2] and Messiah,[3] a quasiclassical calculation is made of the double differential cross section for scattering of neutrons by molecules. Their method is actually equivalent to the mass-tensor approximation. In the papers of Zemach and Glauber[1] and Volkin,[4] they discuss, within the framework of the approximation of small scattering times (Placzek method) the problem of the angular distribution of the scattered neutrons; in the work of Volkin,[4] Rahman,[5] Griffing,[6,7] Young and Koppel,[8] the quantum character of the rotations is taken into account and they sum the partial double differential cross sections for transitions between any two rotational states of the molecule.

The simplicity of the quasiclassical Krieger-Nelkin formula[2] and its wide use in interpreting the results of experiments makes it desirable to establish the limits of its applicability, especially as deviations have already been observed from theoretical calculations made on the quasiclassical approximation.[9-11] A solution of this problem was attempted by Kosaly and Solt,[12] who found the condition for equality of the exact values of the Placzek moments and their values as determined in the quasiclassical approximation. Starting from the moment theorem, these authors propose to correct the Krieger-Nelkin formula. But so far no regular method has been proposed for finding corrections to the Krieger-Nelkin formula in the region of momentum transfers where this formula gives the main properties of the spectrum of scattered neutrons, and there is no simple formula for the neutron spectrum in the region of low momentum transfers, where the quasiclassical formula is not applicable. In the present paper we try to fill this gap.

We shall not be interested in the low-temperature region ($k_BT \ll \Delta E_r$, ΔE_r—separation between rotational levels), since it has already been treated in sufficient detail by Volkin.[4]

1. BASIC FORMULAS

In the Zemach-Glauber formalism the differential cross section for scattering of unpolarized neutrons by gas molecules is given in the form[1]

$$\frac{d^2\sigma}{d\Omega d\varepsilon} = \frac{1}{2\pi}\frac{k}{k_0}\sum_{\nu\nu'}(A_\nu A_{\nu'} + C_\nu^2\delta_{\nu\nu'})\int_{-\infty}^{\infty} d\tau \exp\{-i\tau(\varepsilon + R) - k_BTR\tau^2\}\langle\chi_{\nu\nu'}\quad \langle\chi_{\nu\nu'}(\tau)\rangle = \langle e^{iH\tau}e^{i\kappa\mathbf{r}_\nu}e^{-iH\tau}e^{-i\kappa\mathbf{r}_{\nu'}}\rangle. \quad (1.1)$$

Here $\hbar\mathbf{k}_0$, $\hbar\mathbf{k}$ are the momenta of the neutron in the initial and final states; $\hbar\kappa = \hbar\mathbf{k} - \hbar\mathbf{k}_0$, $\epsilon = \hbar^2(\mathbf{k}^2 - \mathbf{k}_0^2)/2m$ are the change in momentum and energy of the neutron, $R = \hbar^2\kappa^2/2M_{\text{mol}}$, A_ν and C_ν are the amplitudes for coherent and incoherent scattering of the neutron by the ν-th nucleus in the molecule, $\mathbf{r}_\nu = \mathbf{b}_\nu + \mathbf{u}_\nu$ ($\mathbf{b}_\nu$ is the radius vector to the equilibrium position of the ν-th nucleus in the molecule in the center-of-mass system of the molecule, $\mathbf{u}_\nu$ is the displacement of the nucleus from its equilibrium position), $\langle\ldots\rangle$ denotes a statistical average over the states of the system with the Hamiltonian

$$H = H_r + H_v, \qquad H_r = {}^1/{}_2\hbar^2\mathbf{L}\cdot\mathbf{I}^{-1}\cdot\mathbf{L},$$

$$H_v = \sum_\lambda \hbar\omega_\lambda(a_\lambda^+ a_\lambda + {}^1/{}_2), \quad [a_\lambda, a_{\lambda'}^+] = \delta_{\lambda\lambda'},$$

where L_i ($i = 1, 2, 3$) is the angular momentum operator for the molecule, I_{ik} is the moment-of-inertia tensor of the molecule, ω_λ is the frequency of the λ-th normal mode of the molecule, a_λ^+ and

a_λ are the operators for creation and annihilation of the normal vibrations.

The parts H_r and H_V of the Hamiltonian describe, respectively, the rotation and internal oscillations of the molecule. We are assuming that the molecular rotation and vibration do not interact. If we consider the case where the separation of rotation levels is much smaller than the energy of a single vibrational quantum, we easily obtain[2]

$$H_v' \equiv e^{i\varkappa\mathbf{r}_\nu} H_v e^{-i\varkappa\mathbf{r}_\nu} = H_v + V_v + R_v, \; V_v = -\frac{\hbar\varkappa\pi_\nu}{M_\nu},$$

$$R_v = \frac{\hbar^2\varkappa^2}{2M_\nu}, \quad \pi_\nu = i \sum_\lambda \left(\frac{\hbar\omega_\lambda M_\nu}{2}\right)^{1/2} \mathbf{v}_\nu(\lambda)\,(a_\lambda^+ - a_\lambda), \tag{1.2}$$

where $\mathbf{v}_\nu(\lambda)$ are the orthonormal vectors of the normal vibrations of the molecule, M_ν is the mass of the ν-th nucleus. In addition,[2]

$$H_r' \equiv e^{i\varkappa r_\nu} H_r e^{-i\varkappa r_\nu} = H_r + V_r + R_r, \quad R_r = {}^1/_2\hbar^2\varkappa\cdot\mathbf{K}^{\nu\nu}\cdot\varkappa,$$

$$V_r = -{}^1/_2\hbar^2(\varkappa\cdot\mathbf{D}^{\nu T}\cdot\mathbf{I}^{-1}\cdot\mathbf{L} + \mathbf{L}\cdot\mathbf{I}^{-1}\cdot\mathbf{D}^\nu\cdot\varkappa),$$

$$D_{ik}^\nu = -D_{ik}^{\nu T} = -\varepsilon_{ikl}b_{\nu l}, \quad \mathbf{K}^{\nu\nu'} = \mathbf{D}^{\nu T}\cdot\mathbf{I}^{-1}\cdot\mathbf{D}^{\nu'},$$

$$i,\ k,\ l = 1,\ 2,\ 3, \tag{1.3}$$

ϵ_{ikl} is the completely antisymmetric tensor.

Now, using (1.2) and (1.3), we find for $\langle\chi_{\nu\nu}(\tau)\rangle$ the following expression:

$$\langle\chi_{\nu\nu}(\tau)\rangle = \langle\exp\{i\tau(H_r + H_v)\}\exp\{-i\tau(H_r' + H_v')\}\rangle. \tag{1.4}$$

In the present paper we deal only with the incoherent part of the scattering ($\nu = \nu'$). In most cases, for example for hydrogenous compounds, the role of interference terms ($\nu \neq \nu'$) is negligible.

The calculation of $\langle\chi_{\nu\nu}(\tau)\rangle$ is conveniently done by using the following simple relation:[13]

$$\langle e^{i\tau H}e^{-i\tau(H+V)}\rangle = \langle S(\tau)\rangle$$

$$= \exp\left\{-i\int_0^1 d\alpha\int_0^\tau d\tau'\frac{\langle TV(\tau')S_\alpha(\tau)\rangle}{\langle S_\alpha(\tau)\rangle}\right\}$$

$$S_\alpha(\tau) = T\exp\left\{-i\alpha\int_0^\tau d\tau' V(\tau')\right\}, \quad S(\tau) = S_1(\tau),$$

$$V(\tau) = e^{i\tau H}V(0)e^{-i\tau H}, \tag{1.5}$$

where T is the operator for chronological ordering. Expanding $S_\alpha(\tau)$ in the exponent in (1.5) in powers of $V(\tau)$, we find for $S(\tau)$, to terms of fourth order in $V(\tau)$, the following expression:

$$\langle S(\tau)\rangle = \exp\Big\{-i\langle V(0)\rangle\tau - {}^1/_2\int_0^\tau d\tau'\int_0^\tau d\tau''[\langle TV(\tau')V(\tau'')\rangle$$

$$-\langle V(0)\rangle^2] + \frac{i}{6}\int_0^\tau d\tau'\int_0^\tau d\tau''\int_0^\tau d\tau'''[\langle TV(\tau')V(\tau'')$$

$$\times V(\tau''')\rangle - 3\langle TV(\tau')V(\tau'')\rangle\langle V(0)\rangle + 2\langle V(0)\rangle^3]$$

$$+ {}^1/_{24}\int_0^\tau d\tau'\int_0^\tau d\tau''\int_0^\tau d\tau'''\int_0^\tau d\tau''''[\langle TV(\tau')V(\tau'')V(\tau''')$$

$$\times V(\tau''''))\rangle - 3\langle TV(\tau')V(\tau'')\rangle\langle TV(\tau''')V(\tau'''')\rangle$$

$$-4\langle TV(\tau')V(\tau'')V(\tau''')\rangle\langle V(0)\rangle + 12\langle TV(\tau')V(\tau'')\rangle$$

$$\times\langle V(0)\rangle^2 - 6\langle V(0)\rangle^4] + \ldots\Big\}. \tag{1.6}$$

A characteristic feature of (1.6) is that the expansion is made in functions which in many cases, in the terminology of the diagram technique, correspond to coupled diagrams. For example, in the case of a crystal in the harmonic approximation or a Maxwellian gas, the first two terms in (1.6) already give the exact result for the correlation function. The remaining terms in the expansion are identically zero (in the first case as a consequence of Wick's theorem). In the case of a liquid, formula (1.6) allows one to obtain corrections to the Gaussian approximation.

2. CALCULATION AND ANALYSIS OF THE DIFFERENTIAL CROSS SECTION FOR SCATTERING OF SLOW NEUTRONS BY MOLECULES

For simplicity we treat the case of an axially symmetric molecule. We assume that the energy of the incident neutron and the mean energy of thermal motion of the molecule are small compared to the energy of the first vibrational level of the molecule. For this case we rewrite formula (1.4) in the form:

$$\langle\chi_{\nu\nu}(\tau)\rangle = \exp(-i\tau R_v)Z^{-1}\sum_{n_\lambda IKM}\exp\{-\beta(E_{JK} + E_{n_\lambda})\}$$

$$\times\langle JKM, n_\lambda|\exp(i\tau H_v)\exp\{-i\tau(H_v + \delta H_r$$

$$+V_r + R_r + V_v)\}|JKM, n_\lambda\rangle$$

$$\equiv \exp(-i\tau R_v)\langle\exp(i\tau H_v)$$

$$\times\exp\{-i\tau(H_v + \delta H_r + R_r + V_r + V_v)\}\rangle. \tag{2.1}$$

Here $E_{JK} = {}^1\!/_2\hbar^2[pJ(J+1) + (p_3 - p)K^2]$ is the eigenvalue of H_r, $p_i = I_i^{-1}$, $p_1 = p_2 \equiv p \neq p_3$, I_i are the principal moments of inertia of the molecule, $|JKM\rangle$ is the wave function of the rotational state of the molecule, proportional to the Wigner D-function; $J(J+1)$, K, and M are the eigenvalues of the operators L^2, L_3 (the projection along the molecular axis), L_z (the projection along the space fixed axis z); n_λ are the occupation numbers of the vibrational levels, $E_{n_\lambda} = \Sigma\hbar\omega_\lambda(n_\lambda + {}^1\!/_2)$, Z is the partition function for the rotational and vibrational states of the molecular gas, $\delta H_r = H_r - E_{JK}$.

Using (1.6) and keeping of all the terms in $\langle \chi_{\nu\nu}(\tau) \rangle$ that are associated with vibration of atoms in the molecule, only those that are independent of τ, we find for $\langle \chi_{\nu\nu}(\tau) \rangle$ the expression

$$\langle \chi_{\nu\nu}(\tau) \rangle = e^{-2W} \exp \{-ia_1\tau - a_2\tau^2 + ia_3\tau^3 + a_4\tau^4 + \ldots\};$$

$$W = \sum_\lambda \frac{\hbar^2\varkappa^2}{12M_\nu\hbar\omega_\lambda} |\mathbf{v}_\nu(\lambda)|^2, \qquad a_1 = \langle R_r \rangle,$$

$$a_2 = {}^1/_2[\langle V_r^2 \rangle + \langle R_r^2 \rangle - \langle R_r \rangle^2],$$

$$a_3 = {}^1/_6[\langle V_r\delta H_r V_r \rangle + \langle R_r \delta H_r R_r \rangle + \langle V_r^2 R_r \rangle + \langle V_r R_r V_r \rangle + \langle R_r V_r^2 \rangle - 3\langle V_r^2 \rangle\langle R_r \rangle + \langle R_r^3 \rangle - 3\langle R_r^2 \rangle\langle R_r \rangle + 2\langle R_r \rangle^3]. \tag{2.2}$$

The expression for a_4 is quite complicated and will not be given here. In calculating the Debye-Waller factor 2W we have set $\langle n_\lambda \rangle = 0$.

The expression (2.2) is convenient in that, neglecting the term $\langle R_r^2 \rangle - \langle R_r \rangle^2$ in a_2 and the terms with a_3, a_4 in the high temperature limit ($k_BT \gg \Delta EJK$), we arrive at the expression for the correlation function that was obtained by Krieger and Nelkin.[2] Furthermore, with $\langle \chi_{\nu\nu}(\tau) \rangle$ in the form (2.2), we can consider the question of the role of the terms $ia_3\tau^3$, $a_4\tau^4$, i.e., the problem of the validity of the Krieger-Nelkin formula. Substituting (2.2) in (1.1) we verify that the main contribution to the integral comes from the region $\tau \lesssim \tau_0 = \Delta^{-1}$, $\Delta^2 = k_BTR + a_2$. In order for the role of the terms $ia_3\tau^3$, $a_4\tau^4$ to be reduced to corrections to the Krieger-Nelkin formula, we must satisfy the conditions

$$a_3\tau_0^3 < 1, \qquad a_4\tau_0^4 < 1. \tag{2.3}$$

With condition (2.3) the differential cross section for incoherent scattering of neutrons by the ν-th nucleus of the molecule will have the form

$$\frac{d^2\sigma^\nu}{d\Omega\, d\varepsilon} = \frac{k}{2k_0}(A_\nu^2 + C_\nu)^2 \frac{e^{-2W}}{\Delta\sqrt{\pi}} e^{-\xi^2}\left[1 - \frac{a_3}{4\Delta^3}H_3(\xi) + \frac{a_4}{16\Delta^4}H_4(\xi) + \ldots\right], \tag{2.4}$$

where $\xi = (\epsilon + a_1 + R)/2\Delta$, $H_n(\xi)$ is the Hermite polynomial of degree n.

We shall now have to establish when the conditions (2.3) are satisfied. For this purpose we must determine the values of a_2, a_3, a_4. We shall not present the technique for computing matrix elements (one can become familiar with the computation of similar matrix elements from the work of Volkin[4]). We mention only that the computation is conveniently done in the coordinate system fixed in the molecule. Since we are interested in the case of high temperature ($k_BT \gg \Delta E_{JK}$), averages of the type $\langle V_r^2 \rangle$ can be taken in the classical limit. At the same time averages of the type $\langle V_r(\delta H_r)^n V_r \rangle$ ($n \neq 0$) should be calculated using quantum mechanics with passage to the limit of high temperatures only at the end of the computation.

We carry out the further treatment for a spherically symmetric molecule ($p_3 = p$). In this case we have:

$$\langle V_r^2 \rangle = {}^8/_9 B\bar{E}\varkappa^2\mathbf{b}^2, \quad \mathbf{b}^2 = b_{\nu1}^2 + b_{\nu2}^2 + b_{\nu3}^2, \quad B = {}^1/_2\hbar^2 p,$$

$$\bar{E} = {}^3/_2 k_BT, \quad \langle R_r^2 \rangle - \langle R_r \rangle^2 = {}^4/_{45}B^2\varkappa^4\mathbf{b}^4,$$

$$\langle R_r^3 \rangle - 3\langle R_r^2 \rangle\langle R_r \rangle + 2\langle R_r \rangle^3 = {}^6/_5 B^3\varkappa^6\mathbf{b}^6,$$

$$\langle V_r R_r V_r \rangle + \langle V_r^2 R_r \rangle + \langle R_r V_r^2 \rangle - 3\langle V_r^2 \rangle\langle R_r \rangle = {}^{16}/_{45}B^2\bar{E}\varkappa^4\mathbf{b}^4,$$

$$\langle V_r R_r^2 V_r \rangle = {}^{24}/_{35}B^3\bar{E}\varkappa^6\mathbf{b}^6.$$

More complicated calculations are needed for finding expressions of the type $\langle V_r(\delta H_r)^n V_r \rangle$ ($n \neq 0$). Choosing the z axis of the laboratory coordinate system along the vector κ and using the addition theorem for the D-functions, we find

$$\langle V_r(\delta H_r)^n V_r \rangle = {}^4/_{45}B^2\varkappa^2\mathbf{b}^2\langle (E_J - E_{J+1})^n(2J+1)^{-1}(2J+3) \times (4J^2 + 3J + 5) + (E_J - E_{J-1})^n(2J+1)^{-1}(2J-1) \times (4J^2 + 5J + 6)\rangle_J,$$

where the averaging operation $\langle \ldots \rangle_J$ is defined by the formula

$$\langle a_J \rangle_J = \sum_J e^{-\beta E_J}(2J+1)^2 a_J \Big/ \sum_J e^{-\beta E_J}(2J+1)^2.$$

In particular, for n = 1 and n = 2 the preceding expression reduces, respectively, to the relations ($k_BT \gg B$):

$$\langle V_r\delta H_r V_r \rangle = {}^{16}/_9 B^2\bar{E}\varkappa^2\mathbf{b}^2, \quad \langle V_r(\delta H_r)^2 V_r \rangle = {}^{14}/_3 B^2\bar{E}^2\varkappa^2\mathbf{b}^2.$$

At the same time $\langle R_r\delta H_r B_r \rangle \sim B^3\kappa^4 b^4$ with a coefficient of proportionality of order 1.

We proceed to consider the conditions (2.3). To do this we take various limiting cases separately.

1. Low momentum transfers ($\kappa^2 b^2 < 1$). In this case the coefficient has the form

$$\Delta^2 = k_BT(R + {}^4/_3 B\varkappa^2\mathbf{b}^2),$$

while the coefficients a_3 and a_4, which are determined mainly by the expression $\langle V_r(\delta H_r)^n V_r \rangle$ (n = 1, 2), have the form

$$a_3 \approx {}^4/_9 B^2 k_BT\varkappa^2\mathbf{b}^2, \qquad a_4 \approx {}^4/_9 B^2(k_BT)^2\varkappa^2\mathbf{b}^2.$$

In general the criteria (2.3) are not satisfied when $\kappa^2 b^2 < 1$. Consequently the expression (2.4) for the double differential cross section is weakly convergent or even divergent. A fortiori we see that when $\kappa^2 b^2 < 1$ the Krieger-Nelkin formula is not applicable. Kosaly and Solt[12] arrived at practically this same conclusion by using the theorem of moments. In order to obtain a convergent expression for the double differential scattering cross section it is sufficient to note that the divergence of (2.3) is connected with the expression $\langle V_r(\delta H_r)^n V_r\rangle$, proportional to $\kappa^2 b^2$. Thus, going back to (1.4), we can neglect the operator R_r, whose contribution when $\kappa^2 b^2 < 1$ is small compared to the contribution of V_r. In this approximation, using formula (1.6) and retaining terms in the exponent in which the power of κb is no greater than two, we find for the correlation function $\langle\chi_{\nu\nu}(\tau)$

$$\langle\chi_{\nu\nu}(\tau)\rangle = e^{-2W}\exp\left\{-ia_1\tau - \int_0^\tau d\tau'\int_0^\tau d\tau''\langle V_r(\tau')V_r(\tau'')\rangle + \ldots\right\}. \tag{2.5}$$

In the case of a spherically symmetric molecule the function $\langle V_r(\tau')V_r(\tau'')\rangle$ can be easily calculated, and for $k_BT \gg B$ the correlation function $\langle\chi_{\nu\nu}(\tau)$ takes the form

$$\langle\chi_{\nu\nu}(\tau)\rangle = e^{-2W}\exp\{-{}^8/_{45}\varkappa^2 b^2 - {}^2/_{15}Bk_BT\varkappa^2 b^2\tau^2 - a_1\tau + g(\tau) + \ldots\},$$

$$g(\tau) = {}^{32}/_{45}\varkappa^2 b^2(1 - 2Bk_BT\tau^2)\exp(-Bk_BT\tau^2). \tag{2.6}$$

First expanding $\langle\chi_{\nu\nu}(\tau)\rangle$ in powers of $g(\tau)$ and keeping only two terms in the expansion, we find for the double differential cross section for scattering of slow neutrons the expression:

$$\frac{d^2\sigma^\nu}{d\Omega\, d\varepsilon} = \frac{k}{2k_0}(A_\nu^2 + C_\nu^2)e^{-2W}\left\{\frac{1 - 8\varkappa^2 b^2/45}{\Delta_1\sqrt{\pi}} \times \exp\left[-\frac{(\varepsilon + a_1 + R)^2}{4\Delta_1^2}\right] + \frac{16}{45}\frac{\varkappa^2 b^2}{(\pi Bk_BT)^{1/2}}\exp\left[-\frac{(\varepsilon + a_1 + R)^2}{4Bk_BT}\right] \times \frac{(\varepsilon + a_1 + R)^2}{4Bk_BT} + \ldots\right\},$$

$$\Delta_1^2 = k_BT(R + {}^2/_{15}B\varkappa^2 b^2). \tag{2.7}$$

In obtaining (2.7) we restricted ourselves to the first term of the expansion of $\langle\chi_{\nu\nu}(\tau)\rangle$ in the small parameter $\kappa^2 b^2 < 1$. In a definite sense this expansion is the analog of the one-phonon approximation in the theory of scattering of neutrons by a crystal. If we relate (2.7) to the procedure proposed by Egelstaff[14] for determining the spectral density of the autocorrelation function of the momentum of an atom, by introducing the variables $\alpha = \hbar^2\kappa^2/2M_{\mathrm{mol}}k_BT$, $\beta = \epsilon/k_BT$, we easily find from (2.7) the following expression for the spectral density $p(\beta)$ of the autocorrelation function for the momentum of an atom in the molecule:

$$p(\beta) \sim \beta^4\exp\{-\beta^2 k_BT/4B\}, \qquad \beta \neq 0.$$

A peak in the curve of $p(\beta)$ corresponding to the energy transfer $\epsilon \approx 2(Bk_BT)^{1/2}$ should appear in the spectrum of slow neutrons scattered at small angles, completely analogous to what is observed in crystals. Apparently the observed structure of the energy spectrum in the small angle scattering can be explained by using this autocorrelation function for the momenta of the atoms in the molecule.

2. Medium momentum transfers ($1 < \kappa^2 b^2 < k_BT/B$). The coefficients for this case have the form

$$\Delta^2 = k_BT(R + {}^4/_3B\varkappa^2 b^2),$$

$$a_3 = {}^4/_{45}B^2k_BT\varkappa^4 b^4, \qquad a_4 = 0.03B^3k_BT\varkappa^6 b^6. \tag{2.8}$$

One can verify that the conditions (2.3) are satisfied in this case. But this means that (2.4) is applicable. From the derivation of (2.4) it follows that it gives the correct Placzek moments up to fourth order. The second and third terms in (2.4) affect the "wings" of the curve and determine the asymmetry in the neutron spectrum.

3. Large momentum transfers ($\kappa^2 b^2 > k_BT/B$). In this case conditions (2.3) are not satisfied. Formula (2.4) is not applicable. The calculation of the spectrum should be done by Volkin's method,[4] since in this case the operator R_r plays the major role.

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