PHYSICAL REVIEW VOLUME 167, NUMBER 1 5 MARCH 1968

Rotation-Vibration Interaction in Scattering of Slow Neutrons by Spherical-Top Molecules

C. Buzano, F. Demichelis, and M. Rasetti
Istituto di Fisica, Politecnico, Torino, Italy

(Received 12 September 1967)

The scattering of slow neutrons from spherical-top molecules of point group T_d is analyzed and discussed, taking into account the rotation-vibration interaction. After the introduction of the general formulation for the partial differential cross section, the Hamiltonian of the spherical-top molecule is discussed, and the contribution of the rotation-vibration interaction is considered in detail. By using group-theoretical methods, the rotational-vibrational wave function, and then the total wave function, are obtained. Finally, explicit extensive expressions for the intermediate scattering function and the partial differential cross section are derived.

I. INTRODUCTION

In papers[1–4] published in recent years on slow-neutron scattering by molecules, the various authors assume that the molecule is not excited from its ground vibrational state, either thermally or by neutron scattering. Indeed, the previous studies were concerned with the case for which contributions from excited vibrational states could be ignored and the rotation-vibration coupling neglected.

Recently, West, Brugger, and Griffing[5,6] have suggested that the influence on the scattering of vibrationally excited molecules might not be negligible and would have to be considered in making comparisons between theory and experiments.

According to these authors, at a temperature of 306°K, approximately 3% of the CD_4 molecules are in excited vibrational states; the contribution to scattering by the vibrationally excited molecules becomes significant at an energy transfer of about 72 meV, and dominates the scattering at larger energy transfer.

Thus it will be necessary to extend the method of computation so that such contribution to the scattering is included. To do this, the Coriolis force arising from interaction between rotation and vibration must be taken into account.

In this paper, using group-theoretical methods, we discuss the scattering of slow neutrons by a gas of spherical-top molecules of point group T_d, i.e., of tetrahedral molecules containing identical nuclei, and we report an explicit formula for the partial differential cross section, including the rotational-vibrational coupling.

In order to derive the most general expression for the partial differential cross section, according to the treatment of Sinha and Venkataraman[2] we assume that there is only one class of identical nuclei in the molecule and that the group of permutations $\mathcal{P}$ of such nuclei is isomorphous with the point group of the molecule.

II. METHOD OF CALCULATION

A. General Formulation

It is known[1] that the partial differential cross section may be written as

$$\frac{d^2\sigma}{d\Omega dE}=\frac{1}{2\pi\hbar}\frac{k}{k_0}\int e^{-i\omega t}I(\mathbf{Q},t)dt, \qquad (1)$$

where $\mathbf{k}_0$ and $\mathbf{k}$ are the wave vectors of the incident and scattered neutrons, $\hbar\omega=(\hbar^2/2m)(\mathbf{k}^2-\mathbf{k}_0{}^2)$ is the gain of energy, $\hbar\mathbf{Q}$ is the gain in momentum of the neutron, and $I(\mathbf{Q},t)$ is the intermediate scattering function defined by Zemach and Glauber[1]:

$$\begin{aligned}I(\mathbf{Q},t)=\langle\Psi|&\sum_{\substack{\nu,\mu;\\ \text{all nuclei}}}A_\nu A_\mu \exp[i\mathbf{Q}\cdot\mathbf{r}_\nu(t)]\exp[-i\mathbf{Q}\cdot\mathbf{r}_\mu(0)]|\Psi\rangle_T\\ &+\langle\Psi|\sum_{\substack{\nu;\\ \text{all nuclei}}}C_\nu{}^2\exp[i\mathbf{Q}\cdot\mathbf{r}_\nu(t)]\exp[-i\mathbf{Q}\cdot\mathbf{r}_\nu(0)]|\Psi\rangle_T\\ &+\sum_{\substack{\nu,\mu;\ \nu\neq\mu;\\ \text{identical nuclei}}}C_\nu{}^2\langle\Psi|\exp[i\mathbf{Q}\cdot\mathbf{r}_\nu(t)]\exp[-i\mathbf{Q}\cdot\mathbf{r}_\mu(0)]Z(\bar{\Omega})|\Psi\rangle_T. \qquad (2)\end{aligned}$$

The symbol $\langle\cdots\rangle_T$ indicates a thermal average at temperature T, and $\mathbf{r}_\nu(t)$ is the position coordinate of the νth nucleus. The operator $\mathbf{r}_\nu(t)$ is a Heisenberg operator defined by

$$\mathbf{r}_\nu(t)=e^{i(H/\hbar)t}\mathbf{r}_\nu(0)e^{-i(H/\hbar)t}, \qquad (3)$$

where H is the Hamiltonian of the molecule. A_ν, C_ν

---

[1] A. C. Zemach and R. J. Glauber, Phys. Rev. **101**, 129 (1956); **101**, 118 (1956).

[2] S. K. Sinha and G. Venkataraman, Phys. Rev. **149**, 1 (1966).

[3] A. Rahman, J. Nucl. Energy **A13**, 128 (1961).

[4] G. W. Griffing, Phys. Rev. **124**, 1489 (1961).

[5] R. E. West, R. M. Brugger, and G. W. Griffing, Phys. Rev. **148**, 163 (1966).

[6] G. W. Griffing, Phys. Rev. **136**, A988 (1964).

denote, respectively, the coherent and incoherent scattering amplitudes of the νth nucleus;

$$Z(\bar{\Omega})=\frac{\bar{\Omega}(\bar{\Omega}+1)-\eta(S_\nu+1)S_\nu}{S_\nu(S_\nu+1)\eta(\eta-1)}, \tag{4}$$

where S_ν is the spin of each of the identical nuclei, η is the number of identical nuclei in the molecule, and $\bar{\Omega}$ is the total nuclear spin of the identical nuclei.

The last term in Eq. (2) is the contribution of the nuclear spin correlation.

In the case of the vibrational ground state, an expression for this contribution has been derived by Sinha and Venkataraman.[2]

B. Hamiltonian of the Molecule

For molecules with tetrahedral or even higher symmetry (for instance T_d), vibrations which are twofold and threefold degenerate may be present.

In this case the Hamiltonian operator H can no longer be written as a simple sum of the electronic, translational, vibrational, and rotational Hamiltonians:

$$H=H_{\mathrm{el}}+H_{\mathrm{tr}}+H_{\mathrm{v}}+H_{\mathrm{rot}}. \tag{5}$$

In fact, an additional coupling between rotation and vibration (Coriolis coupling) should be taken into account. Let us now consider the influence of the Coriolis interaction and then assume that the anharmonics terms in the potential energy as well as the dependence of the moments of inertia on the normal coordinates are negligible.

The Hamiltonian which describes the rotational-vibrational characteristics of the molecule has the form

$$H=\tfrac{1}{2}\hbar\sum_{s,\sigma}\omega_s\left[\left(\frac{p_{s\sigma}}{\hbar}\right)^2+q_{s\sigma}^2\right]+\tfrac{1}{2}\sum_{\alpha}\frac{(j_{4\alpha}-j_{3\alpha}^*)^2}{I_{\alpha\alpha}^{(e)}}, \tag{6}$$

where

$$q_{s\sigma}=(\omega_s/\hbar)^{1/2}Q_{s\sigma}.$$

Here $Q_{s\sigma}$ are the normal coordinates associated with the normal vibration frequency ω_s; the label s denotes the particular frequency ω_s; and $\sigma=1, 2$ or $1, 2, 3$ depending upon whether the vibration is twofold or threefold degenerate, respectively. $p_{s\sigma}$ are the linear momenta conjugate to $q_{s\sigma}$; $j_{4\alpha}$ is the α component of the total angular momentum $\mathbf{j}_4$ of the molecule; $j_{3\alpha}^*$ is the α component of the internal angular momentum of the nuclei, and is made up of a sum of Coriolis operators, where each operator is associated with a degenerate frequency; $I_{\alpha\alpha}^{(e)}$ are the diagonal components of the moment of inertia $I^{(e)}$ about the α axis; x, y, z are the body-fixed coordinate system; α, β, γ are symbols used to denote x, y, or z.

The Coriolis coupling factor is defined by

$$\zeta_{ss'\sigma\sigma'}^{(\alpha)}=\sum_{\nu}\left(l_{\nu s\sigma}^{(\beta)}l_{\nu s'\sigma'}^{(\gamma)}-l_{\nu s'\sigma'}^{(\beta)}l_{\nu s\sigma}^{(\gamma)}\right), \tag{7}$$

where $(l_{\nu s\sigma}^{(\alpha)}/M_\nu^{1/2})$ are the transformation matrix elements which relate the normal coordinates to the displacement vector of the νth nucleus from its equilibrium position, M_ν being the mass of the νth nucleus.

In spherical-top molecules of tetrahedral symmetry the three moments of inertia are all alike, and, in addition, the Coriolis coupling factors are all equal. (In order to simplify notation, we shall use I instead of $I_{\alpha\alpha}^{(e)}$, and ζ instead of $\zeta_{ss'\sigma\sigma'}^{(\alpha)}$.)

Moreover, there are three species of degenerate vibrational levels: E doubly degenerate, and F_1 and F_2 triply degenerate. However, no Coriolis splitting arises for doubly degenerate vibrational states; indeed from Jahn's rule,[7] the product of the species of the two interacting vibrations $E\times E=A_1+A_2+E$ does not contain the species of the rotation which is, in this case, F_1. On the contrary, the Coriolis interaction does cause a splitting for the triply degenerate vibrational states.

On the basis of these considerations, we can subdivide the Hamiltonian H into two independent parts: the first one containing the factors related to the nondegenerate and twofold-degenerate vibrations (which do not generate internal angular momentum), the second one including terms connected with the threefold-degenerate vibrations and rotations.

Let us then write the Hamiltonian in the following form:

$$H=H_{\mathrm{v}}'+H_{\mathrm{vr}}, \tag{8}$$

where H_{v}' describes the nondegenerate and twofold-degenerate vibrations of the molecule, and H_{vr} describes the rotational-vibrational modes (threefold-degenerate vibrations).

We can now write the unsymmetrized total wave function as a simple product of the electronic, translational, vibrational (for the nondegenerate and twofold-degenerate frequencies), rotational-vibrational, and spin functions, ψ_{el}, ψ_{t}, ψ_{v}', ψ_{vr}, and ξ, respectively:

$$|\Psi\rangle=|\psi_{\mathrm{el}}\rangle|\psi_{\mathrm{t}}\rangle|\psi_{\mathrm{v}}'\rangle|\psi_{\mathrm{vr}}\rangle\xi. \tag{9}$$

Since the molecules are normally to be found in their electronic ground state both before and after the neutron scattering, the function ψ_{el} is invariant under permutation of the matrix elements.

The above assumptions being granted, the translational wave function ψ_{t} transforms according to the identity representation of $\mathcal{P}$.

Since H_{v}' and H_{vr} are invariant under $\mathcal{P}$ we can classify the wave functions ψ_{v}' and ψ_{vr} in terms of the irreducible representations of $\mathcal{P}$. We write these functions as

$$|\psi_{\mathrm{v}}'\rangle\rightarrow|\psi_\gamma{}^{Vs}\rangle,\quad|\psi_{\mathrm{vr}}\rangle\rightarrow|\psi_K{}^{JM}\rangle.$$

S (or M) signifies which particular representation of a given type V (or J) the wave function belongs to; and γ (or K), the row of this representation. Finally, the

---

[7] G. Herzberg, *Infrared and Raman Spectra of Polyatomic Molecules* (D. Van Nostrand, Inc., New York, 1946).

spin function ξ is simply the product of the spin functions for each of the nuclei.

C. Rotational-Vibrational Wave Function

To evaluate the function $|\psi_{\mathrm{vr}}\rangle$ let us write Hamiltonian H_{vr} as a sum of two nonindependent parts:

$$H_{\mathrm{vr}}=H_3+H_4, \tag{10}$$

where

$$H_3=\tfrac{1}{2}h\sum_u \omega_u\left[\left(\frac{\mathbf{p}_u}{h}\right)^2+\mathbf{q}_u{}^2\right], \tag{11}$$

$$H_4=\frac{1}{2I}(\mathbf{j}_4-\zeta\mathbf{j}_3)^2. \tag{12}$$

Here $\mathbf{q}_u$ is the vector with components $q_{u\sigma}$ ($\sigma=1,2,3$) associated with the threefold-degenerate oscillation of frequency ω_u. The $p_{u\sigma}$ are the components of $\mathbf{p}_u$.

In quantum mechanics the components $p_{u\sigma}$, $q_{u\sigma}$ must satisfy the commutation rules

$$\begin{aligned}[][q_{u\sigma},q_{u'\sigma'}]&=[p_{u\sigma},p_{u'\sigma'}]=0,\\ [q_{u\sigma},p_{u'\sigma'}]&=ih\delta_{uu'}\delta_{\sigma\sigma'}.\end{aligned} \tag{13}$$

$\mathbf{j}_4$ is an angular-momentum operator defined by the commutation rules

$$[j_{4\alpha},j_{4\beta}]=ih\sum_\gamma \epsilon_{\alpha\beta\gamma}j_{4\gamma}. \tag{14}$$

$\epsilon_{\alpha\beta\gamma}$ is the antisymmetrical unit tensor of rank three. Clearly the commutation rules (14) must also be satisfied for the component of $\mathbf{j}_4$, in the space-fixed coordinate system (X,Y,Z).

The vector $\mathbf{j}_3{}^*=\zeta\mathbf{j}_3$ is made up of a sum of Coriolis operators and is given by

$$\mathbf{j}_3{}^*=\sum_u \zeta_u(\mathbf{q}_u\times\mathbf{p}_u)=\zeta\sum_u(\mathbf{q}_u\times\mathbf{p}_u)=\zeta\mathbf{j}_3. \tag{15}$$

In the following procedure it is convenient to use the system of Euler angles shown in Fig. 1. The Eulerian angles ψ, ϑ, and φ correlate the orientation of the set of coordinates x, y, and z, fixed with respect to the molecule, to the set X, Y, and Z, fixed in space. Z is the direction of the momentum transfer $\mathbf{Q}$.

Spherical-top molecules of point group T_d, satisfying the assumption that the permutation group of the identical nuclei is isomorphous with the point group of the molecule, have at most one nondegenerate vibration ω_1, one twofold-degenerate vibration ω_2, and two triply degenerate vibrations ω_3 and ω_4.

The operator

$$H_3=H_{33}+H_{34}$$

is the Hamiltonian of a system of two independent three-dimensional isotropic oscillators having total angular momentum

$$\mathbf{j}_3=\mathbf{j}_{33}+\mathbf{j}_{34}. \tag{16}$$

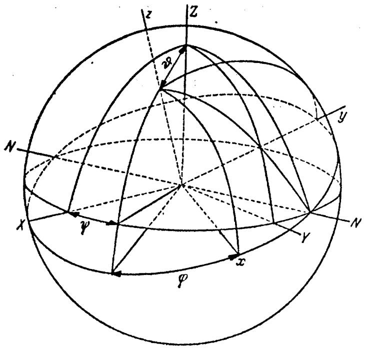

FIG. 1. Eulerian angles relating molecule-fixed axes x, y, z to space-fixed axes X, Y, Z.

The operators H_{3u}, $\mathbf{j}_{3u}{}^2$, and j_{3uz}, the last being the component of $\mathbf{j}_{3u}$ along the z axis, form a complete set of commuting observables.

The eigenvectors $|v_uj_{3u}k_{3u}\rangle$ common to these three observables are described by the quantum numbers v_u, j_{3u}, k_{3u}, and the corresponding eigenvalues are

$$\begin{aligned}H_{3u}|v_uj_{3u}k_{3u}\rangle&=h\omega_u(v_u+\tfrac{3}{2})|v_uj_{3u}k_{3u}\rangle,\\ \mathbf{j}_{3u}{}^2|v_uj_{3u}k_{3u}\rangle&=h^2j_{3u}(j_{3u}+1)|v_uj_{3u}k_{3u}\rangle,\\ j_{3uz}|v_uj_{3u}k_{3u}\rangle&=hk_{3u}|v_uj_{3u}k_{3u}\rangle,\end{aligned} \tag{17}$$

where

$$v_u=0,1,2,\cdots,\quad j_{3u}=v_u,v_u-2,\cdots,\quad \begin{Bmatrix}0 & (v_u\text{ even})\\ 1 & (v_u\text{ odd})\end{Bmatrix}$$

$$k_{3u}=-j_{3u},-j_{3u}+1,\cdots,j_{3u}-1,j_{3u},$$

and u takes the values $u=3$ and $u=4$.

In the $\{\mathbf{q}_u\}$ representation the eigenvector $|v_uj_{3u}k_{3u}\rangle$ is represented by the wave function

$$\begin{aligned}|v_uj_{3u}k_{3u}\rangle\equiv\psi_{v_u,j_{3u},k_{3u}}=&\left\{\frac{2[\frac{1}{2}(v_u-j_{3u})]!}{\Gamma[\frac{1}{2}(v_u+j_{3u})+\frac{3}{2}]}\right\}^{1/2}\\ &\times\rho_u{}^{j_{3u}}\exp(-\tfrac{1}{2}\rho_u{}^2)L_{(1/2)(v_u-j_{3u})}{}^{(j_{3u}+1/2)}(\rho_u{}^2)\\ &\times Y_{j_{3u},k_{3u}}(\vartheta_u,\chi_u),\end{aligned} \tag{18}$$

where $L_n{}^{(k)}(z)$ is the associated Laguerre polynomial of degree n,

$$L_n{}^{(k)}(z)=\frac{e^zz^{-k}}{n!}\frac{d^n}{dz^n}(e^{-z}z^{n+k}), \tag{19}$$

and $Y_{l,m}(\vartheta,\chi)$ is the spherical harmonic of degree l.

Instead of the linear coordinates q_{u1}, q_{u2}, q_{u3} we have introduced the spherical polar coordinates ρ_u, ϑ_u, and χ_u defined by

$$\begin{aligned}q_{u1}&=\rho_u\sin\vartheta_u\cos\chi_u,\\ q_{u2}&=\rho_u\sin\vartheta_u\sin\chi_u,\\ q_{u3}&=\rho_u\cos\vartheta_u.\end{aligned} \tag{20}$$

We can thus form a complete basis of eigenvectors of the Hamiltonian H_3 which should be simultaneously eigenfunctions of commuting observables $\mathbf{j}_{33}{}^2$, $\mathbf{j}_{34}{}^2$, $\mathbf{j}_3{}^2$, $\mathbf{j}_{3z}$ (Ref. 8):

$$|v_3v_4j_{33}j_{34}j_3k_3\rangle=\sum_{k_{33},k_{34}}\langle j_{33}j_{34}k_{33}k_{34}|j_3k_3\rangle|v_3j_{33}k_{33}\rangle\times|v_4j_{34}k_{34}\rangle. \quad (21)$$

On the other hand we observe that, if we neglect the Coriolis coupling factor, the Hamiltonian H_4 takes the form

$$\tilde{H}_4=\frac{1}{2I}\mathbf{j}_4{}^2,$$

which is just the Hamiltonian for a rigid rotator. $\tilde{H}_4$, j_{4z}, j_{4Z} form a complete set of commuting observables; therefore the wave function described by the three quantum numbers j_4, k_4, m_4 will be the matrix element of the irreducible representation of the rotation group

$$|j_4k_4m_4\rangle\equiv[(2j_4+1)/8\pi^2]^{1/2}D_{k_4,m_4}{}^{(j_4)}(\psi,\vartheta,\varphi), \quad (22)$$

where

$$\begin{aligned}\tilde{H}_4|j_4k_4m_4\rangle&=(\hbar^2/2I)j_4(j_4+1)|j_4k_4m_4\rangle,\\ j_{4z}|j_4k_4m_4\rangle&=\hbar k_4|j_4k_4m_4\rangle, \quad (23)\\ j_{4Z}|j_4k_4m_4\rangle&=\hbar m_4|j_4k_4m_4\rangle,\end{aligned}$$

with

$$\begin{aligned}j_4&=0,1,2,\cdots,\\ m_4&=-j_4,-j_4+1,\cdots,j_4-1,j_4,\\ k_4&=-j_4,-j_4+1,\cdots,j_4-1,j_4.\end{aligned}$$

Since $\mathbf{j}_3$ and $\mathbf{j}_4$ are commuting observables, if we follow the same procedure as for the case examined earlier, we obtain a complete basis of eigenvectors of the operator $H=H_3+H_4$ which are simultaneously eigenfunctions of the commuting observables $\mathbf{j}_4{}^2$, $\mathbf{j}_3{}^2$, $\mathbf{J}^2$, J_z, where

$$\mathbf{J}=\mathbf{j}_4-\mathbf{j}_3 \quad (24)$$

and J_z is its z component in the body-fixed system.

We have

$$\begin{aligned}|\psi_{\mathrm{vr}}\rangle&=|v_3v_4m_4j_3j_4JK\rangle\\ &=\sum_{k_3,k_4}\langle j_3j_4-k_3k_4|JK\rangle|v_3v_4j_{33}j_{34}j_3-k_3\rangle|j_4k_4m_4\rangle\\ &=\sum_{k_3,k_4;k_{33},k_{34}}\langle j_3j_4-k_3k_4|JK\rangle\langle j_{33}j_{34}k_{33}k_{34}|j_3k_3\rangle\\ &\quad\times|v_3j_{33}k_{33}\rangle|v_4j_{34}k_{34}\rangle|j_4k_4m_4\rangle. \quad (25)\end{aligned}$$

Equation (25) transforms under the operation of pure rotation $\tilde{R}$ of the proper group $\mathcal{O}$, isomorphic to the improper group T_d, according to

$$\mathcal{O}_{\tilde{R}}|v_3v_4m_4j_3j_4JK\rangle=\sum_{K'}D_{K',K}{}^{(J)}(\tilde{R})|v_3v_4m_4j_3j_4JK'\rangle,$$

where[9]

$$\begin{aligned}&D_{K',K}{}^{(J)}(\tilde{R})\\ &=\sum_{k_3',k_4';k_3,k_4}\langle j_3j_4-k_3k_4|JK\rangle\langle j_3j_4-k_3'k_4'|JK'\rangle\\ &\quad\times(-)^{k_3'-k_3}D_{k_3',k_3}{}^{*(j_3)}(\tilde{R})D_{k_4',k_4}{}^{(j_4)}(\tilde{R})\\ &=\sum_{\mathcal{K}'}\langle j_3j_4-k_3k_4|JK\rangle\langle j_3j_4-k_3'k_4'|JK'\rangle\\ &\quad\times\langle j_{33}j_{34}k_{33}k_{34}|j_3k_3\rangle\langle j_{33}j_{34}k_{33}'k_{34}'|j_3k_3'\rangle\\ &\quad\times D_{k_{33}',k_{33}}{}^{(j_{33})}(\tilde{R})D_{k_{34}',k_{34}}{}^{(j_{34})}(\tilde{R})D_{k_4',k_4}{}^{(j_4)}(\tilde{R}). \quad (26)\end{aligned}$$

The symbol $\mathcal{K}'$ stands collectively for k_3', k_4', k_3, k_4; k_{33}', k_{34}', k_{33}, k_{34}. $D_{K',K}{}^{(j)}(\tilde{R})$ are the irreducible representations of the rotation group; $\tilde{R}$ is the pure rotation associated with the point-group operation R.

The eigenvalues of the Hamiltonian H are

$$\begin{aligned}&\langle v_3v_4m_4j_3j_4JK|H|v_3v_4m_4j_3j_4JK\rangle\\ &\quad=\hbar\omega_3(v_3+\tfrac{3}{2})+\hbar\omega_4(v_4+\tfrac{3}{2})+(\hbar^2/2I)\\ &\qquad\times[j_4(j_4+1)+\zeta^2j_3(j_3+1)]-(\zeta/I)\langle v_3v_4m_4j_3j_4JK|\mathbf{j}_3\\ &\qquad\times\mathbf{j}_4|v_3v_4m_4j_3j_4JK\rangle. \quad (27)\end{aligned}$$

The last term of the right-hand side of Eq. (27) may be obtained by applying the Wigner-Eckart theorem. We find

$$\begin{aligned}&\langle v_3v_4m_4j_3j_4JK|\mathbf{j}_3\cdot\mathbf{j}_4|v_3'v_4'm_4'j_3'j_4'J'K'\rangle\\ &\quad=\delta_{JJ'}\delta_{KK'}(-1)^{J+j_3'+j_4'}\begin{Bmatrix}j_3 & 1 & j_3'\\ j_4' & J & j_4\end{Bmatrix}\\ &\qquad\times\langle v_3v_4j_3\|\mathbf{j}_3\|v_3'v_4'j_3'\rangle\cdot\langle m_4j_4\|\mathbf{j}_4\|m_4'j_4'\rangle, \quad (28)\end{aligned}$$

where

$$\begin{Bmatrix}j_1 & j_2 & j_3\\ J_1 & J_2 & J_3\end{Bmatrix}$$

is the Wigner 6-j symbol.[9,10]

The reduced matrix element for the angular momentum is given by

$$\langle\alpha j\|\mathbf{j}\|\alpha'j'\rangle=\delta_{\alpha\alpha'}\delta_{jj'}\hbar[j(j+1)(2j+1)]^{1/2}. \quad (29)$$

Therefore, the rotational and vibrational energy has the form

$$\begin{aligned}E_{\mathrm{vr}}&=\hbar\omega_3(v_3+\tfrac{3}{2})+\hbar\omega_4(v_4+\tfrac{3}{2})\\ &\quad+\frac{\hbar^2}{2I}[j_4(j_4+1)+\zeta^2j_3(j_3+1)]\\ &\quad+\frac{(-)^{J+1+j_3+j_4}}{I}\hbar^2\zeta\begin{Bmatrix}j_3 & 1 & j_3\\ j_4 & J & j_4\end{Bmatrix}\\ &\quad\times[j_3(j_3+1)(2j_3+1)]^{1/2}[j_4(j_4+1)(2j_4+1)]^{1/2}. \quad (30)\end{aligned}$$

---

[8] We use for the Clebsch-Gordan coefficients the notation of A. Messiah, *Mécanique Quantique* (Dunod Cie., Paris, 1965).

[9] M. Hamermesh, *Group Theory and its Application to Physical Problems* (Pergamon Press, Ltd., London, 1962).

[10] E. P. Wigner, *Group Theory* (Academic Press Inc., New York, 1959).

D. Total Wave Function

As stated above, the wave function $|\psi_v'\rangle$ describes the nondegenerate and the twofold-degenerate vibrations; therefore it is the product of the wave functions

$$|v_1\rangle \equiv \left[\frac{1}{\sqrt{\pi}}\frac{1}{2^{v_1}v_1!}\right]^{1/2} \exp(-\tfrac{1}{2}q_1^2)H_{v_1}(q_1) \tag{31}$$

and

$$\begin{aligned}|v_2l_2\rangle \equiv \{\pi[\tfrac{1}{2}(v_2+l_2)]![\tfrac{1}{2}(v_2-l_2)]!2^{v_2}\}^{-1/2} \\ \times\exp[-\tfrac{1}{2}(q_{21}^2+q_{22}^2)] \\ \times H_{(v_2+l_2)/2}(q_{21})H_{(v_2-l_2)/2}(q_{22}),\end{aligned} \tag{32}$$

where $l_2=v_{21}-v_{22}$, $v_2=v_{21}+v_{22}$; $l_2=v_2, v_2-2,\cdots, -v_2$, and v_{21}, v_{22} may be regarded as the quantum numbers introduced in the two-dimensional isotropic oscillator of frequency ω_2 in the representation $\{\mathbf{q}_2\}$. $H_v(z)$ is the Hermite polynomial of degree v. The wave function $|v_1\rangle$ in Eq. (31) describes the nondegenerate state of frequency ω_1, and the wave function $|v_2l_2\rangle$ in Eq. (32) describes the twofold-degenerate vibration of frequency ω_2.

Finally, the rotational and vibrational energy of the molecule, i.e., the eigenvalue of the Hamiltonian in Eq. (7), will therefore be

$$E=\hbar\omega_1(v_1+\tfrac{1}{2})+\hbar\omega_2(v_2+1)+E_{\mathrm{vr}}. \tag{33}$$

The wave functions (31) and (32) each define a representation of the symmetry group of the molecule and transform independently under the operations of the group. The representation realized by both functions is then the product of the representations of the single functions. Actually we can see in Eq. (32) that the exponential factor is invariant under all group operations, while the representation generated from the product of Hermite polynomials is the symmetric product v_2-fold by itself of the irreducible representation generated by $q_{2\sigma}(\sigma=1,2)$.[11] Analogous considerations are true for Eq. (31).

The reducible basis $|\psi_{\mathrm{t}}\rangle|\psi_{\mathrm{v}}'\rangle|\psi_{\mathrm{vr}}\rangle$ transforms under the operations R of group $\mathcal{P}$, according to

$$\begin{aligned}\mathcal{O}_R(|\psi_t\rangle|\psi_\gamma{}^{VS}\rangle|\psi_K{}^{JM}\rangle) \\ =\sum_{K,'\gamma'} D_{K,'K}{}^{(J)}(\tilde{R})D_{\gamma',\gamma}{}^{(V)}(R) \\ \times(|\psi_t\rangle|\psi_{\gamma'}{}^{VS}\rangle|\psi_{K'}{}^{JM}\rangle),\end{aligned} \tag{34}$$

where $D_{\gamma',\gamma}{}^{(V)}(R)$ denotes the irreducible representation V associated with the twofold-degenerate vibration $(\gamma\equiv l_2)$.

E. Intermediate Scattering Function and Partial Differential Cross Section

According to the procedure followed by Sinha and Venkataraman[2] we now introduce the total symmetrized wave function into Eq. (2) and perform the integration and the average over the spin parts. We obtain

$$\begin{aligned}I(\mathbf{Q},t)=\sum_{R\in\mathcal{P}}\sum_{\lambda}\chi^{*(\lambda)}(R)\sum_{\bar{\Omega}}n_\Lambda(\bar{\Omega})\langle\psi_t|\exp[i\mathbf{Q}\cdot\boldsymbol{\varrho}(t)]\exp[-i\mathbf{Q}\cdot\boldsymbol{\varrho}(0)]|\psi_t\rangle_T\sum_{j_{33},j_{34}}\sum_{\mathcal{A}}P_{\mathcal{A}}(T)\sum_{l_2',l_2}\frac{1}{\mathcal{H}}D_{l_2'l_2}{}^{(V)}(R) \\ \times\sum_{m_4}\sum_{K',K}D_{K',K}{}^{(J)}(\tilde{R})\{\langle v_1v_2l_2|\langle v_3v_4m_4j_3j_4JK|\sum_{\nu,\mu}(A_\mu A_\nu+\delta_{\nu\mu}C_\nu{}^2) \\ \times\exp[i\mathbf{Q}\cdot\boldsymbol{\sigma}_\nu(t)]\exp[-i\mathbf{Q}\cdot\boldsymbol{\sigma}_\mu(0)]|v_3v_4m_4j_3j_4JK'\rangle|v_1v_2l_2'\rangle \\ +\sum_{\substack{\nu,\mu;\ \nu\neq\mu;\\ \text{identical nuclei}}}C_\nu{}^2\langle v_1v_2l_2|\langle v_3v_4m_4j_3j_4JK|\exp[i\mathbf{Q}\cdot\boldsymbol{\sigma}_\nu(t)]\exp[-i\mathbf{Q}\cdot\boldsymbol{\sigma}_\mu(0)]Z(\bar{\Omega})|v_3v_4m_4j_3j_4JK'\rangle|v_1v_2l_2'\rangle\}.\end{aligned} \tag{35}$$

The symbol $\mathcal{A}$ stands collectively for $(v_1v_2v_3v_4j_3j_4J)$.

The position of the νth nucleus is here given by

$$\mathbf{r}_\nu=\boldsymbol{\varrho}+\boldsymbol{\sigma}_\nu, \tag{36}$$

where $\boldsymbol{\varrho}$ is the position of the center of mass of the molecule which describes the translational state only; $\boldsymbol{\sigma}_\nu$ is the vector from the center of mass to the equilibrium position of the νth nucleus in the molecule; $\chi^{(\lambda)}(R)$ is the character of R in the irreducible representation λ; and $n_\Lambda(\bar{\Omega})$ denotes the number of times the representation Λ is contained in the basis of spin functions corresponding to total nuclear spin $\bar{\Omega}$. It is $\Lambda=\lambda^*$ if the identical nuclei are bosons, or $\Lambda=(-)^{P_\alpha}\lambda^*$ if the identical nuclei are fermions. P_α is the order of the permutation α of $\mathcal{P}$; $\mathcal{H}$ is the order of group $\mathcal{P}$.

$P_{\mathcal{A}}(T)$ is the Boltzmann factor for the rotational-vibrational part of the wave function and is given by

$$P_{\mathcal{A}}(T)=\exp\left(-\frac{E}{\mathcal{K}_BT}\right)\bigg/\sum_{\mathcal{A}}n_{\mathcal{A}}\exp\left(-\frac{E}{\mathcal{K}_BT}\right), \tag{37}$$

where $\mathcal{K}_B$ is the Boltzmann constant, T the temperature of the gas, and $n_{\mathcal{A}}$ denotes the total multiplicity of the energy level $\mathcal{A}\equiv(v_1v_2v_3v_4j_3j_4J)$.

The translational matrix element has been evaluated

---

[11] L. Landau and E. Lifchitz, *Mécanique Quantique* (Mir, Moscow, 1966).

by Zemach and Glauber[1] and is given by

$$\langle\psi_t|\exp[i\mathbf{Q}\cdot\boldsymbol{\rho}(t)]\exp[-i\mathbf{Q}\cdot\boldsymbol{\rho}(0)]|\psi_t\rangle_T =\exp\left[-\frac{Q^2}{2M}(i\hbar t+t^2\mathcal{K}_BT)\right], \qquad (38)$$

where M is the mass of the molecule.

On the other hand, taking into account Eq. (3), the time dependence of the rotational-vibrational matrix elements can be made explicit, introducing a complete set of eigenfunctions of the Hamiltonian H between the two factors of the operator in (35). Let us denote these eigenfunctions by using double primes attached to the usual symbols.

$$\begin{aligned}\langle v_1|\langle v_2l_2|\langle v_3v_4m_4j_3j_4JK|&\exp[i\mathbf{Q}\cdot\boldsymbol{\sigma}_\nu(t)]\exp[-i\mathbf{Q}\cdot\boldsymbol{\sigma}_\mu(0)]|v_3v_4m_4j_3j_4JK'\rangle|v_2l_2'\rangle|v_1\rangle\\ &={\sum}''\exp\left[i\frac{E-E''}{\hbar}t\right]\langle v_1|\langle v_2l_2|\langle v_3v_4m_4j_3j_4JK|\exp[i\mathbf{Q}\cdot\boldsymbol{\sigma}_\nu(0)]|v_3''v_4''m_4''j_3''j_4''J''K''\rangle|v_2''l_2''\rangle|v_1''\rangle\\ &\qquad\times\langle v_1''|\langle v_2''l_2''|\langle v_3''v_4''m_4''j_3''j_4''J''K''|\exp[-i\mathbf{Q}\cdot\boldsymbol{\sigma}_\mu(0)]|v_3v_4m_4j_3j_4JK'\rangle|v_2l_2'\rangle|v_1\rangle, \qquad (39)\end{aligned}$$

where $\sum''$ denotes the summation over all the indices with two primes, and E'' is the energy given by Eq. (33) when the quantum numbers v_1'', v_2'', $\cdots$, J'', K'' are introduced.

To obtain the final expression for the intermediate scattering function we have to evaluate some matrix elements of the following type:

$$\langle v_1|\langle v_2l_2|\langle v_3v_4m_4j_3j_4JK|\exp[i\mathbf{Q}\cdot\boldsymbol{\sigma}_\nu(0)] \times|v_3''v_4''m_4''j_3''j_4''J''K''\rangle|v_2''l_2''\rangle|v_1''\rangle. \qquad (40)$$

For this purpose we write the position $\boldsymbol{\sigma}_\nu$ of the nucleus with respect to the center of mass as a sum of two terms:

$$\boldsymbol{\sigma}_\nu=\mathbf{u}_\nu+\mathbf{b}_\nu,$$

where $\mathbf{b}_\nu$ is the equilibrium position of the nucleus in the molecule-fixed system with origin at the center of mass and the z axis directed from the center of mass to one of the identical nuclei; and $\mathbf{u}_\nu$ is the displacement of the nucleus from the equilibrium position, due to molecular vibrations.

According to Zemach and Glauber,[1] $\mathbf{u}_\nu$ can be written as

$$\mathbf{u}_\nu=\sum_{\lambda=1}^{3N-6}\mathbf{c}_\nu^{(\lambda)}Q_\lambda, \qquad (41)$$

where Q_λ are the normal coordinates, $\mathbf{c}_\nu^{(\lambda)}$ is the amplitude vector of the νth nucleus in the λth vibrational mode and ω_λ the frequency of the mode; and N is the number of nuclei in the molecule.

By using our formalism we have

$$\mathbf{u}_\nu=\mathbf{c}_\nu^{(1)}\left(\frac{\hbar}{\omega_1}\right)^{1/2}q_1+\sum_{\sigma=1}^{2}\mathbf{c}_\nu^{(2,\sigma)}\left(\frac{\hbar}{\omega_2}\right)^{1/2}q_{2\sigma} +\sum_{\tau=1}^{3}\left[\mathbf{c}_\nu^{(3,\tau)}\left(\frac{\hbar}{\omega_3}\right)^{1/2}q_{3\tau}+\mathbf{c}_\nu^{(4,\tau)}\left(\frac{\hbar}{\omega_4}\right)^{1/2}q_{4\tau}\right]. \qquad (42)$$

If one puts

$$X_\nu^{(s,\sigma)}=(\hbar/\omega_s)^{1/2}\mathbf{Q}\cdot\mathbf{c}_\nu^{(s,\sigma)} \qquad (43)$$

and takes into account the fact that the components of $\mathbf{b}_\nu$ commute with the normal coordinates and that the latter commute with each other, then the matrix element (40) becomes

$$\begin{aligned}&\langle v_1|\langle v_2l_2|\langle v_3v_4m_4j_3j_4JK|\exp[i\mathbf{Q}\cdot\mathbf{b}_\nu]\exp[iX_\nu^{(1)}q_1]\\ &\quad\times\prod_{\sigma=1}^{2}\exp[iX_\nu^{(2,\sigma)}q_{2\sigma}]\prod_{\tau=1}^{3}\exp[iX_\nu^{(3,\tau)}q_{3\tau}]\prod_{\rho=1}^{3}\exp[iX_\nu^{(4,\rho)}q_{4\rho}]|v_3''v_4''m_4''j_3''j_4''J''K''\rangle|v_2''l_2''\rangle|v_1''\rangle\\ &=\langle v_1|\exp[iX_\nu^{(1)}q_1]|v_1''\rangle\langle v_2l_2|\prod_{\sigma=1}^{2}\exp[iX_\nu^{(2,\sigma)}q_{2\sigma}]|v_2''l_2''\rangle\\ &\qquad\times\langle v_3v_4m_4j_3j_4JK|\exp[i\mathbf{Q}\cdot\mathbf{b}_\nu]\prod_{\tau=1}^{3}\exp[iX_\nu^{(3,\tau)}q_{3\tau}]\prod_{\rho=1}^{3}\exp[iX_\nu^{(4,\rho)}q_{4\rho}]|v_3''v_4''m_4''j_3''j_4''J''K''\rangle. \qquad (44)\end{aligned}$$

Performing the explicit calculation of the matrix elements (44), we will assume in the following that $X_\nu^{(s,\sigma)}$ can be replaced by the constant value

$$(\hbar/\omega_s)^{1/2}\langle\mathbf{Q}\cdot\mathbf{c}_\nu^{(s,\sigma)}\rangle_{\text{av}}$$

obtained by taking its expectation value in the dynamical state defined by the molecular wave function $|\psi\rangle$.

Using Eq. (31) we find[12]

$$\langle v_1|\exp[iX_\nu^{(1)}q_1]|v_1''\rangle=(i/\sqrt{2})^{m_1-\bar{m}_1}[\bar{m}_1!/m_1!]^{1/2}[X_\nu^{(1)}]^{m_1-\bar{m}_1}\exp[-\tfrac{1}{4}(X_\nu^{(1)})^2]L_{\bar{m}_1}^{(m_1-\bar{m}_1)}[\tfrac{1}{2}(X_\nu^{(1)})^2]F_{v_1''}{}^{v_1}(\nu), \qquad (45)$$

---

[12] H. Bateman in *Tables of Integral Transforms* (McGraw-Hill Book Co., New York, 1954), Vols. I, II.

where

$$m_1=\max\{v_1,v_1''\}\,,\quad \bar{m}_1=\min\{v_1,v_1''\}\,,$$

and the constant $F_{v_1''}{}^{v_1}(\nu)$ is defined as

$$\begin{aligned} F_{v_1''}{}^{v_1}(\nu)&=1\,, && v_1+v_1''\text{ even}\\ F_{v_1''}{}^{v_1}(\nu)&=\sqrt{2}/X_\nu{}^{(1)}\,, && v_1+v_1''\text{ odd.} \end{aligned} \tag{46}$$

By an analogous procedure we obtain from Eq. (32)

$$\begin{aligned} \langle v_2l_2|\prod_{\sigma=1}^{2}\exp[iX_\nu{}^{(2,\sigma)}q_{2\sigma}]|v_2''l_2''\rangle&=(i/\sqrt{2})^{m_{21}+m_{22}-\bar{m}_{21}-\bar{m}_{22}}[\bar{m}_{21}!\bar{m}_{22}!/m_{21}!m_{22}!]^{1/2}[X_\nu{}^{(2,1)}]^{m_{21}-\bar{m}_{21}}[X_\nu{}^{(2,2)}]^{m_{22}-\bar{m}_{22}}\\ &\times\exp\{-\tfrac{1}{4}[(X_\nu{}^{(2,1)})^2+(X_\nu{}^{(2,2)})^2]\}L_{\bar{m}_{21}}{}^{(m_{21}-\bar{m}_{21})}[\tfrac{1}{2}(X_\nu{}^{(2,1)})^2]L_{\bar{m}_{22}}{}^{(m_{22}-\bar{m}_{22})}[\tfrac{1}{2}(X_\nu{}^{(2,2)})^2]F_{v_2''l_2''}{}^{v_2l_2}(\nu)\,, \end{aligned} \tag{47}$$

where

$$\begin{aligned} m_{21}&=\max\{\tfrac{1}{2}(v_2+l_2),\tfrac{1}{2}(v_2''+l_2'')\}\,,\quad \bar{m}_{21}=\min\{\tfrac{1}{2}(v_2+l_2),\tfrac{1}{2}(v_2''+l_2'')\}\,,\\ m_{22}&=\max\{\tfrac{1}{2}(v_2-l_2),\tfrac{1}{2}(v_2''-l_2'')\}\,,\quad \bar{m}_{22}=\min\{\tfrac{1}{2}(v_2-l_2),\tfrac{1}{2}(v_2''-l_2'')\}\,, \end{aligned}$$

and the constant $F_{v_2''l_2''}{}^{v_2l_2}(\nu)$ is defined as

$$\begin{aligned} F_{v_2''l_2''}{}^{v_2l_2}(\nu)&=1\,, && \tfrac{1}{2}[v_2+v_2''+l_2+l_2'']\text{ even}\\ &=2/(X_\nu{}^{(2,1)}+X_\nu{}^{(2,2)})\,, && \tfrac{1}{2}[v_2+v_2''+l_2+l_2'']\text{ odd.} \end{aligned} \tag{48}$$

Taking into account Eq. (25), the rotational-vibrational matrix element which is the third term in the right-hand side of Eq. (44) can be written as

$$\begin{aligned} &\langle v_3v_4m_4j_3j_4JK\,|\exp[i\mathbf{Q}\cdot\mathbf{b}_\nu]\prod_{\tau=1}^{3}\exp[iX_\nu{}^{(3,\tau)}q_{3\tau}]\prod_{\rho=1}^{3}\exp[iX_\nu{}^{(4,\rho)}q_{4\rho}]|v_3''v_4''m_4''j_3''j_4''J''K''\rangle\\ &=\sum_{\mathcal{K}''}\langle j_3''j_4''-k_3''k_4''|J''K''\rangle\langle j_{33}''j_{34}''k_{33}''k_{34}''|j_3''k_3''\rangle\langle j_3j_4-k_3k_4|JK\rangle\langle j_{33}j_{34}k_{33}k_{34}|j_3k_3\rangle\langle v_3j_{33}k_{33}|\,\langle v_4j_{34}k_{34}|\\ &\qquad\times\langle j_4k_4m_4|\exp[i\mathbf{Q}\cdot\mathbf{b}_\nu]\prod_{\tau=1}^{3}\exp[iX_\nu{}^{(3,\tau)}q_{3\tau}]\prod_{\rho=1}^{3}\exp[iX_\nu{}^{(4,\rho)}q_{4\rho}]|\,j_4''k_4''m_4''\rangle|v_4''j_{34}''k_{34}''\rangle|v_3''j_{33}''k_{33}''\rangle. \end{aligned} \tag{49}$$

The symbol $\mathcal{K}''$ stands collectively for $k_3'',k_4'',k_3,k_4;\,k_{33}'',k_{34}'',k_{33},k_{34}$.

The selection rules concerning the transition to the state indicated with two primes are included in the Clebsch-Gordan coefficients.

The matrix elements in Eq. (49) can be factorized; furthermore, with the use of Eq. (20), they become

$$\begin{aligned} \prod_{s=3,4}\langle v_sj_{3s}k_{3s}|\exp[iX_\nu{}^{(s,1)}\rho_s\sin\vartheta_s\cos\chi_s]\exp[iX_\nu{}^{(s,2)}\rho_s\sin\vartheta_s\sin\chi_s]&\\ \times\exp[iX_\nu{}^{(s,3)}\rho_s\cos\chi_s]|v_s''j_{3s}''k_{3s}''\rangle\langle j_4k_4m_4|\exp[i\mathbf{Q}\cdot\mathbf{b}_\nu]|\,j_4''k_4''m_4''\rangle&. \end{aligned} \tag{50}$$

We shall calculate separately the two factors appearing in expression (50). The calculation of the second factor can be performed taking into account the fact that[8]

$$\exp[i\mathbf{Q}\cdot\mathbf{b}_\nu]=4\pi\sum_{l=0}^{\infty}\sum_{m=-l}^{l}(-)^m i^l j_l(Qb_\nu)Y_{l,m}(\vartheta,\varphi)Y_{l,m}(\vartheta_\nu,\varphi_\nu)\,, \tag{51}$$

where $b_\nu,\vartheta_\nu,\varphi_\nu$ are the polar coordinates of the νth nucleus in the molecule-fixed system and $j_n(X)$ is the nth spherical Bessel function. We have[4]

$$\begin{aligned} \langle j_4k_4m_4|\exp[i\mathbf{Q}\cdot\mathbf{b}_\nu]|\,j_4''k_4''m_4''\rangle&=[(2j_4+1)(2j_4''+1)]^{1/2}\\ &\times(-)^{k_4-m_4}(\sqrt{4\pi})\sum_{l,m}i^l j_l(Qb_\nu)Y_{l,m}(\vartheta_\nu,\varphi_\nu)[1/(2l+1)^{1/2}]\langle j_4''j_4k_4''-k_4|lm\rangle\langle j_4''j_4m_4''-m_4|l0\rangle. \end{aligned} \tag{52}$$

Now we calculate the first factor in the expression (50), which is a more complex case. It may be verified that on introducing the expression of spherical harmonics in terms of associated Legendre functions $P_{j_{3s},k_{3s}}(\cos\vartheta_s)$, this factor will be explicitly

$$\begin{aligned}
&\langle v_s j_{3s} k_{3s} | \exp[iX_\nu^{(s,1)}\rho_s \sin\vartheta_s \cos\chi_s] \exp[iX_\nu^{(s,2)}\rho_s \sin\vartheta_s \sin\chi_s] \exp[iX_\nu^{(s,3)}\rho_s \cos\vartheta_s] | v_s'' j_{3s}'' k_{3s}'' \rangle \\
&= \frac{1}{2\pi}(-)^{k_{3s}+k_{3s}''}[(2j_{3s}+1)(2j_{3s}''+1)]^{1/2} \left\{ \frac{[\frac{1}{2}(v_s-j_{3s})]![\frac{1}{2}(v_s''-j_{3s}'')]!}{\Gamma[\frac{1}{2}(v_s+j_{3s})+\frac{3}{2}]\Gamma[\frac{1}{2}(v_s''+j_{3s}'')+\frac{3}{2}]} \frac{(j_{3s}-k_{3s})!(j_{3s}''-k_{3s}'')!}{(j_{3s}+k_{3s})!(j_{3s}''+k_{3s}'')!} \right\}^{1/2} \\
&\times \int_0^\infty d\rho_s \rho_s^{j_{3s}+j_{3s}''+2} \exp(-\rho_s^2) L_{(1/2)(v_s-j_{3s})}^{(j_{3s}+1/2)}(\rho_s^2) L_{(1/2)(v_s''-j_{3s}'')}^{(j_{3s}''+1/2)}(\rho_s^2) \int_0^\pi d\vartheta_s P_{j_{3s},k_{3s}}(\cos\vartheta_s) \\
&\times P_{j_{3s}'',k_{3s}''}(\cos\vartheta_s) \exp[iX_\nu^{(s,3)}\rho_s \cos\vartheta_s] \sin\vartheta_s \int_0^{2\pi} d\chi_s \exp[iX_\nu^{(s,1)}\rho_s \sin\vartheta_s \cos\chi_s] \exp[iX_\nu^{(s,2)}\rho_s \sin\vartheta_s \sin\chi_s] \\
&\times \exp[i(k_{3s}''-k_{3s})\chi_s]. \qquad (53)
\end{aligned}$$

We have[13]

$$\begin{aligned}
&\int_0^{2\pi} \exp\{i[X_\nu^{(s,1)}\rho_s \sin\vartheta_s \cos\chi_s + X_\nu^{(s,2)}\rho_s \sin\vartheta_s \sin\chi_s + (k_{3s}''-k_{3s})\chi_s]\} d\chi_s \\
&= 2\pi i^{k_{3s}''-k_{3s}} \exp\left[i(k_{3s}''-k_{3s}) \arctan\frac{X_\nu^{(s,2)}}{X_\nu^{(s,1)}} \right] J_{k_{3s}''-k_{3s}}\{[(X_\nu^{(s,1)})^2+(X_\nu^{(s,2)})^2]^{1/2}\rho_s \sin\vartheta_s\}. \qquad (54)
\end{aligned}$$

Omitting from consideration the constant that in Eq. (54) multiplies the Bessel function

$$J_{k_{3s}''-k_{3s}}\{[(X_\nu^{(s,1)})^2+(X_\nu^{(s,2)})^2]^{1/2}\rho_s \sin\vartheta_s\},$$

the integration over ϑ_s gives

$$\begin{aligned}
&\int_0^\pi P_{j_{3s},k_{3s}}(\cos\vartheta_s) P_{j_{3s}'',k_{3s}''}(\cos\vartheta_s) \exp[iX_\nu^{(s,3)}\rho_s \cos\vartheta_s] J_{k_{3s}''-k_{3s}}\{[(X_\nu^{(s,1)})^2+(X_\nu^{(s,2)})^2]^{1/2}\rho_s \sin\vartheta_s\} \sin\vartheta_s d\vartheta_s \\
&= (\sqrt{\tfrac{1}{2}\pi}) 2^{-(j_{3s}+j_{3s}'')} \sum_{\alpha=0}^{\leqslant \frac{1}{2}(j_{3s}-|k_{3s}|)} \sum_{\beta=0}^{\leqslant \frac{1}{2}(j_{3s}''-|k_{3s}''|)} (-)^{\alpha+\beta} \frac{1}{\alpha!\beta!} \frac{(2j_{3s}-2\alpha)!(2j_{3s}''-2\beta)!}{(j_{3s}-\alpha)!(j_{3s}''-\beta)!(j_{3s}-|k_{3s}|-2\alpha)!(j_{3s}''-|k_{3s}''|-2\beta)!} \\
&\times \{[1+(-)^{j_{3s}+j_{3s}''-|k_{3s}|-|k_{3s}''|}] G_{j_{3s},|k_{3s}|,\alpha}^{j_{3s}'',|k_{3s}''|,\beta}(\rho_s, X_\nu^{(s,\sigma)}) \\
&\qquad + [1-(-)^{j_{3s}+j_{3s}''-|k_{3s}|-|k_{3s}''|}] \tilde{G}_{j_{3s},|k_{3s}|,\alpha}^{j_{3s}'',|k_{3s}''|,\beta}(\rho_s, X_\nu^{(s,\sigma)})\}, \qquad (55)
\end{aligned}$$

where

$$\begin{aligned}
G_{j_{3s},|k_{3s}|,\alpha}^{j_{3s}'',|k_{3s}''|,\beta}(\rho_s, X_\nu^{(s,\sigma)}) &= M_{k_{3s},k_{3s}''} (\xi_s^\nu)^{-h_{3s}} (\eta_s^\nu)^{1-n/2} \left(\frac{d}{d\xi_s^\nu}\right)^{\bar{h}_{3s}} \left(\frac{d}{d\eta_s^\nu}\right)^{n/2} \\
&\times \{(\xi_s^\nu)^{2h_{3s}} (\eta_s^\nu)^{n-1} [(\xi_s^\nu)^2+(\eta_s^\nu)^2]^{-(1/2)[(n+1)/2+h_{3s}]} J_{(n+1)/2+h_{3s}}[(\xi_s^\nu)^2+(\eta_s^\nu)^2]^{1/2}\} \qquad (56)
\end{aligned}$$

and

$$\begin{aligned}
\tilde{G}_{j_{3s},|k_{3s}|,\alpha}^{j_{3s}'',|k_{3s}''|,\beta}(\rho_s, X_\nu^{(s,\sigma)}) &= M_{k_{3s},k_{3s}''} i (\xi_s^\nu)^{-h_{3s}} (\eta_s^\nu)^{(1-n)/2} \left(\frac{d}{d\xi_s^\nu}\right)^{\bar{h}_{3s}} \left(\frac{d}{d\eta_s^\nu}\right)^{(n-1)/2} \\
&\times \{(\xi_s^\nu)^{2h_{3s}} (\eta_s^\nu)^{n} [(\xi_s^\nu)^2+(\eta_s^\nu)^2]^{-(1/2)[(n+2)/2+h_{3s}]} J_{(n+2)/2+h_{3s}}[(\xi_s^\nu)^2+(\eta_s^\nu)^2]^{1/2}\}. \qquad (57)
\end{aligned}$$

We have used the following notation:

$$\begin{aligned}
\xi_s^\nu &= [(X_\nu^{(s,1)})^2+(X_\nu^{(s,2)})^2]^{1/2}\rho_s, \\
\eta_s^\nu &= X_\nu^{(s,3)}\cdot\rho_s, \\
n &= j_{3s}+j_{3s}''-|k_{3s}|-|k_{3s}''|-2\alpha-2\beta, \\
h_{3s} &= \max\{|k_{3s}|,|k_{3s}''|\}, && \text{if } k_{3s}\cdot k_{3s}'' \geqslant 0 \\
&= |k_{3s}|+|k_{3s}''|, && \text{if } k_{3s}\cdot k_{3s}'' < 0; \\
\bar{h}_{3s} &= \min\{|k_{3s}|,|k_{3s}''|\}, && \text{if } k_{3s}\cdot k_{3s}'' \geqslant 0 \\
&= 0, && \text{if } k_{3s}\cdot k_{3s}'' < 0; \\
M_{k_{3s},k_{3s}''} &= 1, && \text{if } k_{3s}\geqslant 0,\ k_{3s}''\geqslant 0 \\
&= (-)^{k_{3s}}(j_{3s}''+k_{3s}'')!/(j_{3s}''-k_{3s}'')!, && \text{if } k_{3s}>0,\ k_{3s}''<0 \\
&= (-)^{k_{3s}''}(j_{3s}+k_{3s})!/(j_{3s}-k_{3s})!, && \text{if } k_{3s}<0,\ k_{3s}''>0 \\
&= (-)^{k_{3s}+k_{3s}''}(j_{3s}+k_{3s})!(j_{3s}''+k_{3s}'')!/[(j_{3s}-k_{3s})!(j_{3s}''-k_{3s}'')!], && \text{if } k_{3s}<0,\ k_{3s}''<0. \qquad (58)
\end{aligned}$$

---

[13] W. Gröbner and N. Hofreiter, *Integraletafel—Bestimmte Integrale* (Springer-Verlag, Göttingen, 1950).

In order to calculate the right-hand side of Eq. (55) we have used the polynomial form of the associated Legendre functions and the Fourier cosine and sine transforms of functions of the type[12]

$$\begin{aligned} f(X) &= X^k(1-X^2)^{(1/2)\nu+m}J_\nu[a(1-X^2)^{1/2}], \quad \text{for} \quad 0\leqslant X\leqslant 1 \\ &= 0, \quad \text{for} \quad 1<X<\infty . \end{aligned} \tag{59}$$

Finally, still neglecting the constants, by integration over ρ_s we obtain

$$\begin{aligned} &\int_0^\infty \rho_s^{j_{3s}''+j_{3s}+2}\exp(-\rho_s^2)L_{(1/2)(v_s-j_{3s})}^{(j_{3s}+1/2)}(\rho_s^2)L_{(1/2)(v_s''-j_{3s}'')}^{(j_{3s}''+1/2)}(\rho_s^2)\{[1+(-)^{j_{3s}+j_{3s}''-|k_{3s}|-|k_{3s}''|}] \\ &\qquad \times G_{j_{3s},|k_{3s}|,\alpha}{}^{j_{3s},''|k_{3s}''|,\beta}(\rho_s,X_\nu^{(s,\sigma)})+[1-(-)^{j_{3s}+j_{3s}''-|k_{3s}|-|k_{3s}''|}]\tilde{G}_{j_{3s},|k_{3s}''|,\alpha}{}^{j_{3s}'',|k_{3s}''|,\beta}(\rho_s,X_\nu^{(s,\sigma)})\}d\rho_s \\ &= \sum_{\dot{\alpha}=0}^{(1/2)(v_s-j_{3s})}\ \sum_{\dot{\beta}=0}^{(1/2)(v_s''-j_{3s}'')} \frac{(-)^{\dot{\alpha}+\dot{\beta}}M_{k_{3s},k_{3s}''}}{\dot{\alpha}!\dot{\beta}!}\ \frac{\Gamma[\frac{1}{2}(v_s+j_{3s}+3)]}{\Gamma(j_{3s}+\dot{\alpha}+\frac{3}{2})[\frac{1}{2}(v_s-j_{3s})-\dot{\alpha}]!}\ \frac{\Gamma[\frac{1}{2}(v_s''+j_{3s}''+3)]}{\Gamma(j_{3s}''+\dot{\beta}+\frac{3}{2})[\frac{1}{2}(v_s''-j_{3s}'')-\dot{\beta}]!} \\ &\qquad \times\Bigg\{[1+(-)^{j_{3s}+j_{3s}''-|k_{3s}|-|k_{3s}''|}]\int_0^\infty \rho_s^{j_{3s}+j_{3s}''+2\dot{\alpha}+2\beta+2}\exp(-\rho_s^2)(\xi_s^\nu)^{-h_{3s}}(\eta_s^\nu)^{1-n/2}\left(\frac{d}{d\xi_s^\nu}\right)^{\bar{h}_{3s}}\left(\frac{d}{d\eta_s^\nu}\right)^{n/2} \\ &\qquad \times\{(\xi_s^\nu)^{2h_{3s}}(\eta_s^\nu)^{n-1}[(\xi_s^\nu)^2+(\eta_s^\nu)^2]^{-(1/2)[(n+1)/2+h_{3s}]}J_{(n+1)/2+h_{3s}}[(\xi_s^\nu)^2+(\eta_s^\nu)^2]^{1/2}\}d\rho_s \\ &\qquad +[1-(-1)^{j_{3s}+j_{3s}''-|k_{3s}|-|k_{3s}''|}]i\int_0^\infty \rho_s^{j_{3s}+j_{3s}''+2\dot{\alpha}+2\dot{\beta}+2}\exp(-\rho_s^2)(\xi_s^\nu)^{-h_{3s}}(\eta_s^\nu)(1-n)/2\left(\frac{d}{d\xi_s^\nu}\right)^{\bar{h}_{3s}}\left(\frac{d}{d\eta_s^\nu}\right)^{(n-1)/2} \\ &\qquad\qquad \times\{(\xi_s^\nu)^{2h_{3s}}(\eta_s^\nu)^{n}[(\xi_s^\nu)^2+(\eta_s^\nu)^2]^{-(1/2)[(n+2)/2+h_{3s}]}J_{(n+2)/2+h_{3s}}[(\xi_s^\nu)^2+(\eta_s^\nu)^2]^{1/2}\}d\rho_s\Bigg\}. \end{aligned} \tag{60}$$

Since ξ_s^ν and η_s^ν are proportional to ρ_s [see relations (58)] the two integrals in the second part of the right-hand side of Eq. (60), leaving out a constant A_s^ν, have the general form

$$\begin{aligned} I_p^{\bar{r}}(\mathfrak{F}) &= \int_0^\infty (\xi_s^\nu)^{\bar{r}}\exp[-a(\xi_s^\nu)^2][d^p/d(\xi_s^\nu)^p]\{(\eta_s^\nu)^k[d^m/d(\eta_s^\nu)^m]\mathfrak{f}(\xi_s^\nu,\eta_s^\nu)\}d\xi_s^\nu \\ &= \int_0^\infty (\xi_s^\nu)^{\bar{r}}\exp[-a(\xi_s^\nu)^2][d^p/d(\xi_s^\nu)^p][\mathfrak{F}(\xi_s^\nu,\eta_s^\nu)]d\xi_s^\nu , \end{aligned} \tag{61}$$

where

$$\bar{r}=j_{3s}+j_{3s}''+2\dot{\alpha}+2\dot{\beta}-h_{3s}+2, \quad p=\bar{h}_{3s}.$$

(We always have $\bar{r}\geqslant 2$, $\bar{r}>p$.) In the first integral on the right side of Eq. (60),

$$k=1-\tfrac{1}{2}n, \quad m=\tfrac{1}{2}n.$$

In the second integral on the right-hand side of Eq. (60),

$$\begin{aligned} &k=\tfrac{1}{2}(1-n), \quad m=\tfrac{1}{2}(n-1), \\ &A_s^\nu=[(X_\nu^{(s,1)})^2+(X_\nu^{(s,2)})^2]^{-1/2(j_{3s}+j_{3s}''+2\dot{\alpha}+2\dot{\beta}+3)}, \\ &a=[(X_\nu^{(s,1)})^2+(X_\nu^{(s,2)})^2]^{-1}. \end{aligned}$$

The integral appearing in Eq. (61) gives the recursion formula

$$I_p^{\bar{r}}=-\bar{r}I_{p-1}^{\bar{r}-1}+2aI_{p-1}^{\bar{r}+1}. \tag{62}$$

Therefore $I_p^{\bar{r}}$ can be calculated for any value with the lower index p, if we can calculate for any value of the upper index q the new integral

$$I_0^q=\int_0^\infty (\xi_s^\nu)^q\exp[-a(\xi_s^\nu)^2]\mathfrak{F}(\xi_s^\nu,\eta_s^\nu)d\xi_s^\nu. \tag{63}$$

In our case this integral takes the form

$$\begin{aligned} I_0^q &= B_q\int_0^\infty (\eta_s^\nu)^r\exp[-b(\eta_s^\nu)^2] \\ &\qquad \times[d^m/d(\eta_s^\nu)^m]\mathfrak{f}(\xi_s^\nu\eta_s^\nu)d\eta_s^\nu \\ &= B_q\bar{I}_m^r , \end{aligned} \tag{64}$$

where

$$\begin{aligned} &B_q=\{[(X_\nu^{(s,1)})^2+(X_\nu^{(s,2)})^2]^{1/2}/X_\nu^{(s,3)}\}^{q+1}, \\ &b=(X_\nu^{(s,3)})^{-2}, \\ &r=q+k, \end{aligned}$$

and $q\geqslant\bar{r}-p$, $r\geqslant 3$, $r>m$ always.

The integral appearing in Eq. (64) satisfies a recursion formula analogous to that of Eq. (62):

$$\bar{I}_m{}^r = -r\bar{I}_{m-1}{}^{r-1} + 2b\bar{I}_{m-1}{}^{r+1}, \tag{65}$$

and it can be calculated for any value of the lower index m if we can calculate for any value of the upper index v the integral

$$\bar{I}_0{}^v = C_v \int_0^\infty \rho_s{}^w \exp(-\rho_s{}^2)\, J_u(c\rho_s) d\rho_s = C_v \tilde{I}_u{}^w, \tag{66}$$

where

$$c = [(X_\nu{}^{(s,1)})^2 + (X_\nu{}^{(s,2)})^2 + (X_\nu{}^{(s,3)})^2]^{1/2},$$
$$v \geqslant r - m.$$

For the first integral on the right-hand side of Eq. (60) we have

$$u = \tfrac{1}{2}(n+1) + h_{3s},$$
$$w = v + h_{3s} + \tfrac{1}{2}(n-3),$$
$$C_v = (X_\nu{}^{(s,3)})^{v+n}[(X_\nu{}^{(s,1)})^2 + (X_\nu{}^{(s,2)})^2]^{h_{3s}} \times [(X_\nu{}^{(s,1)})^2 + (X_\nu{}^{(s,2)})^2 + (X_\nu{}^{(s,3)})^2]^{-1/2[(n+1)/2+h_{3s}]},$$

while for the second integral on the right-hand side of Eq. (60), we have

$$u = \tfrac{1}{2}(n+2) + h_{3s},$$
$$w = v + h_{3s} + \tfrac{1}{2}(n-2),$$
$$C_v = (X_\nu{}^{(s,3)})^{v+n+1}[(X_\nu{}^{(s,1)})^2 + (X_\nu{}^{(s,2)})^2]^{h_{3s}} \times [(X_\nu{}^{(s,1)})^2 + (X_\nu{}^{(s,2)})^2 + (X_\nu{}^{(s,3)})^2]^{-1/2[(n+2)/2+h_{3s}]}.$$

The integral on the right-hand side of Eq. (66) has the value[13]

$$\tilde{I}_u{}^w = \frac{c^u \Gamma(\frac{1}{2}(u+w+1))}{2^{u+1}\Gamma(u+1)} \times \exp(-\tfrac{1}{4}c^2) M(\tfrac{1}{2}(u-w+1); u+1; \tfrac{1}{4}c^2), \tag{67}$$

where $M(a;b;z)$ is the hypergeometric confluent function defined by the Pochammer-Kummer series:

$$M(a;b;z) = \sum_{\nu=0}^{\infty} \frac{\Gamma(a+\nu)\Gamma(b)}{\Gamma(a)\Gamma(b+\nu)} \frac{z^\nu}{\nu!}. \tag{68}$$

Finally the matrix element (49) is given by

$$\begin{aligned}
&\langle v_3 v_4 m_4 j_3 j_4 JK | \exp[i\mathbf{Q}\cdot\mathbf{b}_\nu] \prod_{s=3,4} \prod_{\sigma=1}^{3} \exp[iX_\nu{}^{(s,\sigma)} q_{s\sigma}] | v_3'' v_4'' m_4'' j_3'' j_4'' J'' K'' \rangle \\
&= \sum_{\substack{k_3,k_4,k_{33},k_{34};\\ k_3'',k_4'',k_{33}'',k_{34}''\\ m,l}} \pi^{3/2} i^l \langle j_3'' j_4'' - k_3'' k_4'' | J'' K'' \rangle \langle j_{33}'' j_{34}'' k_{33}'' k_{34}'' | j_3'' k_3'' \rangle \langle j_3 j_4 - k_3 k_4 | JK \rangle \langle j_{33} j_{34} k_{33} k_{34} | j_3 k_3 \rangle \\
&\quad \langle j_4'' j_4 k_4'' - k_4 | lm \rangle \langle j_4'' j_4 m_4'' - m_4 | l0 \rangle (-)^{k_4 - m_4} \left[\frac{(2j_4+1)(2j_4''+1)}{(2l+1)}\right]^{1/2} j_l(Qb_\nu) Y_{lm}(\vartheta_\nu, \varphi_\nu) \\
&\quad \times \prod_{s=3,4} \Bigg(\sum_{\alpha,\dot{\alpha};\beta,\dot{\beta}} (-)^{k_{3s}''+k_{3s}+\alpha+\dot{\alpha}+\dot{\beta}+\beta}\, i^{k_{3s}''-k_{3s}}\, 2^{-(j_{3s}+j_{3s}'')} [(2j_{3s}+1)(2j_{3s}''+1)]^{1/2} \exp\left[i(k_{3s}''-k_{3s}) \arctan\frac{X_\nu{}^{(s,2)}}{X_\nu{}^{(s,1)}}\right] \\
&\quad \times \left\{\frac{[\frac{1}{2}(v_s - j_{3s})]!\,[\frac{1}{2}(v_s'' - j_{3s}'')]!\,(j_{3s}-k_{3s})!\,(j_{3s}''-k_{3s}'')!\,\Gamma[\frac{1}{2}(v_s+j_{3s})+\frac{3}{2}]\,\Gamma[\frac{1}{2}(v_s''+j_{3s}')+\frac{3}{2}]}{(j_{3s}+k_{3s})!\,(j_{3s}''+k_{3s}'')!}\right\}^{1/2} \\
&\quad \times M_{k_{3s},k_{3s}''} A_s{}^\nu \frac{(2j_{3s}-2\alpha)!\,(2j_{3s}''-2\beta)!}{\dot{\alpha}!\,\alpha!\,\dot{\beta}!\,\beta!\,(j_{3s}-\alpha)!\,(j_{3s}''-\beta)!\,(j_{3s}-|k_{3s}|-2\alpha)!\,(j_{3s}''-|k_{3s}''|-2\beta)!} \\
&\quad \times \frac{1}{\Gamma(j_{3s}+\dot{\alpha}+\frac{3}{2})\,\Gamma(j_{3s}''+\dot{\beta}+\frac{3}{2})\,[\frac{1}{2}(v_s-j_{3s})+\dot{\alpha}]!\,[\frac{1}{2}(v_s''-j_{3s}'')+\dot{\beta}]!} \\
&\qquad \times \{[1+(-1)^{j_{3s}+j_{3s}''-|k_{3s}|-|k_{3s}''|}] I_p{}^{\bar{r}}(\mathfrak{F}_{1s}{}^\nu) + i[1-(-)^{j_{3s}+j_{3s}''-|k_{3s}|-|k_{3s}''|}] I_p{}^{\bar{r}}(\mathfrak{F}_{2s}{}^\nu)\} \Bigg).
\end{aligned} \tag{69}$$

Equation (61) defines the integral $I_p{}^{\bar{r}}$; here it is

$$\mathfrak{F}_{1s}{}^\nu = (\eta_s{}^\nu)^{1-n/2} (d/d\eta_s{}^\nu)^{n/2} \{(\xi_s{}^\nu)^{2h_{3s}} (\eta_s{}^\nu)^{n-1} [(\xi_s{}^\nu)^2 + (\eta_s{}^\nu)^2]^{-(1/2)[(n+1)/2+h_{3s}]} J_{(n+1)/2+h_{3s}} [(\xi_s{}^\nu)^2 + (\eta_s{}^\nu)^2]^{1/2}\},$$
$$\mathfrak{F}_{2s}{}^\nu = (\eta_s{}^\nu)^{(1-n)/2} (d/d\eta_s{}^\nu)^{(n-1)/2} \{(\xi_s{}^\nu)^{2h_{3s}} (\eta_s{}^\nu)^{n} [(\xi_s{}^\nu)^2 + (\eta_s{}^\nu)^2]^{-(1/2)[(n+2)/2+h_{3s}]} J_{(n+2)/2+h_{3s}} [(\xi_s{}^\nu)^2 + (\eta_s{}^\nu)^2]^{1/2}\}.$$

By using Eqs. (35), (38), and (39) and taking the Fourier transform of the intermediate scattering function as indicated in Eq. (1), we obtain the following expression for the scattering cross section per unit solid angle and unit interval of outgoing neutron energy:

$$
\begin{aligned}
d^2\sigma/d\Omega dE=&(1/2\pi\hbar)(k/k_0)(2\pi M/Q^2\mathcal{K}_BT)^{1/2}\sum_{R\in\mathcal{P}}\sum_{\lambda}\chi^{*(\lambda)}(R)\sum_{\bar{\Omega}}n_\Lambda(\bar{\Omega})\sum_{j_{33},j_{3u}}\sum_{\mathcal{A}}P_{\mathcal{A}}(T)\\
&\times\sum_{l_2',l_2}(1/\mathcal{H})D_{l_2',l_2}{}^{(V)}(R)\sum_{m_4}\sum_{K',K}D_{K',K}{}^{(J)}(\tilde{R})\Big[\sum_{\nu,\mu}(A_\nu A_\mu+\delta_{\nu\mu}C_\nu{}^2)+\sum_{\substack{\nu,\mu;\ \nu\neq\mu\\ \text{nuclei identical}}}C_\nu{}^2Z(\bar{\Omega})\Big]\\
&\times\sum{}''\exp[-(\omega\hbar+\epsilon)^2M/(2Q^2\mathcal{K}_BT\hbar^2)]\langle v_1|\exp(iX_\nu{}^{(1)}q_1)|v_1''\rangle\langle v_1''|\exp(-iX_\mu{}^{(1)}q_1)|v_1\rangle\\
&\times\langle v_2l_2|\prod_{\sigma=1}^{2}\exp(iX_\nu{}^{(2,\sigma)}q_{2\sigma})|v_2''l_2''\rangle\langle v_2''l_2''|\prod_{\sigma'=1}^{2}\exp(-iX_\mu{}^{(2,\sigma')}q_{2\sigma'})|v_2l_2'\rangle\langle v_3v_4m_4j_3j_4JK|\\
&\times\exp[i\mathbf{Q}\cdot\mathbf{b}_\nu]\prod_{s=3,4}\prod_{\sigma''=1}^{3}\exp(iX_\nu{}^{(s,\sigma'')}q_{s\sigma''})|v_3''v_4''m_4''j_3''j_4''J''K''\rangle\\
&\times\langle v_3''v_4''m_4''j_3''j_4''J''K''|\exp[-i\mathbf{Q}\cdot\mathbf{b}_\mu]\prod_{s'=3,4}\prod_{\sigma'''=1}^{3}\exp(-iX_\mu{}^{(s',\sigma''')}q_{s'\sigma'''})|v_3v_4m_4j_3j_4LK'\rangle,\qquad(70)
\end{aligned}
$$

where

$$\epsilon=E-E''-Q^2\hbar^2/2M.$$

The matrix elements appearing in Eq. (70) are explicitly given in Eqs. (45), (47), and (69). In writing down these matrix elements, the orthogonality properties of Clebsch-Gordan coefficients must be kept in mind.

III. SUMMARY

The present paper presents the results of the computations on the partial differential cross section for scattering of slow neutrons by molecules in the general case including rotational-vibrational interaction. A precise and explicit formula has been derived for spherical-top molecules of point group T_d.

When it is assumed that the molecules are in their ground vibrational state both before and after the scattering process, formula (70) coincides with the formula of Sinha and Venkataraman. This is consistent with Eqs. (17) and (24); indeed, if $v_3=v_4=0$, the angular momentum resulting from the Coriolis coupling is zero and $\mathbf{j}_4=\mathbf{J}$.

Actually Eq. (70) has a complicated form, but, by examining the Boltzmann factor, we can see that only the vibrational terms corresponding to the very low quantum numbers give an appreciable contribution. In the case of methane, for instance, where $\nu_1=2914$, $\nu_2=1526$, $\nu_3=3020$, and $\nu_4=1306$ cm^{-1}, an appreciable contribution to Eq. (70) is given only by the terms for which all the v_i's are zero ($i=1,2,3,4$), or only one of the v_i is equal to 1 while all the others are zero.

Moreover, we point out that Eq. (70) and the rotational-vibrational matrix element have been formulated in such a way as to facilitate the writing of a computation program for a high-speed digital computer. Further studies on this subject are in progress.