

Scattering of slow neutrons by rotating systems of identical nuclei

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Slow neutron scattering (SNS) by rotationally tunneling symmetrical systems of identical nuclei is considered. The doubly differential cross section of noncoherent scattering is calculated with account taken of the identity principle which establishes a correlation between the spin and coordinate (rotational) parts of the wave function, and is expressed in terms of irreducible time correlation functions (ITCF) in which the averaging is over states belonging only to one irreducible representation of the symmetry group of the scattering system. The calculation of the ITCF is carried out for groups of three spin-1/2 nuclei with that tunnel in a retarding potential in a solid around the C_3 axis. The interaction of the rotation with the phonons is assumed to be weak. The temperature dependence of the SNS spectrum is considered in the region of small energy transfers, and the collapse of the tunnel multiplet is explained.

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1. INTRODUCTION

In the theory of slow neutron scattering (SNS) by molecules, which was developed by van Hove¹ and by Zemach and Glauber,² (see also Refs. 3-5), the doubly differential scattering cross section (DDSS) is expressed in terms of the temporal correlation functions (TCF) of the molecular motion. The identity principle, which establishes the correlation between the spin and the coordinate parts of the wave function of the scattering system, was not taken into account in Refs. 1-5. However, when neutrons are scattered by symmetrical systems of identical nuclei in gases and in solids at low temperatures, when the motion is essentially of quantum character, the influence of the exclusion principle may turn out to be appreciable. The clearest example of this is scattering by molecular hydrogen.⁶ Zemach and Glauber⁷ attempted to take into consideration the exclusion principle by averaging the spin-dependent factor in the DDSS over the spin wave functions, which are characterized by a definite value of the total spin and transform like the basis functions of the irreducible representation of the permutation group of identical nuclei, and took into account at the same time the only averaged correlation between the spin and coordinate parts of the wave functions. Hama and Miyagi⁸ investigated the influence of this correlation on the SNS in methane gas and found that the corrections to the Zemach-Glauber approximation are important at temperatures below 10 K. Recent advances in the experimental techniques⁹ have made it possible to obtain high resolution, sufficient for the investigation of small energy-level splittings due to the rotational tunneling of molecules and their fragments in solids.¹⁰⁻¹⁴ Hüller¹⁴ considered SNS by four-proton tetrahedral groups (such as CH_4 and NH_4^+) that tunnel in a crystal low-symmetry potential, and pointed out the importance of correlation effects in SNS in molecular crystals at temperatures ≤ 80 K. In view of the neglect of relaxation processes due to the coupling of the rotation with other degrees of freedom of the crystal, Hüller's theory does not describe the temperature dependence of the SNS spectra, and in particular the tunnel-multiplet collapse observed in Refs. 12 and 13.

In the present paper, the DDSS on symmetrical system of identical nuclei is expressed in terms of the irreducible TCF (ITCF), which were introduced in Ref. 15, and in which the averaging is over states belonging to only one irreducible representation of the symmetry group of the system. The general relations are applicable to SNS in a polycrystal that contains three-proton groups that rotate about a symmetry axis in a retarding potential (the methyl and ammonia groups in molecular crystals). The choice of this system is due to by its great abundance, simplicity, and the fact that it has been relatively well investigated.^{10, 13, 16-19} It is shown with the aid of the considered example that, depending on the singularities of the interaction of the rotation with the phonons, the temperature dependence of the tunnel spectrum of the SNS can vary. Conditions under which a collapse of the tunnel multiplet with increasing scatterer temperature takes place.

2. CROSS SECTION OF NONCOHERENT SCATTERING

We consider SNS in a crystal, assuming for simplicity that all the tops are translationally equivalent and that the scattering is only by nuclei of one sort with spin I . We label the nuclei by a double index na , where $n=1, 2, \dots, N$ is the number of the top and a is the number of the nucleus in it. The incident flux of neutrons is assumed to be unpolarized. The DDSS of the noncoherent scattering by the unpolarized sample, obtained from the general formula for the cross section,⁷ can be represented in the form

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{B^2}{I(I+1)} \frac{k'}{2\pi k} \sum_{na_0} \sum_{\mu=-1}^1 (-1)^\mu \int_{-\infty}^{\infty} dt e^{i\omega t} \quad (2.1)$$

$$\times \text{Sp}[\rho_{eq} I_{1\mu}(na_1) \exp(i\mathbf{Q}\mathbf{R}_{na_1}) e^{iFt} I_{1-\mu}(na_2) \exp(-i\mathbf{Q}\mathbf{R}_{na_2}) e^{-iFt}],$$

where Ω is the solid angle, $\hbar\omega$ is the energy transferred to the neutron, $k = |\mathbf{k}|$, $k' = |\mathbf{k}'|$, \mathbf{k} and \mathbf{k}' are respectively the momenta of the incident and scattered neutrons, ρ_{eq} is the scatterer density matrix, B is the amplitude of noncoherent scattering by one nucleus, \mathbf{R}_{na} is the radius vector of the nucleus with number na , $\mathbf{Q} = \mathbf{k}' - \mathbf{k}$ is the momentum transfer, $\hbar F$ is the Hamiltonian of the scatterer, $I_{1\mu}(na)$, $\mu = 0, \pm 1$ are the spherical components of the spin operators:

$$I_{10}(na) = I_{na}, \quad I_{1\pm 1}(na) = \mp (I_{na\pm 1} I_{na\mp 1}) / \sqrt{2}. \quad (2.2)$$

In the derivation of (2.1) we took into account the fact that the spin states of the nuclei in different tops are not correlated.

Being interested in the contribution made to the SNS by the rotation, we confine ourselves to the lowest term in the expansion of $\exp(i\mathbf{Q} \cdot \mathbf{R}_{na})$ in powers of the vibrational (translational) displacements of the nuclei from the equilibrium position, introducing by the same token the rigid-top approximation. The radius vector \mathbf{R}_{na} can now be regarded as a vector directed from the symmetry center of the n -th top to the na -th nucleus. The resolution attainable in experiment is approximately $0.4 \times 10^{-6} \text{ eV} \approx 100 \text{ MHz}$,⁹ which exceeds by several orders of magnitude the values of the intramolecular and intermolecular nuclear magnetic interactions in molecular crystals. On this basis, we neglect the magnetic interactions in the Hamiltonian F and assume that

$$F = \sum_{n=1}^N [H_r(n) + V_{rp}(n)] + H_p, \quad (2.3)$$

where $H_r(n)$, H_p , and V_{rp} are the respective Hamiltonians of the n -th top, of the phonon reservoir, and of their interaction. The phonon reservoir accounts for all the nonrotational degrees of freedom of the crystal. The Hamiltonian $H_r(n)$ consists of a term corresponding to the rotational kinetic energy, and of a single-particle potential that depends only on the rotational coordinates of the n -th top. The explicit form of $H_r(n)$ is determined by the dimensionality of the problem and by the choice of the rotational coordinates (the Euler angles, the quaternions, etc.²⁰). The possibility of using the single-particle approximation for rotation in crystals [i.e., the absence of the terms $V(n_1, n_2)$ from F] was discussed by Hüller.¹⁴ One can expect this possibility to apply well to molecular crystals with tops if the potential for the rotation is produced mainly by nonrotating atoms and/or if the tops are sufficiently widely spaced. By virtue of the form of the Hamiltonian (2.3), the influence of the rotation of different tops on one another manifests itself in a higher order in V_{rp} than the interaction of the rotation with the phonons. Therefore in the case of a sufficiently weak interaction between the rotation and the phonons, which is the case considered below, we can assume the rotation of the different tops to be independent and we can put $N=1$ in (2.3). We shall accordingly omit the index n from now on.

To calculate the DDSS we construct the eigenfunctions of the Hamiltonian F under the foregoing assumptions. Let the system of identical nuclei contained in the top be invariant to the operation of the point group G . Inasmuch as the energies transferred in the SNS are small compared with the binding energies of the molecules and their fragments, it suffices to take into account only the "physically realizable" symmetry transformations, which do not call for surmounting impenetrable barriers (the Longuet-Higgins rule²¹). This requirement leads in fact to the need for taking into account only rotations of the tops as a whole. We note

that if the rotation of the top occurs in a crystal potential of sufficiently high symmetry, then the Hamiltonian H_r can have a symmetry group higher than G .

To construct the complete single-particle wave functions it is necessary to know the symmetrized spin and coordinate wave functions that transform in accordance with the irreducible representations $\mathcal{D}^\Gamma = \{\mathcal{D}_{ik}^\Gamma(g)\}$ of the group G ($i, k = 1, 2, \dots, s(\Gamma)$, Γ designates the irreducible representation). For this purpose it is necessary to choose the basis spin and coordinate wave functions and actually expand the irreducible representations of the group that they make up in terms of irreducible ones.^{22, 23} The operator of the total spin of the system of identical particles is a symmetrical function of the individual spins, therefore the symmetrized spin wave functions should be eigenfunctions of the square of the total angular momentum and of its projection on one of the coordinate axes, (the z axis), and is characterized by a total spin j and by its projection M on the z axis. Accordingly we designate the spine wave functions by $|\Gamma\alpha j M\rangle$, where the index $\alpha = 1, 2, \dots, \alpha(\Gamma)$ numbers the equivalent irreducible representations that enter in the expansion of the representation of the G group. The coordinate symmetrized wave function will be designated by $|\Gamma\delta i f\rangle$, where the index $\delta = 1, 2, \dots, \alpha(\Gamma)$ numbers the equivalent irreducible representations, and f is the set of additional quantum numbers that characterize the coordinate wave function. Reorientation of the top as a whole corresponds to even permutations of identical particles, which the complete wave functions by virtue of the identity principle, remain unchanged (i.e., they transform in accordance with A representation of the G group), and therefore take the form

$$|\Gamma\alpha\delta j M f\rangle = s(\Gamma)^{-1/2} \sum_{i=1}^{s(\Gamma)} |\Gamma\alpha i j M\rangle |\Gamma\delta i f\rangle, \quad (2.4)$$

where $\bar{\Gamma}$ is the representation adjoint to Γ , and coincides for unitary representation with the complex-conjugate representation²³:

$$\bar{\mathcal{D}}_{ik}^\Gamma(g) = \mathcal{D}_{ik}^{\bar{\Gamma}}(g) = \mathcal{D}_{ik}^{\bar{\Gamma}}(g^{-1}), \quad |\Gamma\alpha i j M\rangle = |\bar{\Gamma}\alpha i j M\rangle^*.$$

To calculate the matrix elements we write down the sums over the numbers of the nuclei, which enter in (2.1), in symmetrized form:

$$\sum_a I_{1\mu}(a) \exp(i\mathbf{Q}\mathbf{R}_a) = \sum_{\Gamma\alpha} \sum_{i=1}^{s(\Gamma)} T_\mu(\bar{\Gamma}\alpha i) B(\Gamma\alpha i), \quad (2.5)$$

where the operators $B(\Gamma\alpha i)$ are linear combinations of the coordinate functions $\exp(i\mathbf{Q} \cdot \mathbf{R}_a)$, which transform like the basis functions of the irreducible representation, while the operators $T_\mu(\bar{\Gamma}\alpha i)$ are linear combinations of the spin operators $I_{1\mu}(a)$, which transform like the basis functions of the adjoint representation $\bar{\Gamma}$. The operators $B(\Gamma\alpha i)$ and $T(\bar{\Gamma}\alpha i)$ can be obtained with aid of the projection operators (N_g is the number of elements in the group G)

$$P_r(\bar{\Gamma}i) = \frac{s(\Gamma)}{N_g} \sum_{g \in G} \mathcal{D}_{i,r}^{\bar{\Gamma}}(g) g, \quad P_s(\Gamma i) = \frac{s(\Gamma)}{N_g} \sum_{g \in G} \mathcal{D}_{i,r}^{\Gamma}(g) g, \quad (2.6)$$

whose properties are^{22, 23}

$$P_r(\Gamma i) T_\mu(\bar{\Gamma}\alpha i') = \delta_{rr'} \delta_{ii'} T_\mu(\Gamma\alpha i), \\ P_s(\Gamma i) B(\bar{\Gamma}\alpha i') = \delta_{rr'} \delta_{ii'} B(\Gamma\alpha i). \quad (2.7)$$

Substitution of (2.4) and (2.5) in (2.1) leads to an expression for the DDSS per top

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{B^2}{I(I+1)} \frac{k'}{2\pi k} \sum_s \frac{1}{s(\Gamma_1)s(\Gamma_2)} \langle \tilde{\Gamma}_{1\alpha_1 i_1 j_1} \| T(\tilde{\Gamma}\alpha i) \| \tilde{\Gamma}_{2\alpha_2 i_2 j_2} \rangle \quad (2.8)$$

$$\times \langle \tilde{\Gamma}_{2\alpha_2 i_2 j_2} \| T^+(\tilde{\Gamma}'\alpha' i') \| \tilde{\Gamma}_{1\alpha_1 i_1 j_1} \rangle \langle B(\Gamma\alpha i; 0) B^+(\Gamma'\alpha' i'; t) \rangle_{i_1 i_2 i_3 i_4}^{\Gamma_1 \delta_1 \Gamma_2 \delta_2}$$

where the summation is over $\Gamma_1, \delta_1, \Gamma_2, \delta_2, \alpha_1, \alpha_2, \Gamma, \alpha, i, \Gamma', \alpha', i', j_1, j_2, i_1, i_2, i_3$, and i_4 . In the derivation of (2.8), we have used the Wigner-Eckart theorem

$$\langle \tilde{\Gamma}_{1\alpha_1 i_1 j_1 M_1} | T_\nu(\tilde{\Gamma}\alpha i) | \tilde{\Gamma}_{2\alpha_2 i_2 j_2 M_2} \rangle = (-1)^{j_1 - M_1} \begin{pmatrix} j_1 & 1 & j_2 \\ -M_1 & \mu & M_2 \end{pmatrix} \langle \tilde{\Gamma}_{1\alpha_1 i_1 j_1} \| T(\tilde{\Gamma}\alpha i) \| \tilde{\Gamma}_{2\alpha_2 i_2 j_2} \rangle, \quad (2.9)$$

and the orthogonality property of the $3j$ symbols, and have introduced "irreducible" time correlation functions (ITCF), in which the summation is over states belonging to specified irreducible representations

$$\langle B(\Gamma\alpha i; 0) B^+(\Gamma'\alpha' i'; t) \rangle_{i_1 i_2 i_3 i_4}^{\Gamma_1 \delta_1 \Gamma_2 \delta_2} = \sum_{j_1 j_2} \rho_{\alpha\alpha'}(\Gamma, \delta, j_1) \langle \Gamma, \delta, i, j_1 | B(\Gamma\alpha i) | \Gamma_2 \delta_2 i_2 j_2 \rangle \quad (2.10)$$

$$\times \langle \Gamma_2 \delta_2 i_2 j_2 | e^{iHt} B^+(\Gamma'\alpha' i'; t) e^{-iHt} | \Gamma_1 \delta_1 i_1 j_1 \rangle,$$

where

$$\rho_{\alpha\alpha'}(\Gamma\delta) = \exp(-\beta E_{\Gamma\delta}) / \sum_{\Gamma\delta} \eta(\Gamma) \exp(-\beta E_{\Gamma\delta}), \quad (2.11)$$

$$\beta = 1/k_B T,$$

is the matrix element of the statistical operator of a scatterer in thermodynamic equilibrium, and $\eta(\Gamma)$ is the nuclear statistical weight of the level $E_{\Gamma\delta}$ (Ref. 23).

In the case of cyclic groups the representations are one-dimensional, there are no indices i in the relations written out above, and the ITCF (2.10) differ from that introduced in Ref. 15 only in the designation of the representation and by a factor that depends on the index of the representation Γ_1 . We note that the cross section for coherent scattering by symmetrical systems of identical nuclei is also expressed in terms of the ITCF (2.10). From the definition (2.10) we get the following properties of the ITCF:

$$\langle B(\Gamma\alpha i; 0) B^+(\Gamma'\alpha' i'; t) \rangle_{i_1 i_2 i_3 i_4}^{\Gamma_1 \delta_1 \Gamma_2 \delta_2} = \langle B(\Gamma\alpha i; -t) B^+(\Gamma'\alpha' i'; 0) \rangle_{i_1 i_2 i_3 i_4}^{\Gamma_1 \delta_1 \Gamma_2 \delta_2}, \quad (2.12)$$

$$\langle B(\Gamma\alpha i; 0) B^+(\Gamma'\alpha' i'; t) \rangle_{i_1 i_2 i_3 i_4}^{\Gamma_1 \delta_1 \Gamma_2 \delta_2} = \langle B^+(\Gamma'\alpha' i'; 0) B(\Gamma\alpha i; -t + i\hbar\beta) \rangle_{i_1 i_2 i_3 i_4}^{\Gamma_1 \delta_1 \Gamma_2 \delta_2}, \quad (2.13)$$

$$\langle B(\Gamma\alpha i; 0) B^+(\Gamma'\alpha' i'; t) \rangle_{i_1 i_2 i_3 i_4}^{\Gamma_1 \delta_1 \Gamma_2 \delta_2} = \langle B(\Gamma'\alpha' i'; 0) B^+(\Gamma\alpha i; -t) \rangle_{i_1 i_2 i_3 i_4}^{\Gamma_1 \delta_1 \Gamma_2 \delta_2}. \quad (2.14)$$

From (2.10) and (2.11) follow also the equalities

$$\langle B(\Gamma\alpha i; 0) B^+(\Gamma'\alpha' i'; t) \rangle_{i_1 i_2 i_3 i_4}^{\Gamma_1 \delta_1 \Gamma_2 \delta_2} = \langle \rho(t) B^+(\Gamma'\alpha' i') \rangle_{i_1 i_2 i_3 i_4}^{\Gamma_1 \delta_1 \Gamma_2 \delta_2} = \sum_{j_1 j_2} \langle \Gamma_1 \delta_1 i_1 j_1 | e^{-iFt} \rho_{\alpha\alpha'} B(\Gamma\alpha i) e^{iFt} | \Gamma_2 \delta_2 i_2 j_2 \rangle \langle \Gamma_2 \delta_2 i_2 j_2 | B^+(\Gamma'\alpha' i') | \Gamma_1 \delta_1 i_1 j_1 \rangle, \quad (2.15)$$

which show that the ITCF can be expressed in terms of solutions of the equation for the scatterer density matrix with initial condition

$$\rho(0) = \rho_{\alpha\alpha'} B(\Gamma\alpha i). \quad (2.16)$$

The use of relations (2.15) and (2.16) is convenient in the case considered by us of a weak interaction of the rotation with the phonons, for in this case there exists a kinetic equation for the rotational density ma-

trix, which is obtained from the density matrix of the scatterer by averaging over the phonon variables.

3. NONCOHERENT SCATTERING BY SYMMETRICAL THREE-SPIN SYSTEMS

We now make the results of the preceding section more concrete for the case of a solid containing tops consisting of three nuclei with spin $1/2$, placed at the vertices of an equilateral triangle. The tops rotate about threefold symmetry axes. The index Γ takes on values 0 (A representation) and ± 1 (E representations). The values of the total spin j of the top are connected with Γ by the relation

$$j = 3/2 - |\Gamma|. \quad (3.1)$$

Taking into account the connection $\tilde{\Gamma} = -\Gamma$, we can write

$$T_\nu(\Gamma) = T_\nu(-\Gamma) = I_{\nu\nu}(1) + \varepsilon^{-\Gamma} I_{\nu\nu}(2) + \varepsilon^\Gamma I_{\nu\nu}(3), \quad (3.2)$$

and

$$B(\Gamma) = 1/2 (e^{iQR} + \varepsilon^{-\Gamma} e^{iQR} + \varepsilon^\Gamma e^{iQR}), \quad (3.3)$$

where $\varepsilon = \exp(2\pi i/3)$. Expression (2.8) reduces to the form

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{8B^2 k'}{3 \cdot 2\pi k} \times \sum_{\Gamma_1 \Gamma_2 \Gamma} | \langle -\Gamma_1; 3/2 - |\Gamma_1| \| T(-\Gamma) \| -\Gamma_2; 3/2 - |\Gamma_2| \rangle |^2 \text{Re } K_{\Gamma_1 \Gamma_2 \Gamma}^{\Gamma_1 \Gamma_2}(\omega), \quad (3.4)$$

and

$$K_{\Gamma_1 \Gamma_2 \Gamma}^{\Gamma_1 \Gamma_2}(\omega) = \int_0^\infty dt e^{i\omega t} \langle B(\Gamma; 0) B^+(\Gamma; t) \rangle_{\Gamma_1 \Gamma_2}. \quad (3.5)$$

To calculate the reduced matrix elements we used the spin functions $|\Gamma j M\rangle \equiv |\Gamma M\rangle$, which take the form²⁴

$$|0^3/2\rangle = |+++\rangle, \quad |1^3/2\rangle = 3^{-1/2} (|-++\rangle + \varepsilon^{-\Gamma} |+-+\rangle + \varepsilon^\Gamma |+-\rangle), \quad (3.6)$$

where, for example, in the state $|++-\rangle$ the spins of nuclei 1 and 2 have projections $+\frac{1}{2}$, while the spin of the nucleus 3 has a projection $-\frac{1}{2}$ on the z axis. States with $M = -3/2$ and $-1/2$ are obtained by replacing $+$ by $-$ in (3.6) from the rules for multiplication of irreducible representations of the C_3 group it follows that the matrix elements in (3.4) differ from 0 only if the following relation is satisfied

$$\Gamma_1 - \Gamma - \Gamma_2 = 0, \pm 3. \quad (3.7)$$

The nonzero reduced matrix elements

$$\langle \Gamma_1; 3/2 - |\Gamma_1| \| T(\Gamma) \| \Gamma_2; 3/2 - |\Gamma_2| \rangle = \langle \Gamma_1 \| T(\Gamma) \| \Gamma_2 \rangle$$

have the values

$$\begin{aligned} \langle 0 \| T(0) \| 0 \rangle &= \sqrt{15}, \quad \langle 1 \| T(0) \| 1 \rangle = \langle -1 \| T(0) \| -1 \rangle = \sqrt{3/5}, \\ \langle 0 \| T(1) \| -1 \rangle &= -\langle 1 \| T(1) \| 0 \rangle = -\langle -1 \| T(1) \| 1 \rangle \\ &= \langle -1 \| T(-1) \| 0 \rangle = -\langle 0 \| T(-1) \| 1 \rangle = -\langle 1 \| T(-1) \| -1 \rangle = \sqrt{6}. \end{aligned} \quad (3.8)$$

From the complex-conjugate relation between the E states $|\Gamma f\rangle^* = |-\Gamma f\rangle$ we get in addition to the symmetry properties (2.12)–(2.14) the relation

$$\langle B(\Gamma; 0) B^+(\Gamma; t) \rangle_{\Gamma_1 \Gamma_2} = \langle B^+(\Gamma; 0) B(\Gamma; t) \rangle_{-\Gamma_1, -\Gamma_2}, \quad (3.9)$$

which makes it possible, in conjunction with Eqs. (2.12)–(2.14), to decrease the number of calculated ITCF.

4. IRREDUCIBLE TIME CORRELATION FUNCTIONS OF ROTATIONAL IN A TRIPLE PERIODIC POTENTIAL

The calculation of the ITCF of a one-dimensional top with Hamiltonian

$$\hbar H_r = \frac{\hbar^2 p^2}{2I} + V(\varphi), \quad p = -i \frac{\partial}{\partial \varphi}, \quad V(\varphi) = V(\varphi + 2\pi/3), \quad (4.1)$$

where I is the moment of inertia, was considered in Ref. 15, where the Hamiltonian of the interaction of the rotation with the phonons V_{rp} was not specified; it was only assumed that the smallness of V_{rp} is sufficient for the existence of a kinetic equation for the rotational density matrix δ defined in the basis of the eigenfunctions $|\Gamma v\rangle$ of the Hamiltonian (4.1) ($\hbar H_r |\Gamma v\rangle = E_{\Gamma v} |\Gamma v\rangle$). The index $v=0, 1, 2, \dots$, number the torsional levels, while Γ numbers the sublevels of the tunnel multiplet. The Hamiltonian of the interaction of the rotation with the phonons will be obtained here in explicit form.

The Hamiltonian of a crystal with uniaxial rigid top can be represented in the general case in the form

$$\hbar F = \frac{\hbar^2 p^2}{2I_0} + U(\varphi - \bar{\varphi}(q); q) + \hbar H_p, \quad (4.2)$$

$$\hbar H_p = \sum_s \frac{P_s^2}{2M_s} + U_p(q), \quad P_s = -i\hbar \frac{\partial}{\partial q_s}, \quad (4.3)$$

where M_s is the mass of the s -th atom of the crystal ($s=1, 2, \dots$), M_0 is the mass of the top, q is the set of displacements q_s of the crystal atoms ($s=1, 2, \dots$) and of the center of gravity of the tops ($s=0$) from the equilibrium positions, I_0 is the moment of inertia of the free top about the rotation axis, and $\bar{\varphi}(q)$ is the position of one of the minima of the rotation potential. If we make the substitutions $\varphi' = \varphi - \bar{\varphi}(q)$ and $q' = q$, and leave out the primes of the variables in the final expression, then the Hamiltonian (4.2) can be represented in the form (2.3), where the Hamiltonian of the phonons is given by expression (4.3), and the Hamiltonian of the top by expression (4.1) in which

$$V(\varphi) = \langle U(\varphi; q) \rangle, \quad \frac{1}{I} = \frac{1}{I_0} + \sum_s \frac{1}{M_s} \left\langle \left[\frac{\partial \bar{\varphi}(q)}{\partial q_s} \right]^2 \right\rangle, \quad (4.4)$$

I is the effective moment of inertia of the top, and the Hamiltonian of the interaction of the rotation with the phonons takes the form

$$\hbar V_{rp} = \hbar V_k + \hbar V' = -\hbar p \sum_s \left[\frac{P_s}{2M_s}, \frac{\partial \bar{\varphi}(q)}{\partial q_s} \right] + \frac{1}{2} [U(\varphi; q) - \langle U(\varphi; q) \rangle], \quad (4.5)$$

where we have neglected terms of order $\bar{\varphi}^{(1)} \bar{\varphi}^{(2)}$ and terms of higher order, which result from the expansion $\bar{\varphi}(q) = \bar{\varphi}^{(0)} + \bar{\varphi}^{(1)} + \bar{\varphi}^{(2)} + \dots$ in powers of q . The angle brackets $\langle \dots \rangle$ denote averaging over the phonon variables, i.e., over the equilibrium states of the system with the Hamiltonian H_p .

In the classical limit we have

$$\sum_s \left[\frac{P_s}{2M_s}, \frac{\partial \bar{\varphi}(q)}{\partial q_s} \right] \rightarrow \sum_s q_s \frac{\partial \bar{\varphi}(q)}{\partial q_s} = \frac{d\bar{\varphi}(q)}{dt}.$$

Therefore in analogy with the vibrational-rotational

interaction in the molecules, the term $\hbar V_k$ and $\hbar V_{rp}$ will be called the Coriolis interaction. This interaction in solids was introduced in Ref. 25, where it was used to explain the temperature dependence of the spin-lattice relaxation due to the rotational tunneling of the methyl groups. The term $U[\varphi - \bar{\varphi}(q); q]$ in (4.2) can be represented in our case of triple symmetry in the form of an expansion in $\cos(3n[\varphi - \bar{\varphi}_n(q)])$, $n=1, 2, \dots$ if it is assumed that only the first term is significant in this expansion, then

$$\bar{\varphi}(q) = \varphi_1(q), \quad U(\varphi - \bar{\varphi}(q); q) = U_1(q) \{1 - \cos(3[\varphi - \bar{\varphi}(q)])\}.$$

Therefore

$$V(\varphi) = \frac{1}{2} \langle U_1(q) \rangle (1 - \cos 3\varphi) = \frac{1}{2} V_3 (1 - \cos 3\varphi), \quad (4.6)$$

$$\hbar V' = \frac{1}{2} [U_1(q) - \langle U_1(q) \rangle] (1 - \cos 3\varphi). \quad (4.7)$$

We assume that the conditions discussed in Ref. 15 for the applicability of the kinetic equation for a rotational density matrix are satisfied. An analysis of this equation has shown that in the infinite chain of equations for the matrix elements $\langle \Gamma_1 v | \rho(t) | \Gamma_2 v \rangle$, in terms of which the low-frequency Fourier components of the ITCF are expressed, under the condition

$$\exp(-\hbar\beta\Omega) = \exp[-\beta(E_{11} - E_{10})] \ll 1, \quad (4.8)$$

which is henceforth assumed satisfied, in the case $|\Gamma_1| = |\Gamma_2|$ it suffices to take into account the two levels $v=0$ and 1 , while at $|\Gamma_1| \neq |\Gamma_2|$ it may be necessary to take into account all the below-barrier levels with $E_{\Gamma v} < V_3$, for which the tunnel splitting $\hbar |\Delta_v| = |E_{1v} - E_{0v}|$ is less than or of the order of the relaxation coefficients $\gamma_{vv}^{\Gamma_1 \Gamma_2}$. The relaxation coefficients $\gamma_{v_1 v_2}^{\Gamma_1 \Gamma_2}$ are expressed in accordance with Ref. 15 in terms of the matrix elements

$$p_{v_1 v_2}^{\Gamma} = \langle \Gamma v_1 | p | \Gamma v_2 \rangle, \quad c_{v_1 v_2}^{\Gamma} = \langle \Gamma v_1 | 1 - \cos 3\varphi | \Gamma v_2 \rangle$$

and the spectral densities

$$j_p(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \langle A(t) A(0) \rangle, \quad A = \sum_s \left[\frac{P_s}{2M_s}, \frac{\partial \bar{\varphi}(q)}{\partial q_s} \right], \quad (4.9)$$

$$j_c(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \langle C(t) C(0) \rangle, \quad C = \frac{1}{2\hbar} [U_1(q) - \langle U_1(q) \rangle]. \quad (4.10)$$

At temperatures satisfying the condition (4.8), the tunnel structure of the torsion levels in the SNS spectrum can manifest itself only for not very high barriers, for which the splitting of the ground level Δ_0 greatly exceeds the attained resolution, which amounts to approximately 100 MHz. If we bear in mind the experiments on SNS from methyl groups,^{10, 13, 18} then we must consider barriers $V_3 \approx 400 \text{ cm}^{-1}$ for which calculation by the quasiclassical formula, obtained in Ref. 26 using the value $r=1.8 \text{ \AA}$ for the interproton distance in the methyl groups and $I \sim I_0$ leads to $\Delta_0 \approx 300 \text{ MHz}$. The value of $\hbar\Omega$ for the methyl group is $100\text{--}200 \text{ cm}^{-1}$. For the ammonia groups $r \approx 1.4 \text{ \AA}$ and $\Delta_0 \approx 300 \text{ MHz}$ at $V_3 \approx 700 \text{ cm}^{-1}$. For such barriers, the third torsion level lies either near the barrier or above the barrier²⁹ and one can assume the inequality $|\Delta_2| > |\gamma_{22}^{\Gamma_1 \Gamma_2}|$, $|\Gamma_1| \neq |\Gamma_2|$, to be satisfied, so that we can confine ourselves to a two-level approximation. The matrix elements p_{vv}^{Γ} for the below-barrier levels differ from zero only because of the finite penetrability of the barrier at $\Gamma=0$, and increase rapidly with increasing v .²⁷

$$\exp(-\hbar\beta\Omega), \Delta_0/|\Delta_1|, (p_{00}/p_{11})^2 \ll 1.$$

Neglecting terms of order $\hbar B\Delta_0 \ll 1$, we obtain for the DDSS

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{B^2 k'}{4} \left\{ (10|B_0^{00}|^2 + |B_0^{11}|^2 + |B_0^{-1-1}|^2) \delta(\omega) + \frac{4}{\pi} (|B_1^{-11}|^2 + |B_1^{-1-1}|^2) \frac{\tau_{EE}}{1 + \omega^2 \tau_{EE}^2} + \frac{4}{\pi} (|B_1^{01}|^2 + |B_1^{0-1}|^2) \right. \quad (4.11)$$

$$\left. \times \left[\frac{\tau_{AE}}{1 + (\omega - \Delta_1)^2 \tau_{AE}^2} + \frac{\tau_{AE}}{1 + (\omega + \Delta_1)^2 \tau_{AE}^2} \right] \right\}, \quad \tau_{EE}^{-1} = \gamma_1^{-1}, \quad \tau_{AE}^{-1} = \gamma_1^{10}, \quad \Delta_1 = \omega_1^{10}, \quad (4.12)$$

$$\gamma_1^{11} + i\omega_1^{11} = \gamma_{00}^{11} + i\omega_0^{11} - \frac{\gamma_{01}^{11} \gamma_{10}^{11}}{i\omega_1^{11} + \gamma_{11}^{11}}, \quad (4.13)$$

where

$$B_\Gamma^{r_1 r_2} = \langle \Gamma_1 0 | B(\Gamma) | \Gamma_2 0 \rangle, \quad \omega_\Gamma^{r_1 r_2} = (E_{r_1} - E_{r_2})/\hbar.$$

Let θ and ϕ be the spherical angles of a vector \mathbf{Q} in a coordinate system whose z axis coincides with the symmetry axis of the top, the zx plane passes through one of the minima of the potential $V(\varphi)$. Then

$$QR_\alpha = QR \sin \theta \cos \left[\varphi - \Phi + \frac{2\pi}{3}(a-1) \right] \quad Q = |\mathbf{Q}|, \quad R = |\mathbf{R}_\alpha|.$$

To calculate the matrix element $B_\rho^{\Gamma_1 \Gamma_2}$ we use the tunnel approximation^{27, 15} for the wave function and expand the exponentials in (3.3) in a Taylor series about the values $\varphi = 0$ and $\pm 2\pi/3$. With accuracy of order $(QR)^2 \hbar/4I\Omega$ we can confine ourselves to the first term of this expansion ($\hbar/4I\Omega \sim 1/40$ for the methyl groups). For the SNS in a polycrystal it is necessary to average the DDSS over the angles θ and ϕ . We denote the result of the averaging by a superior bar

$$\frac{\overline{d^2\sigma}}{d\Omega d\omega} = B^2 \frac{k'}{k} \left\{ \left(1 + 2 \frac{\sin Qr}{Qr} \right) \delta(\omega) + \frac{2}{3\pi} \left(1 - \frac{\sin Qr}{Qr} \right) \frac{\tau_{EE}}{1 + \omega^2 \tau_{EE}^2} + \frac{2}{3\pi} \left(1 - \frac{\sin Qr}{Qr} \right) \times \left[\frac{\tau_{AE}}{1 + (\omega - \Delta_1)^2 \tau_{AE}^2} + \frac{\tau_{AE}}{1 + (\omega + \Delta_1)^2 \tau_{AE}^2} \right] \right\}; \quad (4.14)$$

Here $r = R/\sqrt{3}$ is the distance between the nuclei in the top. Thus, the SNS spectrum in the region of small energy transfers ($\ll \hbar\Omega$) consists of the elastic-scattering line [the first term in the right-hand side of (4.14)] and a tunnel multiplet corresponding to inelastic scattering with change of the quantum number Γ .

If we put in (4.14) formally $\Delta_1 = 0$ and $\tau_{EE} = \tau_{AE} = \tau$, then this expression goes over into the results obtained on the basis of the classical model of reorientation jumps around the C_3 axis through angles $\pm 2\pi/3$.¹⁶⁻¹⁸

In the limit of a vanishing rotation interaction with the phonons $\tau_{EE}, \tau_{AE} \rightarrow \infty$ and we get from (4.14)

$$\frac{\overline{d^2\sigma}}{d\Omega d\omega} = B^2 \frac{k'}{k} \left\{ \left(\frac{5}{3} + \frac{4}{3} \frac{\sin Qr}{Qr} \right) \delta(\omega) + \frac{2}{3} \left(1 - \frac{\sin Qr}{Qr} \right) [\delta(\omega - \Delta_1) + \delta(\omega + \Delta_1)] \right\}, \quad V_{rr} \rightarrow 0. \quad (4.15)$$

The expression in the curly brackets (the scattering law) is given in Ref. 28.

An analysis of the explicit expressions for the ITCF correlation times τ_{EE} and τ_{AE} and for the effective tunneling frequency Δ_1 has shown that their temperature behavior depends on the relations between the quantities

$$a = (p_{01}^{11})^2 j_p(\Omega), \quad b_v(\omega) = 1/2 (p_{vv}^{11})^2 j_p(\omega), \quad c = 1/2 (c_{01}^{11})^2 j_c(\Omega), \\ c_v(\omega) = 1/2 (c_{vv}^{11})^2 j_c(\omega), \quad d = (p_{01}^{11} - p_{01}^{01})^2 j_p(\Omega), \quad |\Delta_0|.$$

The parameters a and c determine the rates of vibrational relaxation (the lifetime and the torsion states relative to transitions with change of the quantum numbers v), while the quantities $b_v(\omega)$ and $c_v(\omega)$ at $\omega = 0$, and $|\Delta_0|$ determine the rates of the phase relaxation in the system of tunnel sublevels of the v -th torsional multiplet. The parameters $b_v(\omega)$, c , $c_v(\omega)$ and d for the below-barrier levels are different from 0 only because of the finite penetrability of the barrier. In the present paper the calculation of the spectral densities $j_p(\omega)$ and $j_c(\omega)$ for the concrete models of the phonon spectrum will not be considered. We discuss briefly two principal limits.

1) Rapid phase relaxation:

$$4b_1(0) \gg a + 2c, \quad b_1(|\Delta_1|) + c_1(|\Delta_1|) \gg a + c.$$

In this case

$$\tau_{EE}^{-1} = 4b_1(0) + (a + 2c) e^{-\hbar\beta a}, \quad (4.16)$$

$$\tau_{AE}^{-1} = b_1(\Delta_0) + c_1(\Delta_0) + (a + c) e^{-\hbar\beta a}, \quad (4.17)$$

$$\Delta_1 = \bar{\Delta}_0 + \bar{\Delta}_1 \frac{a^2 e^{-\hbar\beta a}}{\bar{\Delta}_1^2 + [b_1(|\Delta_1|) + c_1(|\Delta_1|)]^2}, \quad (4.18)$$

where $\bar{\Delta}_v = \Delta_v + \text{Im} \gamma_{vv}$.¹⁰ If the dynamic shifts¹⁰ $\text{Im} \gamma_{vv}$ are small compared with Δ_0 , then the temperature dependence of Δ_1 is determined by the degree of satisfaction of the inequality $b_1(|\Delta_1|) + c_1(|\Delta_1|) \gg a + c$. The better this inequality is satisfied, the smaller the second term in the right-hand of (4.18) and the weaker the temperature dependence of Δ_1 .

2) Fast vibrational relaxation:

$$a + 2c \gg 4b_1(0), \quad a + c \gg b_1(|\Delta_1|) + c_1(|\Delta_1|).$$

In this limit

$$\tau_{EE}^{-1} = 4b_1(0) + 4 \left[b_1(0) + \frac{2ac}{a+2c} \right] e^{-\hbar\beta a}, \quad (4.19)$$

$$\tau_{AE}^{-1} = b_1(\Delta_0) + c_1(\Delta_0) + \left[b_1(|\Delta_1|) + c_1(|\Delta_1|) + (a+c) \frac{\bar{\Delta}_1^2 + 2ac + ad + c^2}{\bar{\Delta}_1^2 + (a+c)^2} \right] e^{-\hbar\beta a}, \quad (4.20)$$

$$\Delta_1 = \bar{\Delta}_0 + \bar{\Delta}_1 \frac{a^2}{\bar{\Delta}_1^2 + (a+c)^2} e^{-\hbar\beta a}. \quad (4.21)$$

It follows from the last expression that if the quantum rate of the transitions between the states with $v = 0$ and $v = 1$ is comparable with the tunnel splitting of the excited level, then at $a \approx c$ the effective tunneling frequency Δ_1 depends strongly on the temperature, and with increasing temperature a collapse of the tunnel multiplet takes place ($\Delta_1 \rightarrow 0$). This effect is observed in SNS from ammonia¹² and methyl¹³ groups. Under the conditions $a^2 \gg \Delta_1^2$ and $a \gg c$, neglecting the dynamic shifts, we obtain from (4.21) the relation $\Delta_1 = \Delta_0 - |\Delta_1| \exp(-i\hbar\beta)$, which was introduced earlier²⁹ from qualitative considerations to explain the experiments on magnetic resonance and which was used in analogy

in Refs. 12 and 13 to describe the temperature dependence of the tunnel frequencies in the SNS spectrum.

We call attention in conclusion to the fact that the ITCF correlation times are not expressed in terms of the lifetimes on the levels. At low temperatures, the main contribution to τ_{EE}^{-1} and τ_{AE}^{-1} is due to phase relaxation.

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Influence of radiation on the motion of channeled particles

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A detailed investigation is made of the influence of radiation on the motion of channeled particles in a crystal. It is shown, in particular, that radiation can lead to an increase in the angular divergence of a particle beam in a channel. The analysis of the relaxation of the transverse energy via radiation takes into account the multiple scattering of the channeled particle in the crystal.

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INTRODUCTION

It is known (see Refs. 1-4 and the bibliography therein) that spontaneous radiative transitions between transverse-energy levels of a channeled particle lead to intense x and gamma radiation, accompanied by relaxation of the transverse energy. For light channeled particles, as noted by us earlier,² the process of radiative damping of the transverse energy can play a noticeable role alongside the nonradiative de-

channeling processes that lead to an increase of the transverse energy. In this case, generally speaking, self-focusing of a beam of channeled particles is possible.

The effect of radiative focusing of a beam of channeled particles was considered also by others.⁵⁻⁷ However, the results of Baryshevskii and Dubovskaya,⁵ as shown by us earlier,⁸ turned out to be completely in error. Wedell's paper⁷ also contains conclusions that