

# THEORY OF ZEEMAN LEVEL WIDTHS OF NUCLEAR SPINS IN SOLID ORTHOHYDROGEN

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The Zeeman level widths of nuclear spins in solid orthohydrogen are calculated. The treatment is based on an equation for the nonequilibrium mean values of the operators characterizing the nonequilibrium state of the spin system, which interacts with the libron system (treated as a thermal bath). This equation is derived by the method of Zubarev's nonequilibrium statistical operator. The correlation functions for the libron subsystem that occur in the expression for the widths are calculated in the Bose approximation.

## 1. Introduction

An isolated system of noninteracting nuclear spins of a molecular orthohydrogen crystal in a constant external magnetic field has a discrete Zeeman energy spectrum. As a result of the interaction with the other degrees of freedom of the crystal the Zeeman levels become quasidiscrete, acquiring a finite width  $\Gamma$ , which is related to the lifetime by the equation  $\Gamma \sim 1/\tau$ . We should mention that the notion of quasistationary states is justified when the widths of the quasidiscrete levels are small compared with the distance between the levels. A broadening of the Zeeman levels in solid orthohydrogen at low temperatures may be due to several physical factors (interactions with lattice vibrations, with the libron subsystem, etc.). Estimates show that at low temperatures the interaction with the lattice vibrations can be neglected and one need only allow for the interaction with the libron system.

At low temperatures the system of orthohydrogen molecules, which form a rigid face-centered cubic lattice and are bound only by the electric quadrupole-quadrupole interaction, is described by the Hamiltonian [1]

$$\mathcal{H}_L = \sum_{ij} \sum_{m,n} \gamma_{ij}^{mn} O_i^m O_j^n. \quad (1)$$

Here, the first sum is over all the sites of the face-centered cubic lattice and  $\gamma_{ij}^{mn}$ , the coupling constants of the quadrupole-quadrupole interaction, are determined in [1]. The operators  $O_i^m$  are related to the operators of the quadrupole moment components of the molecule and are equal to

$$\begin{aligned} O_i^0 &= 3(J_i^z)^2 - 2, \\ O_i^{\pm 1} &= J_i^z J_i^{\pm} + J_i^{\pm} J_i^z, \\ O_i^{\pm 2} &= (J_i^{\pm})^2, \\ J_i^{\pm} &= J_i^x \pm iJ_i^y, \end{aligned} \quad (2)$$

where  $J_i$  is the operator of the angular momentum of the  $i$ -th molecule ( $J = 1$ ), the quantization axis  $z_i$  being taken along the symmetry axis of the molecule  $i$ . In the ground state the molecular lattice of solid orthohydrogen consists of four simple cubic sublattices in each of which the molecules are translationally and orientationally equivalent. The symmetry axes of the molecules are directed along the four different threefold symmetry axes of the crystal, which coincide with the principal diagonals of the cube of the face-centered cubic lattice. In the molecular field approximation it has been found [1, 2] that the state with  $J_i^z = 0$  lies below the states with  $J_i^z = \pm 1$  and is the ground state. Calculations show [1, 3-5] that near the ground state, i.e., in the orientationally ordered phase, there exist collective excitations, known as librions, which are associated with the transitions  $(J_i^z = 0) \rightarrow (J_i^z = \pm 1)$ . Because of the anisotropic nature of the electric quadrupole-quadrupole interaction the gap in the libron energy spectrum is approximately equal

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Joint Institute of Nuclear Research. Translated from *Teoreticheskaya i Matematicheskaya Fizika*, Vol. 6, No. 1, pp. 109-122, January, 1971. Original article submitted June 1, 1970.

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to 10°K. The width of the libron band is about 5°K [note that the energy of the transition ( $J_i = 1$ )  $\rightarrow$  ( $J_i = 3$ ) is 860°K].

In the present paper we shall calculate the Zeeman level widths of the nuclear spins arising from the interaction with the libron system, treated as a thermal bath. As in the study of the longitudinal nuclear spin-libron relaxation [5-7] in solid orthohydrogen, we shall assume that the interaction between the spin and libron subsystems is made up of the intramolecular dipole-dipole interaction between the nuclear spins of the protons and the  $I_i \cdot J_i$  interaction, both interactions being modulated by the quadrupole interaction. The intermolecular dipole-dipole interaction will be neglected (see also [7]).

Now it is clear that the problem can be solved on the basis of perturbation theory for the Green's functions [8]. However, we shall calculate the Zeeman nuclear level widths by means of equations for the nonequilibrium mean values since it is well known that the equations for the nonequilibrium mean values are equivalent to the equations for the corresponding equilibrium Green's functions. This is an expression of Onsager's principle, which is widely employed in the theory of nonequilibrium processes. In the investigation [9], Zubarev and the present authors used the nonequilibrium statistical operator method [10-12] to obtain the corresponding equation for the nonequilibrium mean values of the operators characterizing the nonequilibrium state of a subsystem interacting with a thermal bath. Using this equation, one can very easily calculate the energy shift and the damping; this will become apparent in the present investigation.

## 2. Hamiltonian of the System

We shall consider a nuclear spin system in an external constant magnetic field interacting with a libron system. We write the Hamiltonian of the complete system in the form [6]

$$\mathcal{H} = \mathcal{H}_s + \mathcal{H}_L + V. \quad (3)$$

Here,  $\mathcal{H}_s = -a \sum_i I_i^z$  is the Zeeman Hamiltonian of the nuclear spins;  $I_i^z$  is the operator of the z component of the total nuclear spin of the molecule ( $I = 1$ ) at the site  $i$ ; the quantization axis is along the external magnetic field  $H_0$ ;  $a = \gamma H_0$ , where  $\gamma$  is the gyromagnetic ratio;  $\mathcal{H}_L$  is the Hamiltonian of the libron system (1). The interaction operator has the form

$$V = V_{dd} + V_{I,J}, \quad (4)$$

where

$$V_{dd} = \sum_i \left\{ -d \cdot I_i^3 \cdot J_i^3 + \frac{3}{2} d (I_i \cdot J_i) + 3d (I_i \cdot J_i)^2 \right\} \quad (5)$$

is the operator of the intramolecular interaction of the protons of the molecule. Note that in contrast to (1) the quantization axis for  $J_i$  in the expression (5) is the external magnetic field  $H_0$ ;

$$V_{I,J} = -c \sum_i I_i \cdot J_i, \quad (6)$$

is the operator of the I-J interaction,  $J_i$  in (6) also being quantized along  $H_0$ . We shall use the notation

$$c = 2\mu_i H'; \quad d = \frac{1}{2} \mu_i^2 \langle r^{-3} \rangle,$$

the definition and the numerical value of these quantities can be found in [6]. The interaction operator  $V$  in (4) can be represented in the form

$$V = \frac{3}{4} d \sum_i \sum_{m,n=-2}^2 q_i^m O_i^{-m} - c \sum_i I_i \cdot J_i, \quad (7)$$

where by analogy with (2), we have introduced the operators  $q_i^m$ :

$$\begin{aligned} q_{i0} &= 2[(I_i^z)^2 - 2/3], \\ q_i^{\pm 1} &= I_i^{\pm} I_i^z + I_i^z I_i^{\pm}, \\ q_i^{\pm 2} &= (I_i^{\pm})^2, \\ I_i^{\pm} &= I_i^x \pm i I_i^y, \end{aligned} \quad (8)$$

We must emphasize once more that the operators  $O_i^m$  in (7) have the form (2) but the quantization axis for  $J_i$  is along the external magnetic field  $H_0$ . In calculating the Zeeman level widths in the present paper we shall restrict ourselves in (7) to terms that do not lead to a mixing [13] of states with different  $I_i^z = \lambda$ , where  $1 \leq \lambda \leq 1$ .

$$V = \frac{3}{4} d \sum_i q_i^0 O_i^0 - c \sum_i I_i^z \cdot J_i^z. \quad (9)$$

Since  $[I_i^z, V] = 0$ , the interaction in the form (9) does not lead to longitudinal nuclear spin-libron relaxation [5]. This corresponds to a restriction to elastic scattering of librons by the nuclear spins and is physically justified since the gap in the libron spectrum is much greater than the distance between the Zeeman nuclear levels. Using the equation

$$I_i^z = \sum_{\lambda} \lambda a_{i\lambda}^{\dagger} a_{i\lambda} = \sum_{\lambda} \lambda n_{i\lambda}, \quad (10)$$

it is convenient to introduce  $a_{i\lambda}^{\dagger}$  and  $a_{i\lambda}$ , the operators of creation and annihilation of a spin at site  $i$  with  $z$  component equal to  $\lambda$ . One can show that these operators satisfy the commutation relations

$$\begin{aligned} [a_{i\lambda}, a_{j\sigma}] &= [a_{i\lambda}^{\dagger}, a_{j\sigma}^{\dagger}] = 0, \\ [a_{i\lambda}, a_{j\sigma}] &= \delta_{ij} \delta_{\lambda\sigma} \left( 1 - \sum_{\nu} a_{i\nu}^{\dagger} a_{i\nu} \right) - \delta_{ij} a_{i\sigma}^{\dagger} a_{i\lambda}, \end{aligned} \quad (11)$$

where  $a_{i\lambda} \cdot a_{i\sigma}^{\dagger} = 0$  for  $\lambda \neq \sigma$ ;  $a_{i\lambda}^{\dagger} \cdot a_{i\sigma}^{\dagger} = a_{i\lambda} \cdot a_{i\sigma} = 0$ . Using (10) we rewrite  $\mathcal{H}$ , and  $V$  in the form (see [9])

$$\mathcal{H}_s = \sum_{i\lambda} E_{\lambda}(i) a_{i\lambda}^{\dagger} a_{i\lambda}; \quad E_{\lambda}(i) = -a\lambda, \quad (12)$$

$$V = \sum_{i\lambda} \varphi_{\lambda}(i) a_{i\lambda}^{\dagger} a_{i\lambda}, \quad (13)$$

where

$$\varphi_{\pm 1} = \frac{1}{2} d \cdot O_i^0 \mp c J_i^z; \quad \varphi_0(i) = -d \cdot O_i^0. \quad (13a)$$

### 3. Equation for the Nonequilibrium Mean Values

In order to obtain an equation for the nonequilibrium mean values [9], we shall use Zubarev's nonequilibrium statistical operator method [10-12]. Our starting point will be the Hamiltonian (3), in which  $\mathcal{H}_s$ ,  $\mathcal{H}_L$ , and  $V$  are taken, respectively, in the form (12), (1), and (13). We shall take the mean values of the operators  $a_{i\lambda}$ ,  $a_{i\lambda}^{\dagger}$ , and  $n_{i\lambda}$  [9] as the set of variables describing the nonequilibrium state of the spin system with the Hamiltonian  $\mathcal{H}_s$ . We shall regard the libron system, which is in equilibrium at the temperature  $\theta = 1/\beta$ , as a thermal bath and describe it by means of the Hamiltonian  $\mathcal{H}_L$  (1).

We introduce the quasiequilibrium distribution [10-12]

$$\rho_q(t, 0) = e^{-S(t, 0)}, \quad (14)$$

where

$$S(t, 0) = \Phi(t) + \sum_{i\lambda} \{ f_{i\lambda}(t) a_{i\lambda} + f_{i\lambda}^*(t) a_{i\lambda}^{\dagger} + F_{i\lambda}(t) n_{i\lambda} \} + \beta \mathcal{H}_L, \quad (15)$$

$$\Phi = \ln \text{Sp} \exp \left\{ - \sum_{i\lambda} (f_{i\lambda}(t) a_{i\lambda} + f_{i\lambda}^*(t) a_{i\lambda}^{\dagger} + F_{i\lambda}(t) n_{i\lambda}) - \beta \mathcal{H}_L \right\}. \quad (15a)$$

The first argument of  $\rho_q(t, 0)$  indicates the implicit dependence of this quantity on the time and the second its dependence through the Heisenberg representation;  $f_{i\lambda}(t)$ ,  $f_{i\lambda}^*(t)$ , and  $F_{i\lambda}(t)$  are the parameters conjugate to  $\langle a_{i\lambda}^{\dagger} \rangle_q$ ,  $\langle a_{i\lambda} \rangle_q$ , and  $\langle n_{i\lambda} \rangle_q$  in the sense of nonequilibrium thermodynamics [10-12];  $\langle \dots \rangle_q = \text{Sp}(\rho_q \dots)$ . We may call  $S(t, 0)$  the entropy operator, since  $\langle S(t, 0) \rangle_q$  is the entropy.

We construct the nonequilibrium statistical operator  $\rho(t, 0)$  as follows:

$$\rho(t, 0) = \exp \{ - \widetilde{S(t, 0)} \}, \quad (16)$$

where

$$\widetilde{S(t, 0)} = \varepsilon \int_{-\infty}^0 dt_1 e^{\varepsilon t_1} U_{t_1} S(t + t_1, 0) U_{t_1}^{\dagger} = -\varepsilon \int_{-\infty}^0 dt_1 e^{\varepsilon t_1} U_{t_1} \ln \rho_q(t + t_1, 0) U_{t_1}^{\dagger} \quad (16a)$$

is the quasi-invariant part of the entropy operator;  $\varepsilon \rightarrow +0$  after the thermodynamic passage to the limit in the calculation of the mean values; and  $U_t = \exp(-t \mathcal{H}_s / \hbar)$  is the evolution operator. The parameters  $f_{i\lambda}(t)$ ,  $f_{i\lambda}^*(t)$ , and  $F_{i\lambda}(t)$  can be found from the conditions

$$\langle a_{i\lambda} \rangle = \langle a_{i\lambda} \rangle_{\mathbf{q}}; \langle a_{i\lambda}^+ \rangle = \langle a_{i\lambda}^+ \rangle_{\mathbf{q}}; \langle n_{i\lambda} \rangle = \langle n_{i\lambda} \rangle_{\mathbf{q}}, \quad (17)$$

where  $\langle \dots \rangle = \text{Sp}(\rho(t, 0) \dots)$  denotes averaging with the nonequilibrium statistical operator (16). Fulfillment of the conditions (17) ensures conservation of the normalization after the quasi-invariant part has been taken. The entropy of the system is

$$S(t) = \langle S(t, 0) \rangle = \Phi(t) + \sum_{i\lambda} \{ \langle a_{i\lambda} \rangle f_{i\lambda}(t) + \langle a_{i\lambda}^+ \rangle f_{i\lambda}^*(t) + \langle n_{i\lambda} \rangle F_{i\lambda}(t) \} + \langle \mathcal{H}_L \rangle \beta. \quad (15b)$$

One can therefore show that the mean values in (15b) can be interpreted as generalized thermodynamic coordinates and  $f_{i\lambda}$ ,  $f_{i\lambda}^*$ , and  $F_{i\lambda}$  as thermodynamic forces.

Averaging the equation of motion for  $a_{i\lambda}$  with the nonequilibrium statistical operator (16):

$$i \frac{da_{i\lambda}}{dt} = [a_{i\lambda}, \mathcal{H}] = E_{i\lambda}(t) a_{i\lambda} + \frac{1}{i} [a_{i\lambda}, V]$$

and restricting ourselves to the second order in the weak interaction, we obtain, as in [9, 14],

$$i \frac{d\langle a_{i\lambda} \rangle}{dt} = E_{i\lambda}(t) \langle a_{i\lambda} \rangle + \frac{1}{i} \int_{-\infty}^0 \langle [[a_{i\lambda}, V] V(t_1)] \rangle_{\mathbf{q}} e^{et_1} dt_1. \quad (18)$$

Here,  $V(t_1)$  denotes a representation of the interaction operator  $V$ . Note that in deriving Eq. (18) we used the condition  $\langle \varphi_{\lambda}(i) \rangle_{\mathbf{q}} = 0$ . This can always be achieved as follows. To the Hamiltonian  $\mathcal{H} = \mathcal{H}_s + \mathcal{H}_L + V$  add and subtract  $\langle \varphi_{\lambda}(i) \rangle_{\mathbf{q}}$ . In addition, introduce the quantities  $O_i^{01} = O_i^0 - \langle O_i^0 \rangle_{\mathbf{q}}$  and  $J_i^{Z1} = J_i^{Z0} - \langle J_i^{Z0} \rangle_{\mathbf{q}}$ . Equations (12) and (13) are then replaced by

$$\begin{aligned} \mathcal{H}_s &= \sum_{i\lambda} E_{\lambda}'(i) a_{i\lambda}^+ a_{i\lambda}; & E_{\lambda}'(i) &= -a \cdot \lambda + \langle \varphi_{\lambda}(i) \rangle_{\mathbf{q}}', \\ V &= \sum_{i\lambda} \varphi_{\lambda}'(i) a_{i\lambda}^+ a_{i\lambda}; & \varphi_{\lambda}'(i) &= \varphi_{\lambda}(i) - \langle \varphi_{\lambda}(i) \rangle_{\mathbf{q}}, \end{aligned} \quad (19)$$

and hence,  $\langle \varphi_{\lambda}'(i) \rangle_{\mathbf{q}} = 0$ . Therefore, we can assume that we are dealing with renormalized quantities, whose primes we shall omit to shorten the notation. In addition, the renormalization (19) enables us to find the distance between the peaks of the NMR resonance line in solid orthohydrogen. It is readily verified that the distance  $\delta\nu$  between the peaks is

$$\delta\nu = \left| \frac{3}{2} d (3 \cos^2 \theta - 1) \langle O^0 \rangle \right|,$$

where  $O^0 = 3(JZ)^2 - 2$ , and  $\theta$  is the angle between the quantization axis for  $J_1^Z$  and the direction of the external magnetic field. Averaging  $\delta\nu$  over all directions, we obtain

$$\overline{\delta\nu} = \frac{3}{2} d \langle 2 - (J^Z)^2 \rangle,$$

which agrees with the value of  $\overline{\delta\nu}$  given in [15] (see also [17]).

#### 4. Zeeman Level Widths

We shall follow [9]. We expand the double commutator on the right side of Eq. (18):

$$\begin{aligned} i \frac{d\langle a_{i\lambda} \rangle}{dt} &= E_{i\lambda}(t) \langle a_{i\lambda} \rangle + \frac{1}{i} \int_{-\infty}^0 dt_1 e^{et_1} \langle \varphi_{\lambda}(i), \varphi_{\lambda}(i, t_1) \rangle_{\mathbf{q}} \langle a_{i\lambda} \rangle \\ &+ \frac{1}{i} \sum_{j \neq i} \sum_{\sigma} \int_{-\infty}^0 dt_1 e^{et_1} \langle [\varphi_{\lambda}(i), \varphi_{\sigma}(j, t_1)] \rangle_{\mathbf{q}} \langle a_{j\sigma}^+ a_{j\sigma} a_{i\lambda} \rangle_{\mathbf{q}}. \end{aligned} \quad (20)$$

Following [9], we restrict ourselves in [20] to the linear term in  $\langle a_{i\lambda} \rangle$  (this approximation is discussed in [9]):

$$i \frac{d\langle a_{i\lambda} \rangle}{dt} = E_{i\lambda}(t) \langle a_{i\lambda} \rangle + \frac{1}{i} \int_{-\infty}^0 dt_1 e^{et_1} \langle \varphi_{\lambda}(i), \varphi_{\lambda}(i, t_1) \rangle_{\mathbf{q}} \langle a_{i\lambda} \rangle. \quad (21)$$

Introducing the spectral intensity  $\langle \varphi_{\lambda}(i) | \varphi_{\lambda}(i) \rangle_{\omega}$  (see [8, 16])

$$\langle \varphi_{\lambda}(i) | \varphi_{\lambda}(i) \rangle_{\omega} = \int_{-\infty}^{\infty} \langle \varphi_{\lambda}(i), \varphi_{\lambda}(i, t) \rangle_{\mathbf{q}} e^{i\omega t} dt,$$

we transform Eq. (21) to the form

$$i \frac{d \langle a_{i\lambda} \rangle}{dt} = E_\lambda(i) \langle a_{i\lambda} \rangle + K_\lambda(i) \langle a_{i\lambda} \rangle, \quad (22)$$

where

$$K_\lambda(i) = \int_{-\infty}^{\infty} d\omega \frac{\langle \varphi_\lambda(i) | \varphi_\lambda(i) \rangle_\omega}{\omega + i\varepsilon}. \quad (23)$$

Note that  $\langle \varphi_\lambda(i) | \varphi_\lambda(i) \rangle_\omega \geq 0$ . We now introduce the energy shift  $\Delta E_\lambda(i)$  and the damping  $\Gamma_\lambda(i)$  of the Zeeman level  $\lambda$  by means of the equations

$$\begin{aligned} K_\lambda(i) &= \Delta E_\lambda(i) - \frac{i\Gamma_\lambda(i)}{2}, \\ \Delta E_\lambda(i) &= P \int_{-\infty}^{\infty} \frac{d\omega}{\omega} \langle \psi_\lambda(i) | \varphi_\lambda(i) \rangle_\omega \ll E_\lambda(i), \\ \Gamma_\lambda(i) &= 2\pi \langle \varphi_\lambda(i) | \varphi_\lambda(i) \rangle_{\omega=0}. \end{aligned} \quad (24)$$

It is expedient to replace  $\Gamma_\lambda(i)$  by the averaged quantity

$$\Gamma_\lambda = \frac{1}{N_0} \sum_i \Gamma_\lambda(i), \quad (26)$$

where  $N_0 = 4N$  is the total number of molecules in the crystal and  $N$  is the number of unit cells. Using (13) and (13a), we obtain expressions for the damping (26):

$$\Gamma_0 = \frac{2\pi}{N_0} \cdot d^2 \sum_i \langle O_i^0 | O_i^0 \rangle_{\omega=0}, \quad (27)$$

$$\Gamma_{\pm 1} = \frac{2\pi}{N_0} \sum_i \left\{ \frac{d^2}{4} \langle O_i^0 | O_i^0 \rangle_{\omega=0} \mp cd \langle O_i^0 | J_i^z \rangle_{\omega=0} + c^2 \langle J_i^z | J_i^z \rangle_{\omega=0} \right\}. \quad (28)$$

These expressions are written down in coordinate systems in which the external magnetic field  $H_0$  is the quantization axis for the nuclear spin moment  $I_i$  and for the mechanical angular momentum  $J_i$ . In the orientationally ordered phase the quantization axis  $z$  of the angular momentum  $J_i^z$  coincides with one of the four principal diagonals of the cube of the face-centered cubic lattice. We must therefore go over from a system of coordinates associated with the external magnetic field to a system of coordinates associated with sublattices. One can show that the expressions (27) and (28) can be written in the following form in a sublattice coordinate system:

$$\Gamma_0 = \frac{2\pi}{N_0} d^2 \sum_{\alpha, \mathbf{R}_\alpha} \sum_{m, n=-2}^2 R_{nm}(\omega_\alpha) \langle O^m(\mathbf{R}_\alpha) | O^n(\mathbf{R}_\alpha) \rangle_{\omega=0}, \quad (29)$$

$$\begin{aligned} \Gamma_{\pm 1} &= \frac{2\pi}{N_0} \sum_{\alpha, \mathbf{R}_\alpha} \left\{ \frac{d^2}{4} \sum_{m, n=-2}^2 R_{nm}(\omega_\alpha) \langle O^m(\mathbf{R}_\alpha) | O^n(\mathbf{R}_\alpha) \rangle_{\omega=0} \right. \\ &\quad \left. \mp cd \sum_{n=-1}^1 \sum_{m=-2}^2 S_{nm}(\omega_\alpha) \langle O^m(\mathbf{R}_\alpha) | J^n(\mathbf{R}_\alpha) \rangle_{\omega=0} + c^2 \sum_{m, n=-1}^1 T_{nm}(\omega_\alpha) \langle J^m(\mathbf{R}_\alpha) | J^n(\mathbf{R}_\alpha) \rangle_{\omega=0} \right\}, \end{aligned} \quad (30)$$

where  $\alpha = 1, 2, 3, 4$  is the lattice subscript and  $i \rightarrow (\alpha, \mathbf{R}_\alpha)$ . The transformation matrices have the form

$$R_{nm}(\omega_\alpha) = k_n k_m D_{0n}^{2*}(\omega_\alpha) D_{0m}^{2*}(\omega_\alpha), \text{ a } (5 \times 5) \text{ matrix,}$$

$$S_{nm}(\omega_\alpha) = -l_n k_m D_{0n}^{1*}(\omega_\alpha) D_{0m}^{2*}(\omega_\alpha), \text{ a } (3 \times 5) \text{ matrix,}$$

$$T_{nm}(\omega_\alpha) = l_n l_m D_{0n}^{1*}(\omega_\alpha) D_{0m}^{1*}(\omega_\alpha), \text{ a } (3 \times 3) \text{ matrix,}$$

$$D_{0m}^{1*}(\omega_\alpha) = \left( \frac{4\pi}{2l+1} \right)^{1/2} (-1)^m Y_{lm}(\theta_\alpha, 0).$$

The coefficients  $k_m$  and  $l_n$  have the values:  $k_0 = -1$ ;  $k_{\pm 1} = \pm\sqrt{3/2}$ ;  $k_{\pm 2} = -\sqrt{3/2}$ ;  $l_0 = 1$ ;  $l_{\pm 1} = \mp 1/2$ ;  $\omega_\alpha = (\theta_\alpha, \varphi_\alpha)$  are the axial and azimuthal angles of the quantization axis in the sublattice  $\alpha$  in the coordinate system associated with the external magnetic field.

5. Correlation Functions for the Libron Subsystem  
in the Bose Approximation

We now turn to the calculation of the spectral intensities of the libron subsystem correlation functions in the expressions for the damping (29) and (30).

We start from the Hamiltonian (1)

$$\mathcal{H}_L = \sum_{ij} \sum_{m,n=-2}^2 \gamma_{ij}^{mn} O_i^m O_j^n.$$

Following [1], we introduce the operators

$$\begin{aligned} a_i &= (1/\sqrt{2}) [1 - (J_i^z)^2] J_i^-, \\ b_i &= (1/\sqrt{2}) [1 - (J_i^z)^2] J_i^+ \end{aligned} \quad (31)$$

and the complex conjugate operators  $a_i^+$  and  $b_i^+$ . If these operators act on a state with  $J_i^z = 0$ , they map it to a state with  $J_i^z = \pm 1$ :

$$\begin{aligned} a_i^+ |J_i^z = 0\rangle &= |J_i^z = +1\rangle, \\ b_i^+ |J_i^z = 0\rangle &= |J_i^z = -1\rangle. \end{aligned}$$

One can show that these operators satisfy the commutation relations [1]

$$\begin{aligned} [a_i, a_j^+] &= (1 - 2n_{ai} - n_{bi}) \delta_{ij}, \\ [b_i, b_j^+] &= (1 - n_{ai} - 2n_{bi}) \delta_{ij}, \\ [a_i, b_j^+] &= -b_i^+ a_i \delta_{ij}; \quad [b_i, a_j^+] = -a_i^+ b_i \delta_{ij}, \\ [a_i, b_j] &= [a_i^+, b_j] = 0. \end{aligned} \quad (32)$$

The operators  $J_i^\pm$  and  $J_i^z$  can be expressed in terms of  $a_i$  and  $b_i$ :

$$\begin{aligned} J_i^z &= a_i^+ a_i - b_i^+ b_i; \quad (J_i^z)^2 = a_i^+ a_i + b_i^+ b_i, \\ J_i^+ &= \sqrt{2}(a_i^+ + b_i); \quad J_i^- = \sqrt{2}(a_i + b_i^+), \end{aligned} \quad (33)$$

and thus, by virtue of (2),

$$\begin{aligned} O_i^0 &= 3(a_i^+ a_i + b_i^+ b_i) - 2, \\ O_i^{+1} &= \sqrt{2}(a_i^+ - b_i); \quad O_i^{-1} = (O_i^{+1})^+, \\ O_i^{+2} &= 2a_i^+ b_i; \quad O_i^{-2} = (O_i^{+2})^+. \end{aligned} \quad (33a)$$

In calculating the spectral intensities in (29) and (30) we shall restrict ourselves to the approximation when  $a_i$ ,  $a_i^+$ ,  $b_i$ , and  $b_i^+$  can be regarded as operators that satisfy Bose commutation relations:

$$\begin{aligned} [a_i, a_j^+] &= [b_i, b_j^+] = \delta_{ij}, \\ [a_i, b_j^+] &= [b_i, a_j^+] = [a_i, b_i] = [a_i^+, b_i^+] = 0. \end{aligned} \quad (34)$$

This corresponds to the assumption  $\langle n_a \rangle \ll 1$  and  $\langle n_b \rangle \ll 1$ , which is valid at low temperatures. Substituting the operators (33a) into the Hamiltonian (1) and restricting ourselves to bilinear terms, we obtain

$$\mathcal{H}_L^{(2)} = 2 \sum_{ij} \{ \gamma_{ij}^{00} [2 - 3(a_j^+ a_j + b_j^+ b_j)] + \gamma_{ij}^{+1,-1} (a_i^+ - b_i) (a_j - b_j^+) + \gamma_{ij}^{+1,+1} (a_i^+ - b_i) (a_j^+ - b_j) \} + \text{Hermitian conjugate.} \quad (35)$$

We now go over to the Fourier representation for  $a_i$  and  $b_i$ :

$$\begin{aligned} a(\mathbf{R}_\alpha) &= \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{i\mathbf{k}\mathbf{R}_\alpha} a_\alpha(\mathbf{k}), \\ b(\mathbf{R}_\alpha) &= \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{i\mathbf{k}\mathbf{R}_\alpha} b_\alpha(\mathbf{k}). \end{aligned} \quad (36)$$

We also have

$$\begin{aligned} \gamma_{\alpha\beta}^{+1,-1}(\mathbf{R}_\alpha - \mathbf{R}_\beta') &= \frac{19\Gamma}{4} \frac{1}{N} \sum_{\mathbf{k}} e^{-i\mathbf{k}(\mathbf{R}_\alpha - \mathbf{R}_\beta')} f_{\alpha\beta}(\mathbf{k}), \\ \gamma_{\alpha\beta}^{+1,+1}(\mathbf{R}_\alpha - \mathbf{R}_\beta') &= \frac{19\Gamma}{4} \frac{1}{N} \sum_{\mathbf{k}} e^{-i\mathbf{k}(\mathbf{R}_\alpha - \mathbf{R}_\beta')} g_{\alpha\beta}(\mathbf{k}), \end{aligned} \quad (37)$$

where we have restricted ourselves to the nearest-neighbor approximation and

$$\sum_{\mathbf{R}_\beta'} \gamma_{\alpha\beta}^{00}(\mathbf{R}_\alpha - \mathbf{R}_\beta') = -\frac{19\Gamma}{12}.$$

Using (36) and (37) we can represent the Hamiltonian (35) as

$$\begin{aligned} \mathcal{H}_L^{(2)} = & 19\Gamma \sum_{\alpha, \mathbf{k}} \{a_{\alpha^+}(\mathbf{k}) a_\alpha(\mathbf{k}) + b_{\alpha^+}(\mathbf{k}) b_\alpha(\mathbf{k})\} + \frac{19\Gamma}{2} \sum_{\alpha, \beta, \mathbf{k}} \{g_{\alpha\beta}(\mathbf{k}) [a_{\alpha^+}(-\mathbf{k}) \\ & - b_\alpha(\mathbf{k})] [a_{\beta^+}(\mathbf{k}) - b_\beta(-\mathbf{k})] + g_{\alpha\beta}^*(\mathbf{k}) [a_\alpha(\mathbf{k}) + b_{\alpha^+}(-\mathbf{k})] [a_\beta(-\mathbf{k}) \\ & - b_{\beta^+}(\mathbf{k})] + f_{\alpha\beta}(\mathbf{k}) [a_{\alpha^+}(-\mathbf{k}) - b_\alpha(\mathbf{k})] [a_\beta(-\mathbf{k}) - b_{\beta^+}(\mathbf{k})] + f_{\alpha\beta}^*(\mathbf{k}) \\ & \times [a_\alpha(\mathbf{k}) - b_{\alpha^+}(-\mathbf{k})] [a_{\beta^+}(\mathbf{k}) - b_\beta(-\mathbf{k})]\}. \end{aligned} \quad (38)$$

This Hamiltonian can be diagonalized [8] and one can then find the spectrum of the collective excitations known as librions. Using Wick's theorem, one can calculate the necessary correlation functions for the libron subsystem. This can be done as follows. The canonical transformation

$$\begin{aligned} a_\alpha(\mathbf{k}) &= \sum_{\mu} \{u_{\alpha\mu}(\mathbf{k}) c_\mu(\mathbf{k}) + v_{\alpha\mu}(\mathbf{k}) c_{\mu^+}(-\mathbf{k})\}, \\ b_\alpha(\mathbf{k}) &= \sum_{\mu} \{u_{\alpha\mu}^*(\mathbf{k}) c_\mu(\mathbf{k}) + v_{\alpha\mu}^*(\mathbf{k}) c_{\mu^+}(-\mathbf{k})\}, \end{aligned} \quad (39)$$

where  $c_\mu(\mathbf{k})$  and  $c_{\mu^+}(\mathbf{k})$  are new Bose operators, reduces the Hamiltonian (38) to the diagonal form

$$\mathcal{H}_L^{(2)} = \sum_{\mu, \mathbf{k}} \varepsilon_\mu(\mathbf{k}) c_{\mu^+}(\mathbf{k}) c_\mu(\mathbf{k}) + E_0, \quad (40)$$

where

$$\varepsilon_\mu(\mathbf{k}) = 19\Gamma [1 + 2\varphi_\mu(\mathbf{k})]^{1/2} = 19\Gamma \omega_\mu(\mathbf{k}) \quad (40a)$$

is the libron spectrum;  $\omega_\mu(\mathbf{k})$  are the libron frequencies,  $\mu = 1, 2, \dots, 8$ .

$$E_0 = \frac{1}{2} \sum_{\mu, \mathbf{k}} \varepsilon_\mu(\mathbf{k}) - \frac{5N_0}{6} 19\Gamma.$$

The quantities  $u_{\alpha\mu}(\mathbf{k})$  and  $v_{\alpha\mu}(\mathbf{k})$  have the form

$$\begin{aligned} u_{\alpha\mu}(\mathbf{k}) &= \frac{1}{2} V_{\alpha\mu}(\mathbf{k}) \frac{\omega_\mu(\mathbf{k}) - 1}{\sqrt{\omega_\mu(\mathbf{k})}}, \\ v_{\alpha\mu}(\mathbf{k}) &= \frac{1}{2} V_{\alpha\mu}(\mathbf{k}) \frac{\omega_\mu(\mathbf{k}) + 1}{\sqrt{\omega_\mu(\mathbf{k})}}. \end{aligned}$$

Here  $V_{\alpha\mu}(\mathbf{k})$  are the elements of a unitary matrix and they satisfy the equations

$$\begin{aligned} \sum_{\alpha} \{V_{\alpha\mu}^* V_{\alpha\lambda} + V_{\alpha\mu} V_{\alpha\lambda}^*\} &= \delta_{\mu\lambda}, \\ \sum_{\mu} V_{\alpha\mu} V_{\beta\mu}^* &= \delta_{\alpha\beta}; \quad \sum_{\mu} V_{\alpha\mu} V_{\beta\mu} = \sum_{\mu} V_{\alpha\mu}^* V_{\beta\mu}^* = 0, \end{aligned}$$

and also

$$\begin{aligned} \sum_{\alpha\beta} \{V_{\alpha\mu}^*(\mathbf{k}) V_{\beta\lambda}(\mathbf{k}) f_{\alpha\beta}(\mathbf{k}) - V_{\alpha\mu}^*(\mathbf{k}) V_{\beta\lambda}(\mathbf{k}) g_{\alpha\beta}(\mathbf{k}) \\ + V_{\alpha\beta}(\mathbf{k}) V_{\beta\lambda}^*(\mathbf{k}) f_{\alpha\beta}^*(\mathbf{k}) - V_{\alpha\mu}(\mathbf{k}) V_{\beta\lambda}(\mathbf{k}) g_{\alpha\beta}^*(\mathbf{k})\} &= \varphi_\lambda(\mathbf{k}) \delta_{\mu\lambda}. \end{aligned}$$

The operators  $a_{\alpha}(\mathbf{k})$  and  $b_{\alpha}(\mathbf{k})$  can be expressed in terms of  $c_{\mu}(\mathbf{k})$  and  $c_{\mu^+}(\mathbf{k})$ :

$$\begin{aligned} a_\alpha(\mathbf{k}) &= \frac{1}{2} \sum_{\mu} V_{\alpha\mu}(\mathbf{k}) \frac{1}{\sqrt{\omega_\mu(\mathbf{k})}} \{[\omega_\mu(\mathbf{k}) + 1] c_\mu(\mathbf{k}) + [\omega_\mu(\mathbf{k}) - 1] c_{\mu^+}(-\mathbf{k})\}, \\ b_\alpha(\mathbf{k}) &= \frac{1}{2} \sum_{\mu} V_{\alpha\mu}^*(\mathbf{k}) \frac{1}{\sqrt{\omega_\mu(\mathbf{k})}} \{[\omega_\mu(\mathbf{k}) + 1] c_\mu(\mathbf{k}) + [\omega_\mu(\mathbf{k}) - 1] c_{\mu^+}(-\mathbf{k})\}. \end{aligned} \quad (41)$$

We shall now show how the correlation functions are calculated. As an example, consider  $\langle O^0(\mathbf{R}_\alpha) | O^0(\mathbf{R}_\alpha) \rangle_{\omega=0}$

$$\sum_{\mathbf{R}_\alpha} \langle O^0(\mathbf{R}_\alpha) | O^0(\mathbf{R}_\alpha) \rangle_{\omega=0} = \sum_{\mathbf{k}} \langle O_{\alpha^+}^0(\mathbf{k}) | O_{\alpha^+}^0(-\mathbf{k}) \rangle_{\omega=0} = \sum_{\mathbf{k}, \mathbf{k}_1} \int_{-\infty}^{\infty} d\omega \{ \langle a_{\alpha^+}(\mathbf{k}) | a_{\alpha^+}(-\mathbf{k}) \rangle_{-\omega} \langle a_\alpha(-\mathbf{k}_1) | a_\alpha(\mathbf{k}_1) \rangle_{\omega} \}$$

$$\begin{aligned}
& + \langle a_\alpha(\mathbf{k}) | a_\alpha^+(\mathbf{k}) \rangle_{-\omega} \langle a_\alpha^+(\mathbf{k}_1) | a_\alpha(\mathbf{k}_1) \rangle_\omega \\
& + \langle b_\alpha^+(\mathbf{k}) | b_\alpha^+(-\mathbf{k}) \rangle_{-\omega} \langle b_\alpha(-\mathbf{k}_1) | b_\alpha(\mathbf{k}_1) \rangle_\omega \\
& + \langle b_\alpha(\mathbf{k}) | b_\alpha^+(\mathbf{k}) \rangle_{-\omega} \langle b_\alpha^+(\mathbf{k}_1) | b_\alpha(\mathbf{k}_1) \rangle_\omega \\
& + \langle a_\alpha^+(\mathbf{k}) | b_\alpha^+(-\mathbf{k}) \rangle_{-\omega} \langle a_\alpha(-\mathbf{k}_1) | b_\alpha(\mathbf{k}_1) \rangle_\omega \\
& + \langle a_\alpha(\mathbf{k}) | b_\alpha^+(\mathbf{k}) \rangle_{-\omega} \langle a_\alpha^+(\mathbf{k}_1) | b_\alpha(\mathbf{k}_1) \rangle_\omega \\
& + \langle b_\alpha^+(\mathbf{k}) | a_\alpha^+(-\mathbf{k}) \rangle_{-\omega} \langle b_\alpha(-\mathbf{k}_1) | a_\alpha(\mathbf{k}_1) \rangle_\omega \\
& + \langle b_\alpha(\mathbf{k}) | a_\alpha^+(\mathbf{k}) \rangle_{-\omega} \langle b_\alpha^+(\mathbf{k}_1) | a_\alpha(\mathbf{k}_1) \rangle_\omega.
\end{aligned} \tag{42}$$

Going over to the operators  $c_\mu(\mathbf{k})$  and  $c_\mu^+(\mathbf{k})$ , one can calculate the spectral intensities in (42). As an example, we shall give the two following spectral intensities:

$$\begin{aligned}
& \langle a_\alpha^+(\mathbf{k}) | a_\alpha^+(-\mathbf{k}) \rangle_\omega = \int_{-\infty}^{\infty} \langle a_\alpha^+(-\mathbf{k}) a_\alpha^+(\mathbf{k}, t) \rangle e^{i\omega t} dt \\
& = \frac{1}{4} \sum_{\mu} (V_{\alpha\mu}^*(\mathbf{k}))^2 \frac{\omega_\mu^2(\mathbf{k}) - 1}{\omega_\mu(\mathbf{k})} \left\{ \frac{\delta(\omega - \varepsilon_\mu(\mathbf{k}))}{e^{\beta\varepsilon_\mu(\mathbf{k})} - 1} + \frac{e^{\beta\varepsilon_\mu(\mathbf{k})} \delta(\omega + \varepsilon_\mu(\mathbf{k}))}{e^{\beta\varepsilon_\mu(\mathbf{k})} - 1} \right\},
\end{aligned} \tag{43}$$

$$\langle a_\alpha(\mathbf{k}) | b_\alpha^+(\mathbf{k}) \rangle_\omega = \frac{1}{4} \sum_{\mu} V_{\alpha\mu}^2(\mathbf{k}) \left\{ \frac{(\omega_\mu(\mathbf{k}) + 1)^2}{\omega_\mu(\mathbf{k})} \frac{\delta(\omega - \varepsilon_\mu(\mathbf{k}))}{(e^{\beta\varepsilon_\mu(\mathbf{k})} - 1)} + \frac{(\omega_\mu(\mathbf{k}) - 1)^2 e^{\beta\varepsilon_\mu(\mathbf{k})}}{\omega_\mu(\mathbf{k})} \frac{\delta(\omega + \varepsilon_\mu(\mathbf{k}))}{(e^{\beta\varepsilon_\mu(\mathbf{k})} - 1)} \right\}. \tag{43a}$$

## 6. Results

One can verify that the expressions (29) and (30) for the damping  $\Gamma_0$  and  $\Gamma_{\pm}$ , in which the spectral intensities are calculated according to type by (42), (43), and (43a), have the form

$$\Gamma_0 = \frac{2\pi d^2}{N^2} \sum_{\mathbf{k}, \mathbf{k}_1} \sum_{\mu, \mu'} F_{\mu\mu'}^{(+)}(\mathbf{k}, \mathbf{k}_1) G_{\mu\mu'}^{(1)}(\mathbf{k}, \mathbf{k}_1), \tag{44}$$

where

$$\begin{aligned}
& F_{\mu\mu'}^{(+)}(\mathbf{k}, \mathbf{k}_1) = \delta(\varepsilon_\mu(\mathbf{k}) - \varepsilon_{\mu'}(\mathbf{k}_1)) \frac{\omega_\mu(\mathbf{k}) \omega_{\mu'}(\mathbf{k}_1) + 1}{\omega_\mu(\mathbf{k}) \omega_{\mu'}(\mathbf{k}_1) (e^{\beta\varepsilon_\mu(\mathbf{k})} - 1) (e^{\beta\varepsilon_{\mu'}(\mathbf{k}_1)} - 1)} \\
& \times \{ (\omega_\mu(\mathbf{k}) + 1) (\omega_{\mu'}(\mathbf{k}_1) + 1) e^{\beta\varepsilon_\mu(\mathbf{k})} + (\omega_\mu(\mathbf{k}) - 1) (\omega_{\mu'}(\mathbf{k}_1) - 1) e^{\beta\varepsilon_{\mu'}(\mathbf{k}_1)} \}
\end{aligned} \tag{45}$$

and

$$\begin{aligned}
& G_{\mu\mu'}^{(1)}(\mathbf{k}, \mathbf{k}_1) = \frac{1}{4} \sum_{\alpha} \left\{ \frac{9}{4} R_{00}(\omega_\alpha) [ |V_{\alpha\mu}(\mathbf{k})|^2 |V_{\alpha\mu'}(\mathbf{k}_1)|^2 + V_{\alpha\mu}^2(\mathbf{k}) V_{\alpha\mu'}^2(\mathbf{k}_1) ] \right. \\
& \quad \left. + 6R_{2,0}(\omega_\alpha) [ |V_{\alpha\mu}(\mathbf{k})|^2 (V_{\alpha\mu}^2(\mathbf{k}_1) + V_{\alpha\mu'}^2(\mathbf{k}_1)) ] \right. \\
& \quad \left. + \frac{1}{2} R_{+2,+2}(\omega_\alpha) [ V_{\alpha\mu}^2(\mathbf{k}) V_{\alpha\mu'}^2(\mathbf{k}_1) + V_{\alpha\mu}^2(\mathbf{k}) V_{\alpha\mu'}^2(\mathbf{k}_1) ] + R_{-2,-2}(\omega_\alpha) [ |V_{\alpha\mu}(\mathbf{k})|^2 |V_{\alpha\mu'}(\mathbf{k}_1)|^2 ] \right\}
\end{aligned} \tag{46}$$

is the factor that takes into account the sublattice structure of the orthohydrogen crystal:

$$\Gamma_{\pm 1} = \frac{2\pi}{N^2} \sum_{\mathbf{k}, \mathbf{k}_1} \sum_{\mu, \mu'} \left\{ \frac{d^2}{4} F_{\mu\mu'}^{(+)}(\mathbf{k}, \mathbf{k}_1) G_{\mu\mu'}^{(1)}(\mathbf{k}, \mathbf{k}_1) + \frac{c^2}{4} F_{\mu\mu'}^{(-)}(\mathbf{k}, \mathbf{k}_1) G_{\mu\mu'}^{(2)}(\mathbf{k}, \mathbf{k}_1) \right\}, \tag{47}$$

where

$$\begin{aligned}
& F_{\mu\mu'}^{(-)}(\mathbf{k}, \mathbf{k}_1) = \delta(\varepsilon_\mu(\mathbf{k}) - \varepsilon_{\mu'}(\mathbf{k}_1)) \frac{\omega_\mu(\mathbf{k}) + \omega_{\mu'}(\mathbf{k}_1)}{\omega_\mu(\mathbf{k}) \omega_{\mu'}(\mathbf{k}_1) (e^{\beta\varepsilon_\mu(\mathbf{k})} - 1) (e^{\beta\varepsilon_{\mu'}(\mathbf{k}_1)} - 1)} \\
& \times \{ (\omega_\mu(\mathbf{k}) + 1) (\omega_{\mu'}(\mathbf{k}_1) + 1) e^{\beta\varepsilon_\mu(\mathbf{k})} - (\omega_\mu(\mathbf{k}) - 1) (\omega_{\mu'}(\mathbf{k}_1) - 1) e^{\beta\varepsilon_{\mu'}(\mathbf{k}_1)} \}
\end{aligned} \tag{48}$$

and

$$G_{\mu\mu'}^{(2)}(\mathbf{k}, \mathbf{k}_1) = \frac{1}{4} \sum_{\alpha} T_{00}(\omega_\alpha) [ V_{\alpha\mu}^2(\mathbf{k}) V_{\alpha\mu'}^2(\mathbf{k}_1) - |V_{\alpha\mu}(\mathbf{k})|^2 |V_{\alpha\mu'}(\mathbf{k}_1)|^2 ]$$

is a factor that depends on the lattice structure. To separate explicitly the temperature dependence we shall assume that the dependence of  $G_{\mu\mu'}^{(1,2)}(\mathbf{k}, \mathbf{k}_1)$  on  $\mathbf{k}$  and  $\mathbf{k}_1$  can be neglected. We introduce the spectrum density (see also [5, 7]) whose numerical values are given in [4]:

$$g(\omega) = \frac{1}{8} \frac{\Omega}{(2\pi)^3} \int_{\omega_\mu(\mathbf{k})=\omega} \frac{dS}{|\nabla_{\mathbf{k}} \omega_\mu(\mathbf{k})|}, \tag{49}$$

where  $\Omega$  is the volume of the unit cell. Going over from summation to integration in (45) and (47):



$$\frac{1}{N} \sum_{\mathbf{k}} \rightarrow \frac{\Omega}{(2\pi)^3} \int_{\Omega^*} d^3k,$$

where  $\Omega^*$  is the volume of the first Brillouin zone, we finally obtain

$$\Gamma_0 = \frac{2\pi d^2}{19\Gamma} \cdot 64 \sum_{\mu\mu'} F_{\mu\mu'}^{(+)}(\beta) G_{\mu\mu'}^{(1)}, \quad (50)$$

$$\Gamma_{\pm 1} = \frac{2\pi d^2}{19\Gamma} \cdot 16 \cdot \sum_{\mu\mu'} F_{\mu\mu'}^{(+)}(\beta) G_{\mu\mu'}^{(1)} + \frac{2\pi c^2}{19\Gamma} \cdot 64 \sum_{\mu\mu'} F_{\mu\mu'}^{(-)}(\beta) G_{\mu\mu'}^{(2)}, \quad (51)$$

where

$$F_{\mu\mu'}^{(\pm)}(\beta) = 2 \int_{\omega_{\min}}^{\omega_{\max}} \frac{d\omega e^{\beta 19\Gamma\omega}}{(e^{\beta 19\Gamma\omega} - 1)^2} \left\{ \frac{(\omega^2 + 1)^2/\omega^2}{1} \right\} g_{\mu}(\omega) g_{\mu'}(\omega), \quad (52)$$

$(\omega_{\max} - \omega_{\min})$  is the width of the band of libron waves [4];  $F_{\mu\mu'}^{(\pm)}(\beta)$  are functions that reflect the temperature dependence of the Zeeman level widths of the nuclear spins;  $G_{\mu\mu'}^{(1,2)}$  are quantities that take into account the complicated sublattice structure of the orthohydrogen crystal (see [5]). At low temperatures, (52) becomes

$$F_{\mu\mu'}^{(\pm)}(\beta) = 2 \int_{\omega_{\min}}^{\omega_{\max}} d\omega e^{-\beta 19\Gamma\omega} \left\{ \frac{(\omega^2 + 1)^2/\omega^2}{1} \right\} g_{\mu}(\omega) g_{\mu'}(\omega). \quad (52a)$$

Here,  $F_{\mu\mu'}^{(\pm)}$  and  $G_{\mu\mu'}^{(1,2)}$  must be calculated numerically.

## 7. Discussion

The physical content of our result is the following. If a system of nuclear spins ceases to be isolated, the Larmor frequencies become spread out. This means that the line of the magnetic resonance absorption acquires a certain width, approximately equal to  $\overline{\delta\nu}$ . The temperature and structure dependence follows from (44) and (47). It should be noted that the intermolecular dipole-dipole interaction and the interaction with the phonons should also be taken into account in a rigorous calculation of the width and form of the nuclear magnetic absorption line and not only the interactions which we have considered. In the considered temperature range (well below the phase transition point which is approximately at 1.5°K) the interaction with the librions is stronger than the intermolecular dipole-dipole interaction. It is well known that solid hydrogen belongs to the group of substances with a so-called fine structure of the resonance line ([17, 18] Ch. VII). In fact, the distance between the protons in the  $H_2$  molecule is  $0.75 \cdot 10^{-8}$  cm and the distance between the molecules in solid hydrogen is  $3.75 \cdot 10^{-8}$  cm. Since the dipole-dipole interaction decreases with the distance as  $R^{-3}$ , the hydrogen molecule can be assumed to be isolated in a first approximation and then one can calculate its energy levels in a constant magnetic field. The order of magnitude of the interaction between the nuclear system and the librions is determined by the quantities  $d$  and  $c$ , which characterize the intramolecular dipole-dipole and spin-orbit interaction responsible for the fine structure of the resonance line. For a rigid lattice these interactions are much stronger than the intermolecular dipole-dipole interaction. It has been found experimentally ([18], see also [15]) that at fairly high temperatures, when the molecules rotate almost freely, there is a single absorption line; this is evidently connected with averaging of the intramolecular interactions. With decreasing temperature the rotational motion of the orthohydrogen molecules becomes ordered, the intensity of this line decreases, and a fine structure begins to appear [15, 17-22]. Note that the intermolecular dipole-dipole interaction can be taken into account in the same way as the interaction (7).

Thus, using the nonequilibrium statistical operator method, one can fairly simply calculate the energy shift and the width of the Zeeman levels of the nuclear spins in solid orthohydrogen. It is clear that similar calculations can also be made for a number of other specific problems.

I should like to express my thanks to D. N. Zubarev and N. M. Plakida for helpful discussions.

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