perimental dependence of the amplitude of the β oscillations on H^{-1}.

The small-amplitude oscillations of the thermoelectric power with a period close to that of the β oscillations, observed in a wide range of angles, are clearly due to small-angle scattering and the resultant transfer of electrons from the second zone to the constant-energy surfaces of the four-cornered rings in the third zone. The periods of these oscillations are clearly governed by the extremal sections of the electron tubes.

It follows that the experimental data obtained in our study support the conclusion that a coherent situation in aluminum single crystals with **RRR ~ 20 000 is** realized over the whole p space even in $H \sim 100$ kOe and $T \sim 4°$K. This is the most important of our results.

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Theory of Mössbauer spectra in the presence of spin-spin relaxation

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Compared with other mechanisms, spin-spin relaxation substantially complicates the theory of Mössbauer spectra: the relaxation constants become frequency-dependent functions and, in addition, the number of relaxation parameters increases sharply. For systems of cubic symmetry and 2→0 nuclear transitions a method of analysis of the spectra is developed that makes it possible to extract the frequency dependence of the relaxation functions directly from the experimental data for single-crystal samples without invoking any theoretical models. The structure of these functions is analyzed using the method of moments. It is shown that in the case of dipole-dipole interaction the relaxation can be described with good accuracy by one function, the form of which can be established from experimental data obtained on polycrystalline samples.

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1. INTRODUCTION

The theory of Mössbauer resonance spectra that has been developed up to now (see, e.g., Refs. 1–6) is applicable only in those cases when the fluctuations of the magnetic and electric fields giving rise to relaxation of the spin of the electron shell of the Mössbauer atom are sufficiently rapid, so that the characteristic correlation time τ_c of a fluctuation is much shorter than either the relaxation time τ_r or the Larmor-precession time τ_L of the nuclear spin in the hyperfine magnetic field ($\tau_L = \hbar/A$, where A is the hyperfine-interaction constant).

Both these conditions are fulfilled by a large margin for processes such as spin-lattice relaxation[1,3,4] and relaxation via conduction electrons in metals,[5,6] and in these cases the Mössbauer spectra can be described with the aid of a certain set of relaxation constants $\{\gamma_i\}$. The approximation based on the use of the inequalities $\tau_c \ll \tau_r$ and $\tau_c \ll \tau_L$ is commonly called the "white-noise" approximation.

The position changes radically when we are dealing with spin-spin relaxation. In fact, in this case, τ_c and τ_r are determined by the same interaction (either dipole-dipole or exchange) between the spins of neighboring atoms. If the magnitude J of this interaction turns out to be much larger than the hyperfine-interaction energy the condition $\tau_c \ll \tau_L$ is fulfilled and we can use the white-noise approximation to describe the spectra.[7] Moreover, in irregular systems in the presence of an external magnetic field H, the time $\tau_c \simeq \hbar/\Delta g \mu_B H$ (Δg is the characteristic quantity defining the scatter in the components of the g-tensor), and at sufficiently large H it is possible to reduce τ_c sharply. In this situation the analysis of spin-spin relaxation is also a completely solved theoretical problem.[8] In the remaining cases, however, especially when $J \sim A$, substantial modification of the theory is required. It is completely obvious that the main feature of this new theory, adapted for the description of spin-spin relaxation, should be the appearance of relaxation parameters that are not constants, as in the white-noise approximation, but depend on the energy ω of the absorbed γ quanta. This fact is already well recognized, and, e.g., in Ref. 9 variants for the phenomenological description of the dependence of the parameters $\{\gamma_i\}$ on ω have been proposed.

The appearance of relaxation functions is not the only complication of the theory. On top of this, the number of independent relaxation parameters increases sharply. It is obvious that their number is determined by the symmetry of the environment, and also by the values of the spins of the Mössbauer nuclei in the ground and excited states. In the present work we have confined ourselves to analyzing the simplest situation—a system of cubic symmetry and nuclear transitions of the type $2 \to 0$. (The simpler situation with $1 \to 0$ transitions is not realized for the Mössbauer isotopes.) As we have been able to show, the relaxation process in the given case is determined by just two independent relaxation functions; moreover, it is shown (and this constitutes the main result of the paper) that these functions can be found directly from experimental data without invoking any theoretical models or assumptions about the form of these functions. Section 2 is devoted to this question. In Sec. 4 the properties of the relaxation functions are analyzed using the method of moments, and the simplest models are also briefly discussed.

2. FORMULATION OF THE PROBLEM AND DERIVATION OF THE GENERAL FORMULAS

We shall consider a Mössbauer atom with an electron shell with half-integer total angular momentum, situated in an environment of cubic symmetry. The crystal fields partially lift the degeneracy (with respect to the angular-momentum projections) of the ground term, and in the general case in a field of cubic symmetry the levels are grouped into Γ_6 and Γ_7 doublets and Γ_8 quartets. We shall assume that the ground state of the atom is a Γ_6 (or Γ_7) doublet, and that the excited states are separated from the ground state by an energy $\Delta \gg kT$ (T is the temperature of the sample), so that they can be completely ignored. For the Γ_6 and Γ_7 doublets the hyperfine interaction is isotropic:

$$H_{hf} = A_\varkappa \mathbf{S}_0 \mathbf{I}_\varkappa; \tag{1}$$

here $\varkappa = e, g$; I_g and I_e are the spin of the nucleus in the ground state and excited state, respectively, and S_0 is the effective spin of the doublet, equal to 1/2.

Owing to the interaction (1) hyperfine structure appears in the Mössbauer spectra, but the manner in which it is displayed depends to an important extent on the interaction of the electron spin with its environment. In this work we shall be interested in the influence of the magnetic interaction (exchange or dipole-dipole) between the atomic spins on the spectra of the hyperfine structure. We find it convenient for the following to split the Hamiltonian of the spin-spin interaction into two parts—the interaction between the atoms surrounding the Mössbauer atom:

$$H_B = \frac{1}{2} \sum_{r,r'} J_{rr'}^{ij} S_r^i S_{r'}^j \tag{2}$$

and the interaction between the Mössbauer atom and its environment:

$$H_{AB} = \sum_r J_{r0}^{ij} S_r^i S_0^j . \tag{3}$$

We assume now that resonance γ quanta in the direction of $\mathbf{k}$, with energy ω and polarization η, are incident on such a system.

The starting point of the analysis is usually the following formal expresssion for the absorption spectrum (see, e.g., Ref. 7):

$$\varphi(\omega, \mathbf{k}, \eta) = -\operatorname{Im} F(\omega, \mathbf{k}, \eta), \tag{4}$$

$$F(\omega, \mathbf{k}, \eta) = \sum_{\alpha,\beta} \rho_\alpha \cdot \frac{(\eta^* \mathbf{j}(\mathbf{k}))_{\alpha\beta} (\eta \mathbf{j}^+(\mathbf{k}))_{\beta\alpha}}{\omega - E_\beta + E_\alpha + i\Gamma/2} . \tag{5}$$

Here α (β) labels the state of the whole system when the nucleus is in the ground (excited) state, $E_{\alpha,\beta}$ are the energies corresponding to these states, $\mathbf{j}(\mathbf{k})$ is the operator of a Fourier component of the nuclear-current density, and ρ_α are the diagonal elements of the equilibrium density matrix

$$\rho = Z^{-1} e^{-H/kT}, \tag{6}$$

where H is the sum of the three interactions (1)–(3).

Before proceeding to the transformations of formula (5) we shall discuss the general properties of the function $F(\omega, k, \eta)$ that follow directly from (5). It is not difficult to see that, as a function of the variable ω, $F(\omega, \mathbf{k}, \eta)$ is analytic in the upper half-plane, and, consequently, the dispersion relation

$$F'(\omega,\mathbf{k},\eta)=-\frac{1}{\pi}\int\frac{F''(\omega',\mathbf{k},\eta)}{\omega-\omega'}d\omega' \tag{7}$$

holds for it. Here F' and F'' are the real and imaginary parts of the function F, and the integral is to be understood in the principal-value sense. As we shall see below, the relation (7) can be used effectively in the analysis of the experimental data.

Except in the region of ultralow temperatures, in the density matrix (6) we can always neglect the interaction H_{AB}, and, since in our case all three interactions H_{hf}, H_A and H_B are of the same order, we can assume that the matrix ρ is proportional to unity. In this situation we can write for F the following expression (see Ref. 7):

$$F(\omega,\mathbf{k},\eta)=(2I_g+1)^{-1}\sum_{\substack{m_g m_e\\ m_g' m_e'}}(\eta^*\mathbf{j}(\mathbf{k}))_{m_g m_e}\bar{G}(\omega+i\Gamma/2)_{m_e m_g;m_e' m_g'}(\eta\mathbf{j}^+(\mathbf{k}))_{m_e' m_g'} \tag{8}$$

$$\bar{G}(\omega+i\Gamma/2)_{m_e m_g;m_e' m_g'}=\sum_{\sigma\sigma'}\hat{G}(\omega+i\Gamma/2)_{\sigma m_e \sigma m_g;\sigma' m_e' \sigma' m_g'}, \tag{9}$$

$$\hat{G}(\omega+i\Gamma/2)=(\omega-\hat{L}_A-\hat{M}(\omega)+i\Gamma/2)^{-1}. \tag{10}$$

Here $\hat{L}_A$ is the Liouville superoperator, acting in the space of the operators a of subsystem A (the Mössbauer atom) in accordance with the rule

$$\hat{L}_A a=[H_{hf},a],$$

and its eigenvalues give us the frequencies of the hyperfine transitions in the absence of relaxation; $\hat{M}(\omega)$ is the relaxation superoperator, determining the renormalization of the positions and widths of the individual components of the spectrum. Its general structure is given by the expression (see Ref. 7)

$$\hat{M}(\omega)=P\hat{L}_{AB}Q(\omega-\hat{L}_A-\hat{L}_B-Q\hat{L}_{AB}Q+i\Gamma/2)^{-1}Q\hat{L}_{AB}P, \tag{11}$$

where P and $Q=1-P$ are projection operators (P denotes taking the trace over the variables of the environment, or, as they are commonly called, the variables of the heat bath), and $\hat{L}_B$ and $\hat{L}_{AB}$ are the Liouville superoperators corresponding to the Hamiltonians H_B and H_{AB}.

Since the spin of the ground state is equal to zero, the superoperator $\bar{G}(\omega)$ is transformed into an ordinary operator acting in the space of the functions $|m_e\rangle$, where $|m_e\rangle$ is the wavefunction of the nuclear excited state with spin projection equal to m_e along the axis of quantization.

From the functions $|m_e\rangle$ we construct linear combinations that are basis functions of irreducible representations of the cubic group:

$$|0\rangle\equiv|0\rangle,\quad |\mathrm{II}\rangle=2^{-1/2}(|2\rangle+|-2\rangle), \tag{12}$$

$$|\mathrm{I}\rangle=2^{-1/2}(|1\rangle+|-1\rangle),\quad |\bar{\mathrm{I}}\rangle=2^{-1/2}(|1\rangle-|-1\rangle),\quad |\widetilde{\mathrm{II}}\rangle=2^{-1/2}(|2\rangle-|-2\rangle). \tag{13}$$

The functions (12) realize the irreducible representation Γ_3 and the functions (13) realize the representation Γ_5. In the presence of cubic symmetry the matrix elements of $\bar{G}(\omega)$ within the same representation coincide, e.g.,

$$\langle 0|\bar{G}(\omega+i\Gamma/2)|0\rangle=\langle\mathrm{II}|\bar{G}(\omega+i\Gamma/2)|\mathrm{II}\rangle. \tag{14}$$

Using this fact, and also explicit expressions for the matrix elements of the operator $\mathbf{j}(\mathbf{k})$ in the specific case of $E2$-transitions, we easily find

$$\begin{gathered}\varphi(\omega,\mathbf{k},\eta)=-\mathrm{Im}\,\{{}^2/_5F_1(\omega)+{}^3/_5F_2(\omega)\}\\ +2[(k_x\eta_x)^2+(k_y\eta_y)^2+(k_z\eta_z)^2-{}^1/_5]\,\mathrm{Im}\,\{F_2(\omega)-F_1(\omega)\},\end{gathered} \tag{15}$$

$$F_1(\omega)=\langle\mathrm{II}|\bar{G}(\omega)|\mathrm{II}\rangle,\quad F_2(\omega)=\langle\mathrm{II}|\bar{G}(\omega)|\widetilde{\mathrm{II}}\rangle. \tag{16}$$

In formula (15) and below we shall mean by the vector $\mathbf{k}$ the unit vector along the direction of propagation of the γ quanta; the axes x, y and z are the crystallographic directions [100], [010] and [001], respectively. After averaging over the polarizations we obtain

$$\begin{gathered}\varphi(\omega,\mathbf{k})=-\mathrm{Im}\,\{{}^2/_5F_1(\omega)+{}^3/_5F_2(\omega)\}\\ +({}^3/_5-k_x^4-k_y^4-k_z^4)\,\mathrm{Im}\,\{F_2(\omega)-F_1(\omega)\},\end{gathered} \tag{17}$$

and, finally, for a polycrystalline sample we have

$$\varphi(\omega)=-\mathrm{Im}\,\{{}^2/_5F_1(\omega)+{}^3/_5F_2(\omega)\}. \tag{18}$$

Thus, the problem has been reduced to the determination of the diagonal elements of the operator $\bar{G}(\omega)$ in the states $|\mathrm{II}\rangle$ and $|\widetilde{\mathrm{II}}\rangle$. For this purpose it is necessary for us to proceed to the analysis of the structure of the superoperator $\hat{G}(\omega)$, which acts in a larger space, namely, the product $|m_e\rangle|\sigma\rangle\langle\sigma'|$, where $|\sigma\rangle$ is the space of the wavefunctions of the Γ_6 doublet and $\langle\sigma'|$ is its Hermitian-conjugate space. As the basis states of this space it is convenient to introduce the following:

$$|m_e\rangle E,\quad |m_e\rangle S_0^i. \tag{19}$$

Here E is the unit operator in the space of the spin variables:

$$E=|{}^1/_2\rangle\langle{}^1/_2|+|-{}^1/_2\rangle\langle-{}^1/_2|,$$

and S_0^i are the operators of the spin components:

$$\begin{aligned}S_0^z&={}^1/_2[|{}^1/_2\rangle\langle{}^1/_2|-|-{}^1/_2\rangle\langle-{}^1/_2|],\\ S_0^x&={}^1/_2[|{}^1/_2\rangle\langle-{}^1/_2|+|-{}^1/_2\rangle\langle{}^1/_2|],\\ S_0^y&=\frac{1}{2i}[|{}^1/_2\rangle\langle-{}^1/_2|-|-{}^1/_2\rangle\langle{}^1/_2|].\end{aligned}$$

We note that, according to its definition (11), the relaxation operator $\hat{M}(\omega)$ possesses the following property:

$$\hat{M}(\omega)|m_e\rangle E\equiv 0,\quad E\langle m_e|\hat{M}(\omega)\equiv 0. \tag{20}$$

We proceed now to determine the matrix element $\langle\mathrm{II}|\bar{G}(\omega)|\mathrm{II}\rangle$.

Taking (14) and (9) into account, we obtain

$$\begin{gathered}\langle\mathrm{II}|\bar{G}(\omega)|\mathrm{II}\rangle=\langle 0|\bar{G}(\omega)|0\rangle\equiv(0|\hat{G}(\omega)|0);\\ |0)=|0\rangle E.\end{gathered} \tag{21}$$

We shall find how many times the irreducible representations Γ_3 and Γ_5 are contained in the space (19) in which the superoperator $\hat{G}(\omega)$ acts. According to well-known results of group theory (see, e.g., Ref. 10),

$$(\Gamma_3+\Gamma_5)\times\Gamma_6\times\Gamma_6=\Gamma_2+2\Gamma_4+2\Gamma_3+3\Gamma_5. \tag{22}$$

Thus, to calculate the matrix element (21) it is sufficient to construct from the basis states (19) two orthogonal states transforming according to the representation Γ_3. They can be constructed by the standard rules,[10] but it is sufficient to bear in mind that they are invariant under rotations about the z axis and a second-order symmetry axis perpendicular to it. These conditions are satisfied by the states $|0)$ and $|\Theta_1) = (|1\rangle S_0^- + |-1\rangle S_0^+)$. Using this result and also the property (20) of the relaxation superoperator, we find that the latter appears in the final expression for $\langle 0|\overline{G}(\omega)|0\rangle$ only through the one relaxation function

$$\gamma_1(\omega) \equiv \gamma_{11}(\omega) = (\Theta_1|\hat{\Omega}(\omega)|\Theta_1). \tag{23}$$

Simple direct calculations give us, finally,

$$F(\omega) = \frac{\overline{\omega}+{}^1/_2A-\gamma_1(\omega)}{\overline{\omega}(\overline{\omega}+{}^1/_2A-\gamma_1(\omega))-{}^3/_2A^2} \tag{24}$$

where $\overline{\omega} = \omega + i\Gamma/2$.

It is a slightly more complicated matter to calculate the matrix element

$$\langle\widetilde{\text{II}}|\overline{G}(\omega)|\widetilde{\text{II}}\rangle = (\widetilde{\text{II}}|\hat{G}(\omega)|\widetilde{\text{II}}); \quad |\widetilde{\text{II}}) = |\widetilde{\text{II}}\rangle E.$$

According to (22), here we need to construct three states transforming according to the representation Γ_5. Besides the state $|\widetilde{\text{II}})$ we choose two others, invariant under the subgroup D_4:

$$\begin{aligned} |\Theta_2) &= 2\sqrt{{}^2/_3}|\widetilde{\text{II}}\rangle S_0^z + \sqrt{{}^1/_3}(|1\rangle S_0^+ - |-1\rangle S_0^-), \\ |\Theta_3) &= 2\sqrt{{}^1/_3}|\widetilde{\text{II}}\rangle S_0^z - \sqrt{{}^2/_3}(|1\rangle S_0^+ - |-1\rangle S_0^-). \end{aligned} \tag{25}$$

The states (25) have been chosen in such a way that $|\Theta_3)$ is an eigenfunction of the superoperator $\hat{L}_A$:

$$\hat{L}_A|\Theta_3) = A|\Theta_3).$$

In this subspace the matrix $\hat{G}^{-1}(\omega+i\Gamma/2)$ has the following structure:

$$\begin{pmatrix} \overline{\omega} & -\sqrt{{}^3/_2}A & 0 \\ -\sqrt{{}^3/_2}A & \overline{\omega}+{}^1/_2A-\gamma_{22} & -\gamma_{23} \\ 0 & -\gamma_{32} & \overline{\omega}-A-\gamma_{33} \end{pmatrix}$$

where

$$\gamma_{ik}(\omega) = (\Theta_i|\hat{\Omega}(\omega)|\Theta_k). \tag{26}$$

Finally, straightforward calculations lead to a result coinciding in form with (24):

$$F_2(\omega) = \frac{\overline{\omega}+{}^1/_2A-\gamma_2(\omega)}{\overline{\omega}(\overline{\omega}+{}^1/_2A-\gamma_2(\omega))-{}^3/_2A^2}, \tag{27}$$

$$\gamma_2(\omega) = \gamma_{22}(\omega) + \frac{\gamma_{23}(\omega)\gamma_{32}(\omega)}{\overline{\omega}-A-\gamma_{33}(\omega)}. \tag{28}$$

Thus, the formulas (15), (24) and (27) give us a general expression for the description of the relaxation spectra. It can be seen that the spin-spin relaxation process can be described by specifying only two functions: $\gamma_1(\omega)$ (23) and $\gamma_2(\omega)$ (28). In the case of fast relaxation, when $J \gg A$, the dispersion of the parameters $\gamma_{ik}(\omega)$ can be neglected. Moreover, in this case, γ_{23} and γ_{32} vanish and $\gamma_{11} = \gamma_{22}$. For the absorption spectrum we obtain the known expression[11]

$$\varphi(\omega) = -\operatorname{Im}\frac{\overline{\omega}+{}^1/_2A-\gamma}{\overline{\omega}(\overline{\omega}+{}^1/_2A-\gamma)-{}^3/_2A^2}. \tag{29}$$

We shall discuss the properties of the relaxation functions $\gamma_1(\omega)$ and $\gamma_2(\omega)$ in Sec. 4. We note only that the exact calculation of the frequency dependence of these relaxation parameters starting directly from the microscopic interactions (2) and (3) is a separate and still unsolved problem. Below we concentrate our attention on the following very important fact: the functions $\gamma_1(\omega)$ and $\gamma_2(\omega)$ can be found directly from the experimental data on the absorption spectra in monocrystalline samples. Indeed, performing the measurements for two different directions of incidence of the γ quantum on the crystal, according to (17) we can establish $F_1''(\omega)$ and $F_2''(\omega)$ independently. Using the dispersion relations (7) it is not difficult to find the real parts of the corresponding functions too. Consequently, the experimental data give us, in fact, the frequency behavior of the functions $F_1(\omega)$ and $F_2(\omega)$ as a whole. Inverting the formulas (24) and (27), we find

$$\gamma_{1,2}(\omega) = \overline{\omega} + \frac{A}{2} - \frac{1{,}5A^2F_{1,2}(\omega)}{\overline{\omega}F_{1,2}(\omega)-1}. \tag{30}$$

Thus, experimental data on the absorption spectra can be used directly to establish the frequency dependence of the relaxation parameters introduced above. This seems to us to be important, primarily because there exist difficulties in the theoretical calculation of these parameters that have not yet been overcome.

3. ALLOWANCE FOR INTERFERENCE EFFECTS [12,13]

The theory developed above has as its purpose an application to the analysis of the experimental data for the compound $Cs_2NaYbCl_6$ with the Mössbauer isotope ^{170}Yb. It is well known that, for the latter, interference between the photoeffect and internal-conversion processes accompanying the resonance absorption of γ quanta is manifested in the absorption spectra.[14] In the absence of relaxation processes the effect of this interference reduces to the appearance in formula (5) of an extra exponential factor of the type $e^{i2\xi}$,[15] where ξ is the so-called dispersion amplitude. It is not difficult to show that the presence of relaxation does not change this result. Therefore, allowance for the interference effects reduces to the following: in the formulas (4) and (7) we must replace $F(\omega)$ by

$$F(\omega) = F(\omega)e^{i2\xi}. \tag{31}$$

In the remaining formulas, including (30), $F_{1,2}(\omega)$ has the previous meaning.

4. ANALYSIS OF THE RELAXATION FUNCTIONS

As already noted above, there is still, as yet, no exact direct method of calculating functions of the type $\gamma_{1,2}(\omega)$, although a large amount of theoretical work has recently been carried out, with attempts to obtain analytic approximate formulas for functions of a similar type[16-18] or direct numerical calculations with the aid of a computer.[19,20] The problem of spin-spin relaxation has great significance for electron paramagnetic and nuclear magnetic resonance, and, amongst the methods developed to analyze this process, only the method of moments,[21,22] by virtue of its completeness, can be distinguished as giving the possibility of finding the integral characteristics of the relaxation functions from the Hamiltonians (2), (3). This is the method we shall use below to analyze the functions $\gamma_{1,2}(\omega)$ that arise in our case.

First of all we note that, according to the definitions (23), (26) and (11), we can represent the parameters $\gamma_{ik}(\omega)$ in the following form:

$$\gamma_{ik}(\omega)=\frac{1}{\pi}\int\frac{\tilde{\gamma}_{ik}(\omega')}{\omega-\omega'+i\Gamma/2}\,d\omega', \tag{32}$$

where

$$\tilde{\gamma}_{ik}(\omega')=\pi(\Theta_i|P\hat{L}_{AB}Q\delta(\omega-\hat{L}_A-\hat{L}_B-Q\hat{L}_{AB}Q)Q\hat{L}_{AB}P|\Theta_k). \tag{33}$$

Here δ is the delta-function, dependent on operator arguments. For the functions γ_{ik} it is not difficult to calculate the moments in accordance with the formula

$$\begin{aligned}M_{ik}^{(n)}&=\int\tilde{\gamma}_{ik}(\omega)\,\omega^n\,d\omega\\&=\pi(\Theta_i|P\hat{L}_{AB}Q(\hat{L}_A+\hat{L}_B+Q\hat{L}_{AB}Q)^nQ\hat{L}_{AB}P|\Theta_k).\end{aligned} \tag{34}$$

Taking into account that $\hat{L}_B$ and $\hat{L}_{AB}$ are Liouville superoperators, i.e., their action on an arbitrary operator reduces to the commutation operation, it is not difficult to calculate the first few moments $M_{ik}^{(n)}$. Thus, e.g., we shall find the zeroth moment of the function $\tilde{\gamma}_{11}(\omega)$. In accordance with (34),

$$M_{11}^{(0)}=\pi(\Theta_1|PL_{AB}^2P|\Theta_1).$$

We shall calculate first the result of the action of the superoperator $\hat{L}_{AB}$ on the state $|\Theta_1)$:

$$\begin{aligned}|\Phi_1)=\hat{L}_{AB}|\Theta_1)&=i(|1\rangle+|-1\rangle)\sum_r J_{0r}^{ij}S_r^j\varepsilon^{ixl}S_0^l\\&+i(|1\rangle-|-1\rangle)\sum_r J_{0r}^{ij}S_r^j\varepsilon^{iyl}S_0^l.\end{aligned}$$

Here ϵ^{ikl} is the antisymmetric unit tensor. Finally, for $M_{11}^{(0)}$ we have

$$M_{11}^{(0)}=\pi(\Phi_1|\Phi_1)=\frac{\pi}{8}\sum_r[2(J_{0r}^{ij})^2-(J_{0r}^{xj})^2-(J_{0r}^{yj})^2]. \tag{35}$$

In the case of dipole-dipole interaction,

$$M_{11}^{(0)}={}^1/_2\pi J_0^2a^{-6}S_1={}^1/_2\pi(\Delta\omega)^2. \tag{36}$$

Here a is the lattice constant, $J_0=(g\mu_B)^2$, and S_1 is the lattice sum, which, for lattices with a cubic structure, depends only on the relative (in units of a) distances $R_{rr'}$ between the atoms:

$$S_1=2\sum_r(R_{0r})^{-6}. \tag{37}$$

Thus, the problem has been reduced to the calculation of the standard lattice sums that arise in the analysis of the lineshapes in EPR and NMR spectra.[21,22] In formula (36) we have introduced the quantity $\Delta\omega$, which defines the energy scale of variation of the relaxation parameters. Thus, concrete calculations for the compound $Cs_2NaYbCl_6$ from the formulas (36) and (37) give $S_1=230$ and $\Delta\omega=1.2\times10^3$ MHz, or, in the units adopted in Mössbauer spectroscopy, $\Delta\omega=17$ mm/sec.

As regards the zeroth moments of the functions $\tilde{\gamma}_{22}(\omega)$ and $\tilde{\gamma}_{33}(\omega)$, these coincide exactly with $M_{11}^{(0)}$, while the zeroth moments of the functions γ_{23} and γ_{32} are identically equal to zero. Below we give only the results.

As direct calculations show,

$$M_{11}^{(1)}=M_{22}^{(1)}=M_{33}^{(1)}=-{}^1/_8A(\Delta\omega)^2\pi,\qquad M_{11}^{(2)}=M_{22}^{(2)}=M_{33}^{(2)}=M^{(2)}, \tag{38}$$

$$\begin{aligned}M^{(2)}/\pi={}^{13}/_{16}A^2(\Delta\omega)^2+{}^1/_2AJ_0^3a^{-9}S_2+{}^1/_{16}[10(\Delta\omega)^4-30J_0^4a^{-12}S_3\\+J_0^4a^{-12}S_4].\end{aligned} \tag{39}$$

Here, S_2, S_3 and S_4 are lattice sums of the form

$$S_2=\sum_r(R_{0r})^{-9}\approx280,\qquad S_3=\sum_r(R_{0r})^{-12}\approx720, \tag{40}$$

$$S_4=J_0^{-2}a^6\sum_{r\neq r'}(R_{rr'})^{-6}[{}^8/_3J_{0r}^{ij}J_{0r'}^{ij}-4R_{rr'}^mR_{rr'}^nJ_{0r}^{in}J_{0r'}^{im}]. \tag{41}$$

The sums S_2 and S_3 are computed trivially, and their approximate values for a face-centered cubic lattice are indicated in the formulas (40). The sum S_4 has a more complicated structure, but it too can be estimated easily if we write (41) in the simpler form

$$S_4=12\sum_{r\neq r'}(R_{rr'})^{-8}(R_{0r}R_{0r'})^{-2}\cos\varphi(1-\cos^2\varphi),$$

where φ is the angle between the vectors R_{0r} and $R_{0r'}$. Direct evaluation gives us

$$S_4\approx2800. \tag{42}$$

It is also relatively easy to calculate the third moment:

$$M_{11}^{(3)}=M_{22}^{(3)}=M_{33}^{(3)}=M^{(3)}=\pi[{}^1/_{16}A(\Delta\omega)^4-{}^3/_4AM^{(2)}-{}^3/_{16}A^2M^{(1)}-{}^1/_{64}A^3M^{(0)}]. \tag{43}$$

As regards the moments of the cross functions $\tilde{\gamma}_{23}$ and $\tilde{\gamma}_{32}$, up to and including the third moment we have values identically equal to zero.

The calculation of the higher moments becomes extremely unwieldy. We note that the calculation of the fourth moment in our case is equivalent in practice to the calculation of the sixth moment in calculations of EPR and NMR lineshapes. Moreover, even a knowledge of the fourth and higher moments does not give us the possibility of describing the behavior of the relaxation functions exactly. Nevertheless, we need to analyze the higher moments. This is connected with the following specific circumstance, inherent in Mössbauer spectroscopy.

In fact, in the investigation of spin-spin relaxation in systems of the type described, the extent to which the relaxation parameters $\gamma_1(\omega)$ and $\gamma_2(\omega)$ differ is extremely important. In the case when the difference of the functions $\gamma_1(\omega)$ and $\gamma_2(\omega)$ can be neglected, then, as can be seen from (30), the sole relaxation function $\gamma(\omega) = \gamma_1(\omega) \simeq \gamma_2(\omega)$ can be established from the experimental data for a polycrystalline sample. This, naturally, would extend the number of cases to which the theory developed above is applicable. Below, therefore, we shall make estimates of the fourth moments also. The calculation of $M_{23}^{(4)}$ and $M_{32}^{(4)}$ is performed fairly simply and leads to the result

$$M_{23}^{(4)} = M_{32}^{(4)} = {}^{27}/_{32}\sqrt{2}\, A^2 J_0^4 a^{-12} S_5, \tag{44}$$

$$S_5 = \sum_r (R_{0r})^{-12} [3(R_{0r}^x R_{0r}^y)^2 - (R_{0r}^x)^4]. \tag{45}$$

As already noted above, the exact evaluation of the fourth moment of the functions $\tilde{\gamma}_{11}$ and $\tilde{\gamma}_{22}$ is extremely complicated, but we can make an approximate estimate. For example, the fourth moment $M_{11}^{(4)}$ will be a sum of positive terms, of which one has the structure (44), (45), where the minus sign must be replaced by a plus sign in the square brackets in (45). The value of the corresponding term will then be approximately thirty times greater then the quantity $M_{23}^{(4)}$. In fact, we have here a small parameter, associated with the following physical circumstance. If we go over from cubic to spherical symmetry, i.e., replace the lattice sums that arise in the calculation of the moments by integrals over regions of spherical symmetry, the values of all the moments for the functions $\tilde{\gamma}_{32}$ and $\tilde{\gamma}_{23}$ will be rigorously equal to zero. In other words, in the lattice sum (45) the terms in the square brackets cancel each other to a considerable extent. As regards the moments of the functions from the diagonal elements, cancellations of this type is not observed in them. This situation is maintained in the calculation not only of the fourth moment but also in higher moments. Therefore, for the estimates we can assume that the off-diagonal elements, in order of magnitude, amount to

$$\gamma_{23}(\omega),\ \gamma_{32}(\omega) \approx \alpha \gamma_{11}(\omega). \tag{46}$$

where α is a small parameter defined by the relation

$$\alpha = \sum_r (R_{0r})^{-12} [3(R_{0r}^x R_{0r}^y)^2 - (R_{0r}^x)^4] \Big/ \sum_r (R_{0r})^{-12} [3(R_{0r}^x R_{0r}^y)^2 + (R_{0r}^x)^4], \tag{47}$$

and, as already noted above, for a face-centered cubic lattice $\alpha = 1/30$. We note that (46) is certainly an overestimate of the functions γ_{23}, γ_{32}. In estimating the fourth moment $M_{11}^{(4)}$ we discarded a large number of other positive terms, having a structure different from that of (44). The contribution of these discarded terms depends on the relative value of $\Delta\omega$ and A, and, for the specific substance $Cs_2NaYbCl_6$, by our estimate, $\alpha \leqslant 0.001$. Since the cross functions appear quadratically in $\gamma_2(\omega)$, we can certainly neglect this term, i.e., put

$$\gamma_2(\omega) = \gamma_{22}(\omega). \tag{48}$$

However, even when we have made the approximation (46), the functions $\gamma_1(\omega)$ and $\gamma_2(\omega)$ still differ from each other. It is not difficult to calculate the difference of their fourth moments, and we obtain the result

$$M_{11}^{(4)} - M_{22}^{(4)} = \frac{27}{32} A^2 J_0^4 a^{-12} S_5, \tag{49}$$

i.e., in the given case the difference is also small and is linear in the parameter α. In the first approximation, with good accuracy we can set the relaxation functions equal:

$$\gamma_1(\omega) = \gamma_2(\omega) \equiv \gamma(\omega). \tag{50}$$

This gives the possibility of using experimental data on polycrystalline samples. We specially emphasize that the validity of the approximation (50) can be easily checked experimentally by using monocrystalline samples.

We shall discuss briefly approximate models to describe the relaxation functions. In the relaxation superoperator $\hat{M}(\omega)$ we neglect the interaction H_{AB} in the denominator. Immediately, we easily find that $\gamma_{23} = \gamma_{32} = 0$; in addition, the condition (50) is fulfilled, and for $\gamma(\omega)$ we have the following representation:

$$\gamma(\omega) = (\Delta\omega)^2 [K(\omega - A) + K(\omega + {}^3/_2 A)], \tag{51}$$

where $K(\omega)$ is the spin autocorrelation function:

$$K(\omega) = \langle S_r^z | (\omega - \hat{L}_B + i\Gamma/2)^{-1} | S_r^z \rangle. \tag{52}$$

According to (51), the function $\gamma''(\omega)$ is a set of two peaks, concentrated about the positions of the resonance frequencies corresponding to the initial hyperfine interaction. However, even for such a substantially simpler correlator, exact methods of determining its form do not exist. Again, we can only calculate a series of its moments and then approximate by some standard function—either a Gaussian or a Lorentzian distribution, as was done in Ref. 9. Inasmuch as the moments of the function $K(\omega)$ are all finite, it appears to us that the Gaussian distribution is preferable:

$$K(\omega) = \frac{1}{\pi} \left(\frac{\sqrt{\pi}}{4\Delta\omega} \right) \int \frac{d\omega'}{\omega' + i\Gamma/2} \exp\left\{ -\left(\frac{\omega - \omega'}{\Delta\omega} \right)^2 \right\}. \tag{53}$$

The formula (53) can be used for a rough comparison of the form of the relaxation functions with the experimental data obtained, and can in no way be regarded as the result of an exact theory.

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Nonlinear electromagnetic absorption of short-wave sound in metals

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An analysis is made of the electromagnetic contribution to the nonlinear coefficient of absorption of short-wave sound in a metal, when the wavelength is much smaller than the free-path length of the electrons. It is shown that, depending on the value of the ratio of the wavelength of the sound to the skin depth (with anomalous skin effect), there are two possible types of dependence of the absorption coefficient on the sound intensity. In the high-frequency range, the deformation contribution to the absorption dominates; the absorption coefficient decreases, with increase of the sound intensity S, as $S^{-1/4}$. In the low-frequency range, the electromagnetic contribution to the nonlinear absorption coefficient dominates, although in the linear range the electromagnetic and deformational contributions are of the same order of magnitude. The sound-absorption coefficient initially increases with increase of the sound intensity as $S^{1/2}$ but then begins to decrease as $S^{1/4}$. The value of the nonlinear absorption coefficient may exceed the value of the linear coefficient. This type of dependence is due to the nonlinear character of the shielding of the priming eddy currents, which lead to the electromagnetic absorption, by conductivity currents. The estimates made show that the predicted dependences are completely accessible for experimental investigation.

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INTRODUCTION

The propagation in metals of short-wave sound, whose wavelength $2\pi/q$ is much smaller than the free-path length l of the electrons, has been repeatedly investigated both experimentally and theoretically. In an overwhelming majority of the experiments, what was studied was the propagation of sound of small intensity, so that the experimental situation was well described by a theory, developed in a number of papers, that is linear in the sound-wave amplitude. By comparison of the experimental results with the theory, the basic mechanisms responsible for sound absorption have been established. For longitudinal sound, a typical mechanism is the so-called deformation mechanism,[1] due to modulation of the energy of the electrons in the field of the sound wave. In the case of transverse sound, along with the deformational absorption, an important role is played by the so-called electromagnetic absorption,[2] caused by Joule losses during flow in the metal of eddy currents produced by the sound wave. In metals with a complicated Fermi surface, and also in propagation of mixed sound modes containing longitudinal and transverse components, the two mechanisms of absorption may compete.

Experimentalists are now able to introduce into metallic crystals sound waves of sufficiently large intensity for study fo the characteristics of their propagation in the nonlinear range. It has therefore become timely to develop a nonlinear theory of absorption of short-wave sound in the range

$$ql \gg 1. \tag{1}$$

For the deformation mechanism of absorption, such a theory has already been developed.[3-5] A qualitative picture describing nonlinear deformational absorption consists of the following. It is well known[1] that what is

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