

# Effect of resonance scattering of phonons on spin-lattice relaxation of paramagnetic centers

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A crystal is considered with two types of local centers. The rapidly relaxing local centers interact strongly with the lattice and lead to resonance scattering of phonons. The nature of the rapidly relaxing local centers is arbitrary. The problem of scattering of phonons by the rapidly relaxing local centers is solved. The results are applied to spin-lattice relaxation of paramagnetic centers that are introduced additionally into the crystal (direct interaction between the paramagnetic centers and the rapidly relaxing centers is not assumed). The results are discussed in connection with the low-temperature thermal conductivity of the lattice. Spin-lattice relaxation of the paramagnetic centers, in which account is taken of the resonance scattering of phonons, possesses characteristic singularities in the temperature dependence. It is proportional to the concentration of the rapidly relaxing local centers and may exceed the rate of the direct process by several orders of magnitude.

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## 1. INTRODUCTION

The presence in a crystal of local centers that interact strongly with the lattice leads to a change in the sound propagation velocity,<sup>[1]</sup> a decrease in the thermal conductivity,<sup>[2]</sup> and an increase in the lattice specific heat.<sup>[3]</sup> This indicates a change in the properties of the system of phonons in a lattice with local centers. Usually, spectroscopic study of local centers that interact strongly with the lattice is made difficult by their fast relaxation (in what follows, we shall call them rapidly relaxing local centers). In this case, ordinary paramagnetic centers added to a crystal with rapidly relaxing local centers can serve as a probe for the study of the properties of the rapidly relaxing local centers. Actually, the spin-lattice relaxation of the paramagnetic centers is sensitive to a change in the properties of the phonon system.

A number of authors<sup>[4-6]</sup> have already considered the problem of the indirect interaction of two paramagnetic centers through a phonon field, the so-called exchange of virtual phonons. This interaction appears in second-order perturbation theory for two paramagnetic centers of identical frequency. This interaction was expected to be manifested in a shift of the frequency of the paramagnetic centers or in a change in the relaxation of the system of paramagnetic centers. However, these effects were not observed experimentally, probably because of the difficulties that arise in distinguishing the effect of exchange of virtual phonons against the background of other interactions in the system of paramagnetic centers.<sup>[7]</sup>

Meanwhile, in the consideration of two types of local centers, the indirect interaction via the phonon field could appear as a cross-relaxation from the paramagnetic centers to the rapidly relaxing local centers. Actually, Moore and Satten<sup>[8]</sup> showed convincingly in an experiment that such a cross-relaxation exists in the absence of direct interaction between the fast and slow centers. The system of fast centers in this case was close to the conditions of a 'phonon neck.' The theory developed by Kochelaev and Aminov<sup>[5]</sup> and by Orbach and Tachiki<sup>[6]</sup> is inapplicable to such a system, since the effect of exchange of virtual phonons is absent in the case of noncoincidence of the resonance frequencies of the two types of centers. Moreover, such a theory is

inapplicable when some of the local centers are essentially fast centers.

The interpretation of the experiment of Moore and Satten<sup>[8]</sup> based on the phenomenological balance equations without account of the virtual phonon exchange between the spins of the fast centers, is incorrect.

An interesting aspect of the problem under analysis is the connection between the paraelectric and paramagnetic centers. The paraelectric centers are always rapidly relaxing local centers, and direct interaction between the nonparamagnetic and paraelectric centers is absent. One should expect an effect of the paraelectric centers on the relaxation of paramagnetic centers via the phonon system.

## 2. RELAXATION OF PARAMAGNETIC CENTERS IN A LATTICE WITH RAPIDLY RELAXING LOCAL CENTERS

The Hamiltonian of the system has the form

$$\mathcal{H} = \mathcal{H}_s + \mathcal{H}_{in}^a + \mathcal{H}_L + \mathcal{H}_{in}^b + \mathcal{H}_b, \quad (1)$$

$\mathcal{H}_a$  and  $\mathcal{H}_{int}^a$  are the energy of the paramagnetic centers and the operator of their interaction with the lattice;  $\mathcal{H}_L$  is the Hamiltonian of the lattice;  $\mathcal{H}_b$  and  $\mathcal{H}_{int}^b$  are the energy of the rapidly relaxing local centers and the operator of their interaction with the lattice.

According to the theory of kinetic equations for the system of paramagnetic centers, the usual Markoffian kinetic equation can be introduced for the system of paramagnetic centers<sup>[9]</sup> if the correlation time  $\tau_C$  in the dissipative system is smaller than the relaxation time  $\tau_a$  of the paramagnetic centers. We consider the lattice and the rapidly relaxing local centers together as the dissipative system. The state of the rapidly relaxing local centers is of no importance here except in order that there be a short correlation time in such a coupled system. At low temperatures, scattering from defects is the basic correlation mechanism in the phonon system.<sup>[3]</sup> Here we are interested in the resonant scattering of phonons by the rapidly relaxing local centers. Thus, it is required that the probability of resonance scattering of phonons  $\tau_0^{-1} \gg \tau_a^{-1}$ . Under this condition, the paramagnetic centers transfer energy to the

phonon system, which is distorted by the interaction with the rapidly relaxing local centers.

As usual, we assume that

$$\mathcal{H}_{int}^{\alpha} = \sum_i \sum_{\mu} C_{m\mu}^{(i)} V_{\mu}^{(i)} u_{m\mu}^{\mu}, \quad (2)$$

$C_{m\mu}^{(i)}$  is the constant of spin-phonon interaction of the  $i$ th paramagnetic center,  $u_{m\mu}^{\mu}$  is the displacement of the  $m$ th neighbor with the  $i$ th paramagnetic center of the lattice ion along the  $\mu$  axis and  $V_{\mu}^{(i)}$  are the spin operators. Then the probability of relaxation of the paramagnetic center from level 1 to level 2 is equal to

$$W_{12}^{(i)} = \sum_{m\mu} C_{m\mu}^{(i)2} |V_{\mu}^{(i)}|^2 \langle u_{m\mu}^{\mu}(\tau) u_{m\mu}^{\mu} \rangle_{\omega_{12}}, \quad (3)$$

$$\langle \dots \rangle_{\omega_{12}} = \frac{1}{2\pi} \int_{-\infty}^{\infty} \text{Sp} \{ e^{-\beta \mathcal{H}'} \dots \} e^{i\omega_{12}\tau} d\tau / \text{Sp} e^{-\beta \mathcal{H}'}. \quad (4)$$

In (4),

$$\mathcal{H}' = \mathcal{H}_L + \mathcal{H}_b + \mathcal{H}_{int}^{\alpha}.$$

The problem reduces to calculation of the Fourier transform of the correlation function (4) with the Hamiltonian  $\mathcal{H}'$ .

### 3. CALCULATION OF THE CORRELATOR

For simplicity, we consider rapidly relaxing local centers with effective spin  $S = 1/2$  and an isotropic  $g$  factor. This assumption simplifies the problem strongly, but it can be expected that the qualitative conclusions will remain valid at  $S > 1/2$ . The Hamiltonian of centers of nonparamagnetic nature, for example, the paraelectric centers, can also be written in the form of a spin Hamiltonian with a fictitious spin.<sup>[10]</sup> At low temperatures, the lattice can be regarded as harmonic. We direct the  $Z$  axis along the external magnetic field. Then

$$\mathcal{H}' = \sum_p \omega_p a_p^+ a_p + \sum_{j=1}^{N_b} \left\{ \omega_{0j} S_z^j + \sum_{\alpha} \sum_p B_p^{\alpha} S_{\alpha}^j (a_p - a_p^+) \exp(ipR_j) \right\}. \quad (5)$$

Here  $B_p^{\alpha}$  is the constant of interaction of the rapidly relaxing local centers with the lattice;  $\alpha = Z, +, -$ ;  $p$  numbers the wave vector  $p$  and the branch  $j_p$ ;  $R_j$  is the radius vector of the  $j$ th rapidly relaxing local center;  $\omega_p$  is the energy of the  $p$ th phonon;  $a_p^+$  and  $a_p$  are the creation and annihilation operators of the  $p$ -th phonon.

To calculate the Fourier transform of the lattice correlator in (4), we introduce the equal-time temperature Green's function  $G_{kk} = \langle \langle a_k - a_k^+; a_{-k} - a_{-k}^+ \rangle \rangle$  and, in accord with Zubarev,<sup>[11]</sup> we write the set of coupled differential equations. In order to obtain the final values of the correlator for  $B_p^{\alpha}$ , which satisfy the criterion of perturbation theory, it is sufficient to limit ourselves to eleven equations for the following Green's functions.<sup>1)</sup>

$$\langle \langle a_k \mp a_{-k}^+; a_{-k} - a_k^+ \rangle \rangle, \quad \langle \langle S_{\alpha}; a_{-k} - a_k^+ \rangle \rangle, \\ \langle \langle S_{\alpha} (a_k \mp a_{-k}^+); a_{-k} - a_k^+ \rangle \rangle; \quad \alpha = Z, +, -, \quad S_{\pm} = S_X \pm iS_Y.$$

We convert to the Fourier transforms, and after the well-known procedure of factorization of the Green's function of higher order<sup>[11, 12]</sup> and solution of the system, we find

$$G_{kk}(\omega) = -\frac{1}{\pi} \left\{ \frac{\omega^2 - \omega_k^2}{\omega_k} - 4 \sum_{j=1}^{N_b} \frac{B_k^+ B_{-k}^- \langle S_z^j \rangle}{\Delta} \right\}^{-1}; \quad (6)$$

$\Delta$  is expressed in terms of the determinant of the matrix of coefficients  $a_{\alpha\alpha'}$  of the system of third-order

linear equations for the Green's functions  $\langle \langle S_{\alpha}; a_{-k} - a_k^+ \rangle \rangle$ . For  $\omega_{0j} \gg |B_p^{\alpha}|$ , we obtain

$$\Delta = \frac{\omega - \omega_{0j}}{2\omega_{0j}} + \frac{\omega^2 - \omega_{0j}^2}{2\omega\omega_{0j}} a_{zz} + \frac{\omega - \omega_{0j}}{2\omega_{0j}} a_{++} + \frac{\omega + \omega_{0j}}{2\omega_{0j}} a_{--}; \quad (7)$$

$$a_{zz} = 2 \sum_p B_p^+ B_{-p}^- (2n_p + 1) \left\{ \frac{\omega + \omega_{0j}}{(\omega + \omega_{0j})^2 - \omega_p^2} + \frac{\omega - \omega_{0j}}{(\omega - \omega_{0j})^2 - \omega_p^2} \right\}, \\ a_{\pm\pm} = 2 \sum_p B_p^+ B_{-p}^- \frac{(2n_p + 1) \omega \mp 2 \langle S_z^j \rangle \omega_p}{\omega^2 - \omega_p^2} \\ + \sum_p B_p^z B_{-p}^z \frac{(2n_p + 1) (\omega \pm \omega_{0j}) \mp \theta_p \omega_p}{(\omega \pm \omega_{0j})^2 - \omega_p^2}. \quad (8)$$

$$\theta_p = 2 \sum_{j'=j}^{N_b} \langle S_z^{j'} \rangle \exp \{ i p (R_j - R_{j'}) \}, \quad n_p = n(\omega_p) = [\exp(\beta\omega_p) - 1]^{-1}.$$

With the help of (6)–(8), we obtain an expression for the Fourier transform of the correlator  $\mathcal{G}_{kk}(\omega)$  corresponding to the Green's function  $G_{kk}(\omega)$ :

$$\mathcal{G}_{kk}(\omega) = \frac{1}{\pi} \frac{n(\omega) \Gamma_k(\omega)}{[ (\omega^2 - \omega_k^2) / 2\omega_k + \Pi_k(\omega) ]^2 + \Gamma_k^2(\omega)}, \quad (9)$$

$$\Gamma_k(\omega) = 2 \sum_{j=1}^{N_b} B_k^+ B_{-k}^- \langle S_z^j \rangle \frac{D}{\Delta^2 + D^2}, \quad \Pi_k(\omega) = -2 \sum_{j=1}^{N_b} B_k^+ B_{-k}^- \frac{\langle S_z^j \rangle \Delta}{\Delta^2 + D^2}, \quad (10)$$

$$D = D_+ + D_-, \quad D_{\pm} = -\pi \sum_p B_p^+ B_{-p}^- \left\{ \delta(\omega \mp \omega_p) \left[ \frac{\omega}{\omega_{0j}} (2n_p + 1) \pm 2 \langle S_z^j \rangle \right] \right. \\ \left. + \frac{\omega^2 - \omega_{0j}^2}{2\omega\omega_{0j}} (2n_p + 1) [\delta(\omega \pm \omega_{0j} - \omega_p) + \delta(\omega \pm \omega_{0j} + \omega_p)] \right\} \\ - \frac{\pi}{2} \sum_p |B_p^z|^2 \frac{\omega \mp \omega_{0j}}{2\omega_{0j}} [\delta(\omega \pm \omega_{0j} - \omega_p) (2n_p + 1 \mp \theta_p) \\ + \delta(\omega \pm \omega_{0j} + \omega_p) (2n_p + 1 \pm \theta_p)]. \quad (11)$$

As was to be expected, the polarization and mass operators, which essentially determine the contribution of the phonon  $k$  to the relaxation of the paramagnetic center, depend strongly on the detuning between the frequencies of the paramagnetic centers and the rapidly relaxing local centers,  $\omega - \omega_{0j}$ , so that, for a sufficiently large detuning  $\Pi_k(\omega)$ ,  $\Gamma_k(\omega) \rightarrow 0$ , and (9) transforms into a  $\delta$  function, which leads to a direct process of relaxation. The largest value of  $\Gamma_k(\omega)$  is achieved as  $\Delta \rightarrow 0$  and, consequently,  $\Pi_k(\omega) \sim \Delta$  can be neglected in this region.

The change in the dispersion of the vibrational spectrum of the lattice, which  $\Pi_k^{(\omega)}$  describes, must be taken into account, for example, in experiments on the measurement of the sound velocity in the crystal.<sup>[1, 12, 13]</sup>  $\Gamma_k^{(\omega)}$  with  $\omega = \omega_k$  describes the probability of resonant scattering of the phonon by the rapidly relaxing local center, and is equal to the reciprocal of the lifetime of the phonon, which determines the coefficient of thermal conductivity of the lattice at low temperatures. The quantity  $\theta_p$  in (8), (11) depends on the character of the distribution of the rapidly relaxing local centers, and in the case of a random distribution,  $\theta_p \sim \delta(p)$  and we do not have to take it into account.

### 4. CALCULATION OF THE RATE OF RELAXATION OF THE PARAMAGNETIC CENTERS

To estimate the rate of relaxation (4), we put in formulas (7)–(11)

$$|B_p^x|^2 = |B_p^y|^2 = |B_p^z|^2 = \omega_p V / N_0, \quad (12)$$

where  $N_0$  is the number of elementary cells in the crystal and  $V$  is a constant that does not depend on  $p$ , has the dimensions of energy, and characterizes the

amount of interaction of the rapidly relaxing local centers with the lattice.

Substituting (9) in (4) with account of (12), we find after summation over all acoustic phonons for the probability of relaxation transition of the paramagnetic center from the upper state 1 to the lower state 2

$$W_{12} = W_{12 \text{ dir}} \left\{ 1 + \frac{2}{3\pi} \left( \frac{\omega_D}{\omega} \right)^3 \gamma \right\}. \quad (13)$$

This formula is obtained in the Debye model of the lattice under the approximation  $\omega \ll \omega_D$ ,  $\gamma \ll 1$  ( $\omega_D$  is the Debye frequency):

$$\begin{aligned} \gamma = c \int_0^{\infty} d\omega_{0j} & \left[ \frac{3\pi}{2} \left( \frac{\omega}{\omega_D} \right)^3 \left( \frac{\omega}{\omega_{0j}} \text{cth} \frac{\beta\omega}{2} + \text{th} \frac{\beta\omega_{0j}}{2} \right) \right] \\ & \times \text{th} \frac{\beta\omega_{0j}}{2} \rho(\Omega - \omega_{0j}) \left/ \left\{ \left[ \frac{\omega^2 - \omega_{0j}^2}{2V\omega_{0j}} + \text{th} \frac{\beta\omega_{0j}}{2} \right]^2 \right. \right. \\ & \left. \left. + \left[ \frac{3\pi}{2} \left( \frac{\omega}{\omega_D} \right)^3 \left( \frac{\omega}{\omega_{0j}} \text{cth} \frac{\beta\omega}{2} + \text{th} \frac{\beta\omega_{0j}}{2} \right) \right]^2 \right\} \right\}, \quad (14) \end{aligned}$$

$W_{12 \text{ dir}}$  is the rate of the direct relaxation of the paramagnetic centers in an ideal lattice, and  $\beta = \hbar/kT$ . For the probability of relaxation transition from the lower to the higher level, we must replace  $\omega$  in (13) and (14) by  $-\omega$ .  $c = N_n/N_0$  is the concentration of rapidly relaxing local centers in the crystal. The integral in (14) appears after we go from summation over all the rapidly relaxing local centers to integration over the frequencies  $\omega_{0j}$ ;  $\rho(\Omega - \omega_{0j})$  is a function of the inhomogeneous scatter of the frequencies ( $\Omega$  is the mean frequency of the rapidly relaxing local centers). It follows from (14) that the inhomogeneous broadening will play a fundamental role if the width of the inhomogeneity is

$$\sigma > 10 \left( \frac{\omega}{\omega_D} \right)^3 \frac{kT}{\hbar\omega_{0j}} V \quad (kT > \hbar\Omega).$$

In this case, the quasi-Lorentzian distribution set off in (14) by the square brackets can be replaced by a  $\delta$  function. Then

$$\gamma_{\text{inhom}} = \pi c V \int_0^{\infty} d\omega_{0j} \left( \frac{\omega^2 - \omega_{0j}^2}{2\omega_{0j}} + V \text{th} \frac{\beta\omega_{0j}}{2} \right) \text{th} \frac{\beta\omega_{0j}}{2} \rho(\Omega - \omega_{0j}) d\omega_{0j}. \quad (15)$$

Thus, the relative increase in the rate of relaxation  $\Delta W_{12}/W_{12 \text{ dir}} = (W_{12} - W_{12 \text{ dir}})/W_{12 \text{ dir}}$  can reach a value  $\sim c(\omega_D/\omega)^3 V/\sigma$ .

In the range of temperatures  $kT > \hbar\Omega$  with  $V \ll kT$ , it follows from (15) that

$$\begin{aligned} \Delta W_{12} = & \left( \frac{\omega_D}{\omega} \right)^3 c \frac{V}{\sigma} W_{12 \text{ dir}} \text{th} \left[ \beta\omega \left( 1 + \frac{V}{2kT} \right) \right] \\ & \cdot \left( 1 + \frac{3}{2} \frac{V}{kT} \right)^{-1} \left( \Omega - \omega \left( 1 + \frac{V}{2kT} \right) \right). \quad (16a) \end{aligned}$$

Here the dependence of the rate of relaxation on the concentration is linear, and  $\Delta W_{12}$  depends only weakly on the temperature. In the region  $kT \ll \hbar\Omega$ ,

$$\Delta W_{12} = \pi c V \rho(\Omega - \omega - V) (\omega_D/\omega)^3 W_{12 \text{ dir}}. \quad (16b)$$

The dependence of  $\Delta W_{12}$  on the detuning has a bell-shaped character or is more complicated, depending on the shape of the inhomogeneous line, and the maximum of  $\Delta W_{12}$  is shifted by an amount  $\sim V$  with a change in temperature.

The inhomogeneous scatter of the frequencies usually has a Gaussian shape. We assume

$$\rho(\Omega - \omega_{0j}) = \frac{1}{\sigma\sqrt{\pi}} \exp \left[ - \left( \frac{\Omega - \omega_{0j}}{\sigma} \right)^2 \right]. \quad (17)$$

Substituting (17) in (15), we calculate  $\gamma_{\text{inhom}}$  for various  $\omega$ ,  $\sigma$ , and  $V$  as a function of the temperature. It is

seen that in the general case either an increase or a decrease of  $\Delta W_{12}$  with temperature may be observed. For the most typical parameters, however, one would expect only a very weak dependence on the temperature.

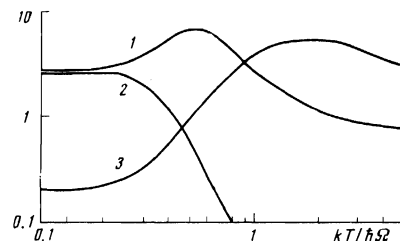
If the nonuniformity of the frequencies is unimportant in comparison with the phonon broadening of the levels of the rapidly relaxing local centers, i.e., if  $\sigma < (\sqrt{2})\pi(\omega/\omega_D)^3 kTV/\hbar\Omega$ , all the qualitative conclusions are preserved. The dependence on the detuning will be sharper. The largest attainable decrease in the relaxation in this case is equal to  $\Delta W_{12}/W_{12 \text{ dir}} \sim 10^{-2}(\omega_D/\omega)^6 c$ . At exact resonance,  $\Delta W_{12}$  does not depend on the amount of interaction of the rapidly relaxing local centers with the lattice, but the location of the exact resonance depends on  $V$ .

## 5. DISCUSSION AND COMPARISON WITH EXPERIMENT

**A. Relaxation.** The experiments of Moore and Satten<sup>[8]</sup> on the relaxation of Pr and Er in ethyl sulfate can be compared directly with the theory that has been presented, since the rates of spin-lattice relaxation amount to  $\tau_b^{-1} \sim 10^4$  Hz and  $\tau_a^{-1} \sim 10^2$  Hz, respectively. The rate of relaxation of Er in such a mixed system is determined by the quantity  $r = \tau_a^{-1} + r_{ab}$ , where  $r_{ab}$  is the rate of cross-relaxation between the centers  $a$  and  $b$  via phonons. In<sup>[8]</sup>, cross-relaxation through dipole-dipole interaction was excluded by the fact that, for Pr,  $g_{\perp} = 0$  (anisotropic  $g$  factor). The rate  $r_{ab}$  is proportional to the concentration of fast centers, as follows from the experiment and from our theory. The temperature dependence of  $r_{ab}$ , according to Eq. (17) of<sup>[8]</sup>, should be absent, while experiment demonstrates a significant increase in  $r_{ab}$  with increasing  $T$ . The theory used by Moore and Satten is inapplicable to the case of Pr and Er in ethyl sulfate, as we have already noted in the Introduction.

In the figure, curve 3 corresponds approximately to the considered case (the interval 1.1–2.2°K in units of  $\hbar\omega$  (Pr) amounts to 0.6 to 1.2) and describes qualitatively the relaxation of Er in ethyl sulfate with Pr. Quantitatively, Eq. (16b) gives the correct order of decrease of the rate of relaxation of Er—a factor of 10–10<sup>2</sup>.

The characteristic feature of the mechanism of decrease in the relaxation time that we have considered is the inverse proportionality of  $W_{12}$  to the temperature. At low temperatures, so far as is known, this is the only mechanism with such a temperature dependence. It is interesting to observe that such a decrease in the rate of relaxation with increase in temperature was observed in the work of Deigen et al.<sup>[14]</sup> in the range



Temperature dependences of  $\gamma'/c$  calculated from Eqs. (15), (17) for the following values of  $V$ ,  $\sigma$ ,  $\omega$  (in units of  $\hbar\Omega$ ): curve 1— $V = 0.2$ ,  $\sigma = 0.5$ ,  $\omega = 0.5$ ; 2— $V = 0.2$ ,  $\sigma = 0.1$ ,  $\omega = 0.7$ ; 3— $V = 0.2$ ,  $\sigma = 0.1$ ,  $\omega = 0.95$ .  $\gamma' = \coth(\beta\omega/2)\gamma$ .

16–20°K. Equation (15) describes quantitatively the decrease in the time of the  $F_A(Li^+)$  centers at  $c = 2 \times 10^{-7}$  (which corresponds to a random distribution of  $Li^+$  in KCl with a concentration of  $n \sim 10^{18} \text{ cm}^{-3}$  and of F centers with a concentration of  $4 \times 10^{-17}$ ), if we assume that  $V \lesssim \Omega$ ,  $\sigma \approx 3 \times 10^{11} \text{ Hz}$ . Here the quantity  $\Omega \approx 6 \times 10^{11} \text{ Hz}$  characterizes the tunnel splitting of the noncentral  $F_A(Li^+)$  in KCl.<sup>[14]</sup>

**B. Thermal conductivity.** The interaction of the rapidly relaxing local centers with the lattice appears in experiments on the thermal conductivity of crystals.<sup>[2,15,16]</sup> The value of the thermal conductivity depends on  $\Gamma_k(\omega_k)$ . In the case  $\sigma \gg V$ , in accord with (10), (11),

$$\Gamma_k(\omega_k) = V \omega_k c \rho (\omega_k - \Omega) \text{th}(\beta \omega_k / 2). \quad (18)$$

This expression is identical with the phenomenological formula of Orbach,<sup>[17]</sup> with the help of which he succeeded in describing the experiment of Morton and Rosenberg<sup>[2]</sup> on paramagnetic salts, where the thermal resistance was measured as a function of the magnetic field.

In the case  $\sigma < V$ , Eq. (18) for  $\Gamma_k(\omega_k)$  leads to an expression for the thermal resistance

$$\lambda^{-1} = \frac{32\pi^2 v V^2}{\omega_v^2 k^2 T J_0} c \frac{(\Omega/kT) \text{th}(\Omega/2kT)}{[(\Omega/kT)^2 - J_1/2J_0]^2 + [J_2/J_0 - (J_1/2J_0)^2]^2}. \quad (19)$$

Here

$$J_0 = \int_0^{\theta/T} \text{th} \frac{x}{2} \frac{e^x dx}{(e^x - 1)^2 x}, \quad J_1 = 2 \int_0^{\theta/T} \text{th} \frac{x}{2} \frac{e^x dx}{(e^x - 1)^2},$$

$$J_2 = \int_0^{\theta/T} \text{th} \frac{x}{2} \frac{e^{2x} dx}{(e^x - 1)^2}.$$

The function  $\lambda^{-1}$  is also bell-shaped in the magnetic field, but its width depends strongly on the temperature. The divergence in  $J_0$  at  $x \ll 1$  is unimportant, since in this region it is necessary to take into account the scattering of phonons on the boundaries of the crystal, similar to what is done in<sup>[18]</sup>.

## 6. CONCLUSIONS

In the experiments on relaxation in a crystal with impurities of two types, an effective cross-relaxation exists between them even in the absence of direct interaction. This is connected with the fact that the local centers, which interact strongly with the lattice, shorten the lifetime of the phonons and thus open a channel for transfer of energy from the paramagnetic center to phonons not only of resonant frequency but also of higher frequencies. This effect of the rapidly relaxing local centers on the system of phonons also appears in the thermal conductivity and in the change of dispersion of the phonons.

The temperature dependence of the rate of relaxation of the paramagnetic centers, with account of the reso-

nance scattering of the phonons by the rapidly relaxing local centers, has a number of characteristic features. This function is represented in the figure for a number of parameters. The value of the rate of relaxation can exceed the probability of the direct process by several orders of magnitude.

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<sup>1)</sup>For arbitrary spin, we would be obliged to solve  $3(2S+1)^2 - 1$  equations.

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