Propagation of nonequilibrium phonons in crystals with two-level systems

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A theory is derived for the propagation of heat pulses in crystals which contain two-level centers with an energy gap Δ under the condition $\Delta \simeq k_B T^*$, where $k_B T^*$ is the average energy of the phonons in the heat pulse. In addition to the elastic scattering due to point defects, there is a strong Raman scattering of phonons in this case, with a pumping of two-level centers. This interaction causes a redistribution of energy between nonequilibrium phonons and two-level centers. It slows down the energy transfer in the crystal substantially. An analytic expression is derived for the response of a bolometer. A family of curves is plotted to show possible shapes of the temperature dependence of the time at which the signal maximum arrives. The nature of these curves is determined by the relation between the length of the crystal and the average distance over which a phonon diffuses before a Raman-scattering event occurs.

Nonequilibrium phonons injected into a crystal at low temperatures can undergo a variety of interactions. The nature and intensity of these interactions determine the nature of the signal recorded by a detector. The propagation of heat pulses in crystals under conditions such that the scattering of the nonequilibrium phonons is important (in contrast with ballistic propagation of these phonons) has been studied most thoroughly for the case of a defective anharmonic crystal. In such a crystal, defects cause an intense elastic scattering of phonons. Anharmonic processes, which are slower at low temperatures, are responsible for changes in the spectrum of nonequilibrium phonons as they propagate. Various regimes for the propagation of these phonons may be realized, depending on the relation between the intensities of elastic and anharmonic scattering.[1-4]

If a crystal contains two-level systems with an energy $\Delta \simeq \hbar\bar{\omega}$, where $\bar{\omega}$ is a characteristic frequency of the injected nonequilibrium phonons, the interaction with two-level systems may turn out to be the most intense interaction for resonant phonons ($\hbar\omega = \Delta$). To describe transport processes under these conditions, it is convenient to distinguish three subsystems: the resonant phonons, the two-level systems (spins), and the nonresonant phonons. We take account of all interaction processes which couple these subsystems.

The interaction between nonresonant and resonant phonons either is due exclusively to anharmonic effects or is manifested indirectly, through the spins. The interaction between the nonresonant phonons and the spins is in turn due to processes involving several phonons. These processes correspond to a perturbation theory of higher order than that in which we find the one-phonon processes in which resonant phonons interact with spins. Apparently the most important of these processes are the Raman scattering of phonons by spins[5] and three-phonon spin-lattice interactions.[6] An equilibrium in the system consisting of the resonant phonons plus the spins is thus reached far more rapidly than a complete equilibrium is reached throughout the system.

In order to describe time-varying transport processes in this system, it is important to know the time scales involved. If we are discussing the evolution of a heat pulse over a time interval no longer than the time scales for the relaxation to a complete equilibrium in the system, then we can discuss transport processes independently in the system of nonresonant phonons and in the system of resonant phonons plus the spins. Under these conditions, the two-level systems may have a negligible effect on the propagation of nonequilibrium phonons, because most of the energy of the heat pulse is carried by nonresonant phonons. The integral response of the bolometer will be essentially independent of whether the crystal contains two-level systems. The occupation numbers of the two-level systems, on the other hand, become functions of the coordinates and the time because of their interaction with resonant phonons. If the intensity of radiative transitions between corresponding states is sufficiently high, one can observe a luminescence induced by the heat pulse from any point in the crystal. One can also study the transport of electronic excitation through the crystal and the characteristics of the resonant phonons.[7-11]

We will not be discussing that case here. Our purpose in this paper is to analyze instead the case in which energy is redistributed among the various subsystems as the nonequilibrium phonon propagates. As we will see below, such a situation is completely feasible experimentally. We would expect that a large fraction of the energy of the nonresonant nonequilibrium phonons might be transferred under these conditions and would accumulate in the subsystem consisting of the resonant phonons plus the spins; such events would have some important observable consequences.

We consider a crystal containing N^* two-level systems with a spectrum $g(\varepsilon)$ ($g(\varepsilon) \neq 0$ at $\varepsilon \simeq \Delta$). We assume that the crystal has a fairly large number of defects; i.e., we assume that there is a strong elastic scattering of phonons in the crystal. For simplicity we restrict the analysis to the case in which the propagation of the nonequilibrium phonons can be described in a one-dimensional problem. Assuming that the temperature of the heat reservoir is low, $T_0 < 4.2$ K, and assuming that the heating of the generator is slight, $\delta T/T \ll 1$, we completely ignore effects associated with the lattice anharmonicity. For typical insulators at $T < 4.2$ K, the time scales for the coalescence and decay of phonons through lattice anharmonicity are on the order of 10^{-3} s. We thus assume from the outset that the basic features which stem from the specific nature of the interaction in this case are manifested far earlier.

In the absence of two-level systems, with a strong

phonon scattering by defects, and without an anharmonic scattering of phonons, the motion of the nonequilibrium phonons in the crystal can be described by a one-dimensional diffusion equation for each group of phonons with a fixed energy $\hbar\omega$ and a diffusion coefficient $D(\omega) = (1/3)V_s^2\tau_0(\omega)$, where $\tau_0(\omega)$ is the time scale of the elastic relaxation of phonons with defects, and V_s is the sound velocity.

We now consider the propagation of nonresonant nonequilibrium phonons in a crystal which does have two-level systems. We denote by $\tau_R(\omega)$ the lifetime of a phonon with an energy $\hbar\omega$ with respect to Raman scattering accompanied by the excitation or deexcitation of the two-level system. By virtue of the discussion above, we assume $\tau_R \gg \max\{\tilde{\tau}_0, \tilde{\tau}_{fs}\}$, where $\tilde{\tau}_{fs}$ is the lifetime of the resonant phonons with respect to capture by a two-level system. Over a time $\tau_R \gg \tau_0$, the nonresonant phonons undergo many elastic collisions, and they travel a distance on the order $l_R(\omega) \simeq (D(\omega)\tau_R(\omega))^{1/2}$. In each event of Raman scattering of phonons by a two-level system, the energies of the phonons before and after the collision differ by $\pm\Delta$. On the average, therefore, each phonon of the group of nonresonant nonequilibrium phonons gives up its energy to the spins over a time $\tau_R(\omega)$, having diffused a distance $l_R(\omega)$ over this time. We thus have a Raman pumping of the spin subsystem by the nonequilibrium phonons. Just how important this process is depends on the relation between $l_R(\omega)$ and the sample thickness L. If $l_R(\omega)/L < 1$, there is a high probability that each phonon will give up an energy Δ to spins, on the average, as it diffuses through the crystal. Since we have $\Delta \simeq \hbar\omega \simeq k_B T^* \simeq k_B T_0$ by assumption, the energy accumulated in the system of spins is on the same order of magnitude as the energy of the injected nonequilibrium phonons. The condition $l_R(\omega)/L < 1$ can be rewritten as $\tau_R(\omega) < L^2/D(\omega)$. For phonons with a frequency $\bar{\omega}$, this condition means $\tau_R(\bar{\omega}) < L^2/D(\bar{\omega}) \simeq t_1$, where t_1 is the arrival time of the maximum of the deviation of the phonons from equilibrium in the scattering of phonons by the defects.

If $\tau_R(\bar{\omega}) \gg t_1$, the Raman pumping cannot cause any significant redistribution of heat-pulse energy between the subsystem of the nonresonant phonons and the subsystem consisting of the resonant phonons plus the spins. The Raman pumping can be quite pronounced even under the condition $\tau_R(\bar{\omega})/t_1 > 1$. The reason is that the decay of the number of nonequilibrium phonons at $t > t_1$ is known to be very slow, $N(t) \propto t^{-1/2}$, in the case of one-dimensional diffusion. For this reason, even at $t > t_1$ the crystal still has a substantial fraction of the nonequilibrium phonons. There is a high probability that these phonons can transfer their energies to spins in Raman-scattering events.

Previous discussions of the possibility of an accumulation of energy comparable to the energy of the heat pulse in the system consisting of the resonant phonons plus the spins have implicitly assumed that the heat capacity of the spin subsystem is greater than that of the lattice. At temperatures $T < 4.2$ K this condition holds for typical insulators if the concentration N^* of two-level systems with $\Delta \simeq k_B T_0$ is at least 10^{17} cm^{-3}. Below we assume that this condition holds.

In addition to the Raman pumping of the spin subsystem, we need to consider spin-lattice relaxation. Here we will consider only two channels for this relaxation: direct and Raman processes (although we could also take into account

Orbach relaxation, for example). Apparently the most probable situation is that in which direct processes are predominant, i.e., the situation $\tau_{sd} \ll \tau_{sR}$, where τ_{sd} and τ_{sR} are the spin-lattice relaxation times for direct and Raman processes, respectively. The relation between the times $\tilde{\tau}_{fs}$ and τ_{sd} and that between τ_R and τ_{sR} depend on the spin concentration N^*. If $\tilde{\tau}_{fs}/\tau_{sd} \ll 1$, a capture effect occurs for the resonant phonons. Under these conditions, with Raman pumping of the spin subsystem, the energy of the heat pulse accumulates in the spin subsystem, and the resonant phonons are responsible for the transport of electronic excitation through the crystal over time intervals no greater than τ_{sR}. In the case $\tilde{\tau}_{fs}/\tau_{sd} \gg 1$, the spin system is an intermediate participant. First, the nonresonant nonequilibrium phonons transfer energy to the spin system in the course of the Raman scattering. The spins quickly relax, emitting resonant phonons. As a result, the energy and number of resonant phonons increase sharply. If the interaction of the resonant phonons with the spins is very intense, the diffusion coefficient for the resonant phonons and thus the velocity at which an electronic excitation moves through the crystal are low. For this reason, Raman relaxation processes must be taken into account. An alternative approach here might be to consider spectral diffusion, as a result of which the spatial diffusion would be due to phonons in the wings of the corresponding line.[8,9]

An analysis of the propagation of nonequilibrium phonons under these conditions should obviously be based on the solution of a system of three coupled kinetic equations for the distribution functions of the nonresonant nonequilibrium phonons, the resonant nonequilibrium phonons, and the spins.

Let us assume that the sample is plate-shaped with Z axis along the normal to the surface of the plate. We denote by $N(k,j;z,t)$, $\tilde{N}(k,j;z,t)$ and $\delta f_\Lambda(z,t)$ the nonequilibrium increments in the distribution functions of the nonresonant phonons, the resonant phonons, and the spins, respectively. Here k is the phonon quasimomentum and j is the index of the phonon branch. The quantity $\delta f_\Lambda(z,t)$ describes the probability for a spin to be in an excited state. The system of kinetic equations for $N(k,j;z,t)$, $\tilde{N}(k,j;z,t)$, and $\delta f_\Lambda(z,t)$ is

$$\frac{\partial N}{\partial t} + V_s\xi\frac{\partial N}{\partial z} + I_{fd}\{N\} + I_{fs}\{N,\delta f_\Lambda\} = a\delta(z)\delta(t),$$

$$\frac{\partial \tilde{N}}{\partial t} + V_s\xi\frac{\partial \tilde{N}}{\partial z} + I_{fd}\{\tilde{N}\} + I_{fs}\{\tilde{N},\delta f_\Lambda\} = \tilde{a}\delta(z)\delta(t), \qquad (1)$$

$$\frac{\partial f_\Lambda}{\partial t} + I_{sf}\{\delta f_\Lambda, N, \tilde{N}\} = 0.$$

Here $I_{fd}\{N\}$ and $I_{fd}\{\tilde{N}\}$ are the integrals for the collisions of phonons with defects; $I_{fs}\{N,\delta f_\Lambda\}$ is the integral for the collisions of nonresonant phonons with spins; $I_{sf}\{\delta f_\Lambda,N,\tilde{N}\}$ is the integral for spin-phonon collisions; ξ is the cosine of the angle between the phonon wave vector k and the Z axis; the quantity $a(\omega)$ describes the spectrum of the phonons which are generated; and $\tilde{a} \equiv a(\omega = \Delta/\hbar)$. In the one-dimensional problem, the phonon distribution function depends on ξ alone. For simplicity we have omitted from Eqs. (1) the index j, which specifies the phonon branch. After we linearize the collision integrals $I_{fs}\{N,\delta f_\Lambda\}$, $I_{fs}\{\tilde{N},\delta f_\Lambda\}$, and $I_{sf}\{\delta f_\Lambda,N,\tilde{N}\} = I_{sf}^d\{\delta f_\Lambda,\tilde{N}\} + I_{sf}^R\{\delta f_\Lambda,N\}$, where $I_{sf}^d\{\delta f_\Lambda,N\}$ and $I_{sf}^R\{\delta f_\Lambda,N\}$ describe, respectively, direct and

Raman spin-lattice relaxation, and after we carry out several simple manipulations, we can rewrite kinetic equations (1) as follows:

$$\frac{\partial N}{\partial t} + V_s \xi \frac{\partial N}{\partial z} + \frac{N}{\tau}$$

$$= \frac{F}{\tau} + \frac{1}{\tau_R'}\left(\frac{n}{f_\vee}\right)^2 \exp\{\beta(\hbar\omega+\Delta)\}\,\delta f_\wedge$$

$$+ AF_< + BF_> + a\delta(z)\delta(t),$$

$$\frac{\partial \tilde{N}}{\partial t} + V_s \xi \frac{\partial \tilde{N}}{\partial z} + \frac{\tilde{N}}{\tilde{\tau}} = \frac{\tilde{F}}{\tilde{\tau}_0} + \frac{\delta f_\wedge}{\tilde{\tau}_{fs}}\,\mathrm{cth}^2\left(\frac{\beta\Delta}{2}\right) + \tilde{a}\delta(z)\delta(t),$$

$$\frac{\partial \delta f_\wedge}{\partial t} + \frac{\partial f_\wedge}{\tau_s} = \frac{\tilde{F}}{\tau_{sd}}\,\mathrm{th}^2\left(\frac{\beta\Delta}{2}\right) + \frac{1}{N^\bullet}\sum_k \frac{F}{\tau_R'}. \tag{2}$$

Here

$$\frac{1}{\tau} = \frac{1}{\tau_R} + \frac{1}{\tau_0},\quad \frac{1}{\tau_s} = \frac{1}{\tau_{sd}} + \frac{1}{\tau_{sR}},\quad F_< \equiv F(\hbar\omega-\Delta)$$

$$F_> \equiv F(\hbar\omega+\Delta),\quad \frac{1}{\tilde{\tau}} = \frac{1}{\tilde{\tau}_0} + \frac{1}{\tilde{\tau}_{fs}},\quad \tilde{\tau}_0 \equiv \tau_0\left(\omega=\frac{\Delta}{\hbar}\right),$$

$$\tilde{\tau}_{fs} \equiv \tau_{fs}\left(\omega=\frac{\Delta}{\hbar}\right),\quad \beta = \frac{1}{k_B T}.$$

The quantities τ_R and τ_R' are introduced by means of

$$\frac{1}{\tau_R} = \frac{1}{\tau_{R1}} + \frac{1}{\tau_{R2}},\quad \frac{1}{\tau_R'} = \frac{1}{\tau_{R1}} - \frac{1}{\tau_{R2}},$$

$$\frac{1}{\tau_{R1}} = \frac{\pi}{2}\frac{N^*}{N_0}a_0^3\left(\frac{\Lambda}{mV_s^2}\right)^2 f_\wedge \frac{n(\omega-\Delta/\hbar)}{n(\omega)}$$

$$\times \omega(\omega-\Delta/\hbar)\rho(\omega-\Delta/\hbar),$$

$$\frac{1}{\tau_{R2}} = \frac{\pi}{2}\frac{N^*}{N_0}a_0^3\left(\frac{\Lambda}{mV_s^2}\right)^2 f_\vee \frac{n(\omega+\Delta/\hbar)}{n(\omega)}$$

$$\times \omega(\omega+\Delta/\hbar)\rho(\omega+\Delta/\hbar), \tag{3}$$

Here a_0^3 is the volume of the unit cell;

$$A \equiv \frac{1}{\tau_{R1}}e^{\beta\Delta}\left(\frac{n(\omega)}{n(\omega-\Delta/\hbar)}\right)^2,$$

$$B \equiv \frac{1}{\tau_{R2}}e^{-\beta\Delta}\left(\frac{n(\omega)}{n(\omega+\Delta/\hbar)}\right)^2,$$

$f_\wedge$, $f_\vee$, and n are the equilibrium occupation numbers of the spins in the excited and ground states and of phonons; Λ is the strain-energy constant for the spin-lattice interaction; M is the mass of the two-level center; and N_0 is the number of unit cells in the lattice. The times τ_{R1} and T_{R2} describe the Raman scattering of phonons by spins accompanied by excitation and deexcitation, respectively.

Solving the last of Eqs. (2), we find

$$\delta f_\wedge(z,t) = \int_{-\infty}^{t} dt'\exp\{-(t-t')/\tau_s\}\left\{\mathrm{th}^2\frac{\beta\Delta}{2}\frac{\tilde{F}(z,t')}{\tau_{sd}}\right.$$

$$\left. + \int_0^\infty d\omega\,\rho(\omega)\frac{F(z,t')}{\tau_R'(\omega)}\right\}. \tag{4}$$

The first two equations of system (2) can easily be recast as integral equations with the help of the Green's function $G(z,t) = \theta(t)e^{-t/\tau}\delta(z-V_s\xi t)$ for the equation

$$\frac{\partial G}{\partial t} + V_s\xi\frac{\partial G}{\partial z} + \frac{G}{\tau} = \delta(z)\delta(t).$$

The kernels of the integral equations contain exponential functions of the type $e^{-(t-t')/\tau}$, so the integration is dominated by the region determined by the condition $(t-t')/\tau \approx 1$. For this reason, we expand the functions of the variable $z - V_s\xi(t-t')$ in powers of the parameter $V_s\xi(t-t')/z \approx V_s\tau/z \ll 1$. Since we will be needing only phonon distribution functions averaged over angle, $F(z,t) \equiv \langle N(z,t)\rangle$ and $\tilde{F}(z,t)$, we give the resulting expressions for the Fourier transforms of these functions, $F(q,\Omega)$ and $\tilde{F}(q,\Omega)$:

$$F(q,\Omega) = \frac{1}{1-\tau/\tau_0 L(q,\Omega)}\left\{\Psi(q,\Omega) + \frac{\tau}{\tau_R'}\left(\frac{n}{f_\wedge}\right)^2\right.$$

$$\times \exp\{\beta(\hbar\omega+\Delta)\}\,L(q,\Omega)\,\delta f_\wedge(q,\Omega)$$

$$\left. + \tau L(q,\Omega)[AF_<(q,\Omega) + BF_>(q,\Omega)]\right\}. \tag{5}$$

Here

$$\Psi(q,\Omega) \approx a\tau L(q,\Omega),$$

and

$$L(q,\Omega) = \frac{1}{1+i\Omega\tau}\left[1 - \frac{q^2 D\tau}{(1+i\Omega\tau)^2} + \frac{9}{5}\frac{q^4 D^2\tau^2}{(1+i\Omega\tau)^4}\right],$$

$$\tilde{F}(q,\Omega) = \frac{\tilde{\Psi}(q,\Omega) + \tilde{L}(q,\Omega)\tau\Phi(q,\Omega)}{1-(\tilde{\tau}/\tilde{\tau}_0)\tilde{L}(q,\Omega) - (\tilde{\tau}/\tilde{\tau}_{fs}\tau_{sd})(\Omega_s+i\Omega)^{-1}},$$

$$\Phi(q,\Omega) = \frac{1}{\tilde{\tau}_{fs}}\mathrm{cth}^2\left(\frac{\beta\Delta}{2}\right)\frac{1}{\Omega_s+i\Omega}\int_0^\infty d\omega\,\rho(\omega)\frac{F(q,\Omega)}{N^\bullet\tau_R'(\omega)}. \tag{6}$$

where $\Omega_s \equiv 1/\tau_s$. Here, as above, all quantities with a tilde are found from the corresponding quantities without the tilde at $\omega = \Delta/\hbar$. Finally, taking the Fourier transform of (4), we find

$$\delta f_\wedge(q,\Omega) = \frac{1}{\Omega_s+i\Omega}\left[\frac{1}{\tau_{sd}}\mathrm{th}^2\left(\frac{\beta\Delta}{2}\right)\tilde{F}(q,\Omega)\right.$$

$$\left. + \int_0^\infty d\omega\,\rho(\omega)\frac{F(q,\Omega)}{N^\bullet\tau_R'(\omega)}\right]. \tag{7}$$

Instead of kinetic equations (2) we thus now have a new system of equations for the Fourier transforms of the angle-

averaged distribution functions (5) and (6), of the nonresonant and resonant nonequilibrium phonons, and (7) of spins. This system of equations is not closed, since a term containing distribution functions of phonons with an energy differing by $\pm \Delta$ from the energy of the phonons of the group under consideration has appeared on the right side of (5). Let us analyze the role played by these terms. We introduce

$$X(q,\Omega) = \int_0^\infty d\omega\, \rho(\omega)\, \frac{F(q,\Omega)}{N^*\tau_R'(\omega)}, \quad X_0(q,\Omega)$$

$$= \int_0^\infty d\omega\, \rho(\omega)\, \frac{\Psi(q,\Omega)}{N^*\tau_R'(\omega)(1-(\tau/\tau_0)L(q,\Omega))},$$

$$Y(q,\Omega) = \int_0^\infty d\omega\rho(\omega)\left(\frac{n}{f_\vee}\right)^2 \exp\{\beta(\hbar\omega+\Delta)\}\,\frac{\lambda}{N^*\tau_R'(\omega)},$$

$$X_{>,<}(q,\Omega) = \int_0^\infty d\omega\, \rho(\omega)\,[AF_<(q,\Omega)+BF_>(q,\Omega)]\frac{\lambda}{N^*},$$

where

$$\lambda \equiv \frac{\tau}{\tau_R'}\,\frac{L(q,\Omega)}{1-(\tau/\tau_0)L(q,\Omega)} \approx \frac{\tau_R}{\tau_R'}\,\frac{1}{1+q^2D\tau_R+i\Omega\tau_R}.$$

From (5) we easily find the equation

$$X(q,\Omega) = X_0(q,\Omega) + Y(q,\Omega)\delta f_\wedge(q,\Omega) + X_{>,<}(q,\Omega). \tag{8}$$

Using (6) and (7), we find from (8)

$$X(q,\Omega) = \left\{1 - \frac{Y(q,\Omega)}{\Omega_s+i\Omega}\left[1+\frac{\tilde{\tau}}{\tilde{\tau}_{fs}\tau_{sd}}\frac{1}{\Omega_s+i\Omega}\right.\right.$$

$$\times\left.\left.\frac{\tilde{L}(q,\Omega)}{1-(\tilde{\tau}/\tilde{\tau}_0)\tilde{L}(q,\Omega)-(\tilde{\tau}/\tilde{\tau}_{fs}\tau_{sd})(\tilde{L}(q,\Omega)/(\Omega_s+i\Omega))}\right]\right\}^{-1}$$

$$\times\left\{X_0(q,\Omega)+\frac{Y(q,\Omega)}{\Omega_s+i\Omega}\frac{1}{\tau_{sd}}\mathrm{th}^2\left(\frac{\beta\Delta}{2}\right)\right.$$

$$\times\frac{\tilde{\Psi}(q,\Omega)}{1-(\tilde{\tau}/\tilde{\tau}_0)\tilde{L}(q,\Omega)-(\tilde{\tau}/\tilde{\tau}_{fs}\tau_{sd})(\tilde{L}(q,\Omega)/(\Omega_s+i\Omega))}$$

$$\left.+X_{>,<}(q,\Omega)\right\}. \tag{9}$$

Using the explicit expressions for A and B, we find the following expressions for $X_{>,<}(q,\Omega)$:

$$X_{>,<}(q,\Omega) = \frac{1}{N^*}\int_0^\infty d\omega\,\rho(\omega)\left(\lambda_>\frac{\tau_R'}{\tau_{R2}}+\lambda_<\frac{\tau_R'}{\tau_{R1}}\right)\frac{F(q,\Omega)}{\tau_R'}. \tag{10}$$

The symbols $>$ and $<$ mean that the arguments of the corresponding function are $\hbar\omega-\Delta$ and $\hbar\omega+\Delta$, respectively. If $\hbar\omega<\Delta$, all quantities with the $<$ should be set equal to zero. In deriving (10) we used Eqs. (3) and some relations which follow from them when the variable ω is replaced by $\omega\pm\Delta/\hbar$. Using (7)–(9), we find

$$\delta f_\wedge(q,\Omega)[1-\Lambda(q,\Omega)Y(q,\Omega)] = \overline{\delta f}_\wedge(q,\Omega)+\Lambda(q,\Omega)X_0(q,\Omega)$$

$$+\Lambda(q,\Omega)\frac{1}{N^*}\int_0^\infty d\omega\,\rho(\omega)\left(\lambda_>\frac{\tau_R'}{\tau_{R2}}+\lambda_<\frac{\tau_R'}{\tau_{R1}}\right)\frac{F(q,\Omega)}{\tau_R'}, \tag{11}$$

where

$$\Lambda(q,\Omega) \equiv \frac{1}{\Omega_s+i\Omega}\left[1+\frac{\tilde{\tau}\tilde{L}(q,\Omega)}{\tilde{\tau}_{fs}\tau_{sd}}\right.$$

$$\times\left.\frac{1}{(\Omega_s+i\Omega)[1-(\tilde{\tau}/\tilde{\tau}_0)\tilde{L}-\tilde{\tau}\tilde{L}/\tilde{\tau}_{fs}\tau_{sd}(\Omega_s+i\Omega)]}\right],$$

$$\overline{\delta f}_\wedge(q,\Omega) = \frac{\mathrm{th}^2(\beta\Delta/2)\,\tilde{\Psi}}{\tau_{sd}(\Omega_s+i\Omega)[1-(\tilde{\tau}/\tilde{\tau}_0)\tilde{L}-\tilde{\tau}\tilde{L}/\tilde{\tau}_{fs}\tau_{sd}(\Omega_s+i\Omega)]}.$$

Now using expressions (5) for $F(q,\Omega)$, and carrying out several simple manipulations, we find

$$\delta f_\wedge(q,\Omega)\left\{1-\Lambda(q,\Omega)\int_0^\infty d\omega\rho(\omega)\left(\frac{n}{f_\vee}\right)^2\exp\{\beta(\hbar\omega+\Delta)\}.\right.$$

$$\left.\times\frac{\lambda}{N^*}\left[\frac{1}{\tau_R'}+\frac{\lambda_<}{\tau_{R1}}+\frac{\lambda_>}{\tau_{R2}}\right]\right\}$$

$$= \overline{\delta f}_\wedge(q,\Omega)+\Lambda(q,\Omega)\int_0^\infty d\omega\rho(\omega)\frac{1}{N^*(1-(\tau/\tau_0)L)}$$

$$\times\frac{\Psi}{\tau_R'}\left(1+\frac{\bar{\lambda}}{\lambda}\right)$$

$$+\Lambda(q,\Omega)\int_0^\infty d\omega\rho(\omega)\frac{F}{N^*}\left(\frac{\bar{\lambda}_<}{\tau_{R1}}+\frac{\lambda_>}{\tau_{R2}}\right), \tag{12}$$

where

$$\bar{\lambda} \equiv \lambda\tau_R'\left(\frac{\lambda_<}{\tau_{R1}}+\frac{\lambda_>}{\tau_{R2}}\right).$$

We can also carry out further iterations. Using (5), we clearly see the structure of the solution. An effective relaxation time $1/\bar{\tau}_R = 1/(\tau_R')+(\lambda_</\tau_{R1})+(\lambda_>/\tau_{R2})+(\bar{\lambda}_</\tau_{R1})+(\bar{\lambda}_>/\tau_{R2})+...$ is singled out in the integrand on the left side of (12). An effective relaxation time $1/\tau_R = 1/\tau_R'(1+\bar{\lambda}/\lambda+...)$ is also singled out on the right side. Clearly, allowance for all these contributions is a correct way to deal with the incoming terms in the collision integral in the case of Raman scattering of phonons by spins. The times $\bar{\tau}_R$ and $\bar{\bar{\tau}}_R$ introduced in this manner are generally functions of q and Ω. This point is not surprising, since the occupation numbers for the nonequilibrium phonons depend on z and t. Since the relaxation times of the phonons depend strongly on their energy, the times $\bar{\tau}_R$ and $\bar{\bar{\tau}}_R$ may nevertheless depend weakly on q and Ω. The reason is the significant difference between the length scales of the Raman interactions for phonons with energies $\hbar\omega$ and $\hbar\omega\pm\Delta$. For phonons with an energy $\hbar\omega-\Delta$, for example, the parameter $(q^2D\tau_R)_<$ may be large in comparison with unity, while for phonons with an energy $\hbar\omega+\Delta$ it may be small in comparison with unity. Whatever the case, the iterative procedure converges well, as can be seen from (12). The incoming terms give rise to a small change in one of the poles in the function $\delta f_\wedge(q,\Omega)$ [and also in the functions $F(q,\Omega)$ and $\bar{F}(q,\Omega)$]. This pole describes the behavior of the heat pulse after a long time (more on this below). Accordingly, we will simplify the calculations by completely discarding the contribution of the incoming terms. System of equations (5)–(7) then becomes closed.

Discarding the corresponding terms from (12), we find

$$\delta f_\wedge(q,\Omega) = \left\{ \Omega_{sR} + i\Omega\left(1 + \frac{\tilde{\tau}}{\tau_{sd}}\right) + q^2 \tilde{D}\frac{\tilde{\tau}}{\tau_{sd}} - \right.$$

$$\left. - \int_0^\infty d\omega \rho(\omega)\frac{\lambda(q,\Omega)}{N^*\tau_R'}\left(\frac{n}{f_\smile}\right)^2 \exp\{\beta(\hbar\omega+\Delta)\}\right\}^{-1}$$

$$\times \left\{ \frac{\widetilde{\Psi}}{\tau_{sd}}\frac{\text{th}^2(\beta\Delta/2)}{1-(\tilde{\tau}/\tilde{\tau}_0)\tilde{L}(q,\Omega)} + \int_0^\infty d\omega \rho(\omega)\frac{1}{N^*\tau_R'} \right.$$

$$\left. \times \frac{\Psi}{1-(\tau/\tau_0)L(q,\Omega)} \right\},$$

$$F(q,\Omega) = \lambda(q,\Omega)\pi_R'\left[a + \frac{1}{\tau_R'}\left(\frac{n}{f_\smile}\right)^2\right.$$

$$\left. \times \exp\{\beta(\hbar\omega+\Delta)\}\,\delta f_\wedge(q,\Omega)\right],$$

$$\tilde{F}(q,\Omega) = \tilde{\lambda}(q,\Omega)\tau_R'\left[\tilde{a} + \frac{1}{\tilde{\tau}_{fs}}\text{cth}^2\left(\frac{\beta\Delta}{2}\right)\delta f_\wedge(q,\Omega)\right]. \tag{13}$$

We can now calculate the energy characteristics of the heat pulse. The nonequilibrium increment in the energy of the system of spins is evidently related to $\delta f_\wedge(z,t)$ by $E_s(z,t) = N^*\delta f_\wedge(z,t)\Delta$. The nonequilibrium increment in the energy of the resonant phonons is evidently given by $E_{rf}(z,t) = \Delta\rho(\Delta)\delta\Delta\tilde{F}(z,t)$, where $\delta\Delta$ is the broadening of the distribution energy of two-level systems. It is convenient to write the total energy of the system of nonequilibrium nonresonant phonons and spins:

$$E_{rf+s} = E_{rf}(z,t) + E_s(z,t) = \tilde{\tau}_{fs}\tilde{a}\Delta\rho(\Delta)\delta\Delta + E_s(z,t)\left(1 + \frac{\tilde{\tau}_{fs}}{\tau_{sd}}\right).$$

We can discard the first term here, since it describes the injection of resonant phonons into the crystal from the $z = 0$ plane at $t = 0$.

We thus have

$$E_{rf+s}(z,t) \approx \left(1 + \frac{\tilde{\tau}_{fs}}{\tau_{sd}}\right)E_s(z,t).$$

Under the condition $\tilde{\tau}_{fs} \ll \tau_{sd}$, most of the energy is in the spin subsystem, while under the condition $\tilde{\tau}_{fs} \gg \tau_{sd}$ the opposite is true. We denote by $E_f(z,t)$ the energy of the nonresonant phonons:

$$E_f(z,t) = \int_0^\infty d\omega\,\rho(\omega)\hbar\omega F(z,t).$$

Using (13), we find an expression for the total energy $E(z,t) = E_f(z,t) + E_{rf+s}(z,t)$:

$$E(z,t) = \int_0^\infty d\omega\,\rho(\omega)\hbar\omega a(\omega)\frac{1}{(2\pi)^2}\int_{-\infty}^{+\infty} dq\,e^{-iqz}$$

$$\times \int_{-\infty}^{+\infty} d\Omega\,e^{i\Omega t}\frac{1}{i\Omega + q^2 D + 1/\tau_R}$$

$$+ \frac{\Delta}{(2\pi)^2}\int_{-\infty}^{+\infty} dq\,e^{-iqz}\int_{-\infty}^{+\infty} d\Omega\,e^{i\Omega t}\left\{i\Omega + \left(1 + \frac{\tilde{\tau}}{\tau_{sd}}\right)^{-1}\right.$$

$$\times \left[\Omega_{sR} + q^2\tilde{D}\frac{\tilde{\tau}}{\tau_{sd}} - \int_0^\infty d\omega\,\rho(\omega)\frac{\lambda(q,\Omega)}{N^*\tau_R'}\left(\frac{n}{f_\smile}\right)^2\right.$$

$$\left.\left. \times e^{\beta(\hbar\omega+\Delta)}\right]\right\}^{-1}\left\{\left[\rho(\Delta)\tilde{a}\delta\Delta + \int_0^\infty d\omega\,\rho(\omega)\lambda(q,\Omega)a\right]\right.$$

$$\left. \times \left[1 + \int_0^\infty d\omega\rho(\omega)\hbar\omega\frac{\lambda(q,\Omega)}{N^*\Delta}\left(1 + \frac{\tilde{\tau}_{fs}}{\tau_{sd}}\right)^{-1}\left(\frac{n}{f_\smile}\right)^2 e^{\beta(\hbar\omega+\Delta)}\right]\right\}. \tag{14}$$

The first integral describes the usual diffusion of nonequilibrium phonons with a diffusion coefficient $D(\omega)$ for each group of nonequilibrium phonons. The only difference from ordinary diffusion is that the number of phonons is not conserved; it decreases, because of the excitation of the two-level systems. We denote this term by $E_1(z,t)$. Carrying out the integration, we find

$$E_1(z,t) = \int_0^\infty d\omega\rho(\omega)\hbar\omega a\frac{1}{2(\pi D(\omega)t)^{1/2}}$$

$$\times \exp\left\{-\frac{z^2}{4D(\omega)t} - \frac{t}{\tau_R(\omega)}\right\}. \tag{15}$$

The second term in (14) evidently describes a more complex process, in which an energy exchange among nonresonant phonons, resonant phonons, and spins is important. We denote this term by $E_2(z,t)$. In it, we discard the term $\rho(\omega)\tilde{a}\delta\Delta$, since $\delta\Delta/\Delta$ is small. In addition, it is easy to show that

$$\int_0^\infty d\omega\rho(\omega)\hbar\omega\frac{\lambda(q,\Omega)}{N^*\Delta}\left(1 + \frac{\tilde{\tau}_{fs}}{\tau_{sd}}\right)^{-1}\left(\frac{n}{f_\smile}\right)^2$$

$$\times \exp[\beta(\hbar\omega+\Delta)] \ll 1,$$

since the heat capacity of the spin system is higher than that of the phonons. Also assuming $\tilde{\tau}_{fs} \ll \tau_{sd}$, we discard terms which are small in comparison with the remaining terms by a factor on this order. We find

$$E_2(z,t) = \frac{\Delta}{(2\pi)^2}\int_{-\infty}^{+\infty} dq\,e^{-iqz}\int_{-\infty}^{+\infty} d\Omega\,e^{i\Omega t}\left\{i\Omega + \left(1 + \frac{\tilde{\tau}}{\tau_{sd}}\right)^{-1}\right.$$

$$\times \left[\Omega_{sR} + q^2\tilde{D}\frac{\tilde{\tau}}{\tau_{sd}}\right.$$

$$\left.\left. - \int_0^\infty d\omega\rho(\omega)\frac{\lambda(q,\Omega)}{N^*\tau_R'}\left(\frac{n}{f_\smile}\right)^2\exp[\beta(\hbar\omega+\Delta)]\right]\right\}^{-1}$$

$$\times \int_0^\infty d\omega\rho(\omega)\lambda(q,\Omega)a. \tag{16}$$

It is difficult to pursue the analysis of general expression (16) further. The reason is that (16) contains an average over the phonon distribution, and the quantities $\lambda(q,\Omega)$ and $(1/\tau_R')\lambda(q,\Omega)$ (and also the sign of $1/\tau_R'$), which are to be averaged, depend strongly on the phonon energy. The

expression for $\lambda(q,\Omega)$ contains the quantities $D(\omega)\tau_R(\omega)$ and $\tau_R(\omega)$. For Rayleigh scattering of phonons by impurities we would have $D(\omega)\propto\omega^{-4}$. For Raman scattering of phonons under the condition $\omega>\Delta$ we again have $\tau_R(\omega)\propto\omega^{-4}$. Consequently, the conclusions reached below will be only qualitative.

Let us assume that the result we find when we take the average over the phonon distribution in (16) is determined by some predominant group of phonons with energies on the order of ω_d. Since the function $\lambda(q,\Omega)$ depends very strongly on the phonon energy, ω_d will generally be a function of the average phonon energy and the thickness of the sample. We assume $\omega_d\simeq k_B T$, and we ignore the L dependence of ω_d. In place of

$$\int_0^\infty d\omega\rho(\omega)\lambda(q,\Omega)a$$

we then write

$$\frac{1}{1+q^2D\tau_R+i\Omega\tau_R}\int_{\omega_m}^\infty d\omega\rho(\omega)\frac{\tau_R}{\tau_R'}a(\omega),$$

where D and τ_R now correspond to the predominant group of phonons. The function in the integrand in

$$\int_0^\infty d\omega\rho(\omega)\frac{\tau_R}{\tau_R'}a(\omega)$$

is a sign-varying function. Its sign corresponds to either a pumping of the spin system in the course of Raman scattering of phonons or a drawing of energy in a collision of a phonon with a two-level system in the excited state. In the integration of the function $\rho(\omega)\lambda(q,\Omega)a$ over $d\omega$, high-frequency phonons are predominant, since the relation $|\lambda(q,\Omega)|\propto 1/|1+q^2D\tau_R+i\Omega\tau_R|\ll 1$ holds for the low-frequency phonons. For this reason, we have replaced the lower integration limit by ω_m, under the assumption

$$\int_{\omega_m}^\infty d\omega\rho(\omega)\frac{\tau_R}{\tau_R'}a(\omega)>0.$$

This assumption corresponds to a pumping of the spin subsystem by the nonequilibrium phonons. In place of

$$\int_0^\infty d\omega\rho(\omega)\frac{\lambda(q,\Omega)}{N^*\tau_R'}\left(\frac{n}{f_V}\right)^2\exp\{\beta(\hbar\omega+\Delta)\}$$

we write

$$\frac{\Omega_{sR}'}{1+q^2D\tau_R+i\Omega\tau_R},$$

where we have introduced the new quantity Ω_{sR}' by means of

$$\Omega_{sR}=\int_0^\infty d\omega\rho(\omega)\frac{1}{N^*\tau_R}\left(\frac{\tau_R}{\tau_R'}\right)^2\left(\frac{n}{f_V}\right)\exp\{\beta(\hbar\omega+\Delta)\}.$$

In place of (16) we find a far simpler expression for $E_2(z,t)$:

$$E_2(z,t)=\frac{\Delta}{(2\pi)^2}\int_{\omega_m}^\infty d\omega\rho(\omega)\frac{\tau_R}{\tau_R'}a\int_{-\infty}^{+\infty}dq\int_{-\infty}^{+\infty}d\Omega$$

$$\times\frac{\exp\{i\Omega t-iqz\}}{(i\Omega+\Omega_{sR}+q^2\tilde{D}\tilde{\tau}_{fs}/\tau_{sd})(1+q^2D\tau_R+i\Omega\tau_R)-\Omega_{sR}'}. \qquad (17)$$

Integrating over $d\Omega$ in (17), we finally find

$$E_2(z,t)=\frac{\Delta}{2\pi}\int_{\omega_m}^\infty d\omega\rho(\omega)\frac{a}{\tau_R'}\int_{-\infty}^{+\infty}dq\frac{e^{-iqz}}{(\Omega_1-\Omega_2)}(e^{-\Omega_2 t}-e^{-\Omega_1 t}), \qquad (18)$$

where $i\Omega_1$ and $i\Omega_2$ ($\Omega_1>0$ and $\Omega_2>0$) are the poles of the integrand in (17). These poles are given by

$$\Omega_{1,2}=\frac{1}{2\tau_R}\left\{1+\Omega_{sR}\tau_R+q^2\left(D+\tilde{D}\frac{\tilde{\tau}_{fs}}{\tau_{sd}}\right)\tau_R\right.$$

$$\left.\pm\left[\left(1+q^2D\tau_R-\Omega_{sR}\tau_R-q^2\tilde{D}\frac{\tilde{\tau}}{\tau_{sd}}\tau_R\right)^2+4\Omega_{sR}'\tau_R\right]^{1/2}\right\}. \qquad (19)$$

It is obvious from (19) that the relation $\Omega_2\ll\Omega_1$ holds. Setting $\Omega_{sR}\tau_R\ll 1$ and $\Omega_{sR}'\tau_R\ll 1$ and expanding the square root in (19) in a power series, we find

$$\Omega_1\approx\frac{1}{\tau_R}+q^2 D,$$

$$\Omega_2\approx\Omega_{sR}+q^2\tilde{D}\frac{\tilde{\tau}}{\tau_{sd}}-\Omega_{sR}'\left[1+q^2\left(D-\tilde{D}\frac{\tilde{\tau}_{fs}}{\tau_{sd}}\right)\tau_R\right]^{-1}.$$

$$(20)$$

It follows immediately that the maximum of the signal $E_2(z,t)$ is determined by the contribution from the pole $\Omega=i\Omega_2$ and occurs long after arrival time t_1. The integration over dq in (18) can be carried out easily by numerical methods, with the help of (19) or (20).

Let us examine the contribution $E_1(z,t)$ in more detail. If $\tau_R(\overline{\omega})>L^2/D(\overline{\omega})$, we can discard the second term in the argument of the exponential function in (15) for time intervals t which are not greatly longer than t_1. The reason is that Raman-pumping processes are unimportant for these time intervals. In this case expression (15) corresponds to ordinary diffusion. The time at which the maximum of the deviation of the phonons from equilibrium arrives is t_1, and the dependence of this time on the temperature and the thickness of the sample is $t_1\propto T^4 L^2$. In the case $\tau_R(\overline{\omega})<L^2/D(\overline{\omega})$, Raman pumping processes play an important role, and the arrival time of the maximum of the phonon deviation from equilibrium is on the order of $[t_1\tau_R(\overline{\omega})]^{1/2}$, proportional to L and independent of the temperature.

We now consider $E_2(z,t)$. We first note that the absolute value of $E_2(z,t)$ near the maximum is on the same order of magnitude as $E_1(z,t)$ as can be seen by comparing (15) and (17). Consequently, the heat pulse does indeed transfer a substantial fraction of its energy to the spin subsystem as it propagates through the crystal. Figure 1 shows the results of a numerical integration of (18). Specifically, this figure shows a family of curves of the arrival time of the second
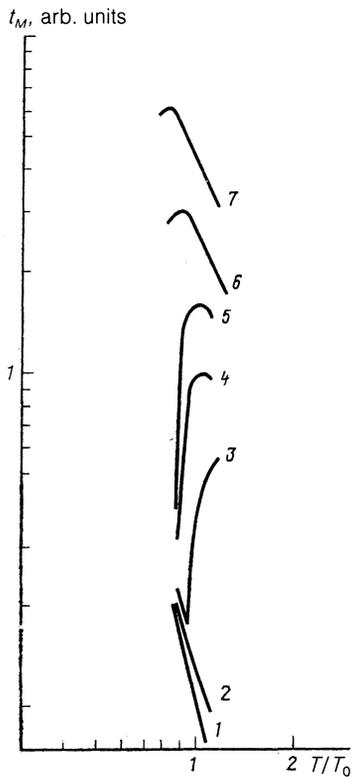
t_M, arb. units

FIG. 1. Calculated temperature dependence of the arrival time of the second maximum of the signal. 1—$B = 8$; 2—$B = 4$; 3—$B = 1$; 4—$B = 0.4$; 5—$B = 0.25$; 6—$B = 0.1$; 7—$B = 0.05$.

maximum of the signal, t_2, versus the crystal temperature. The various curves in this family correspond to different values of the characteristic parameter $B = (l_R/L)^2$ at some temperature T_0. These curves have regions with positive and negative derivatives.

The second maximum of the signal stems from the effective pumping of the two-level systems by the heat pulse. Putting aside the contribution from long-wavelength phonons, which are incapable of transferring energy to the two-level systems, we can draw the following picture of the energy transport through the crystal.

Because of the strong elastic scattering, the phonons are delayed times on the order of τ_R in the crystal. On the average, there is a high probability that each phonon will transfer an energy on the order of its own energy to the two-level systems over a time interval τ_R. Over a time τ_{sd}, the excitation energy of a center is converted into the energy of resonant phonons. If the concentration of two-level systems is high, however, these phonons experience a capture effect and cannot bring about a rapid spatial transport of energy. The effective spatial transport of energy therefore results from the slower process of Raman relaxation of the centers. This relaxation occurs over times on the order of τ_{sR}; as a result, nonresonant phonons with a far larger diffusion coefficient are formed.

After an event in which a center has relaxed, the emitted phonon reaches the bolometer after a time $t \lesssim t_1$ if, in the course of its motion, it does not excite another two-level system. The scale of this signal delay (the arrival time of the second maximum of the signal) in the crystal is determined

by the time τ_{sR} under these conditions. The delay time and the temperature dependence of the position of the second maximum of the signal also depend on the probability for a repeated Raman pumping of the two-level systems by phonons emitted by the excited centers in the course of Raman relaxation. The result can be described at a qualitative level as follows. We assume that at the average experimental temperature T_0 we have $B(T_0) \simeq 1$. At $T < T_0$, the condition $B(T) > B(T_0)$ holds. After relaxation, the phonons reach the bolometer without obstacle. The characteristic delay of the signal is determined by the value of τ_{sR}. It decreases with increasing T (see curves 1 and 2 and also the low-temperature part of curve 3 in Fig. 1). At $T > T_0$, on the other hand, events of a repeated pumping of the centers by phonons emitted in the course of the relaxation become important, because of the rapid increase in both the probability for Raman pumping and the relaxation rate of the centers. In this case there is a "capture" of nonresonant phonons. The signal delay is far longer in this case than in the case $T < T_0$. In a narrow transition region, the arrival time of the second maximum of the signal increases sharply with increasing temperature. With a further increase in the temperature, the effective diffusion coefficient for excitations in the spin system is determined in order of magnitude by l_R^2 (the mean square of the distance traveled by the phonons emitted in the course of the relaxation of the centers before they undergo another Raman-pumping event), divided by the excitation transfer time. The latter differs from τ_{sR} since the rate of change of the nonequilibrium increment in the spin distribution function depends on the deviation of the phonons from equilibrium. The appearance of a region with a negative derivative $\partial t_2/\partial T$ is associated with this circumstance.

At a qualitative level, these results give a satisfactory explanation of the data of the corresponding experiments.[12] Specifically, the case of high concentrations of erbium ions[12] in the proposed systematics corresponds to the condition of an intense Raman pumping of the spin subsystem by the phonons (the parameter B is small at a certain average experimental temperature). The decreasing part of the upper curves in Fig. 1 apparently gives a good description of the experimental results of Ref. 12. As the temperature is lowered in samples of the same composition, and/or when the sample thickness is changed, however, the intensity of the Raman pumping changes. This change should ultimately lead to a change in the sign of the derivative of the temperature dependence of the arrival time of the second signal maximum. The temperature dependence itself becomes very strong in this case. The case of low concentrations of erbium ions[12] evidently corresponds to large values of the parameter $B > 1$ at the average experimental temperature. As in the experiments, the temperature dependence of t_2 is very strong, and we have $(\partial t_2/\partial T) > 0$. In order to observe a behavior like that shown in Fig. 1, it would be important to use samples of identical thickness. The reason why this circumstance is important is that the characteristic frequency of the predominant group of phonons depends on the thickness of the crystal. If experiments are instead carried out with crystals of identical composition (with a fixed concentration of two-level systems), and the parameter B is varied by carrying out the measurements in crystals differing in thickness, the series of $t_2(T,B)$ curves should of course be qualitatively the same as the series in Fig. 1. However, the behavior of the

parameter B as a function of the crystal thickness is not a simple question.

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