Propagation of phonons in crystals containing anisotropic resonant-scattering centers

K. L. Aminov and B. Z. Malkin
V. I. Ul'yanov-Lenin Kazan' State University

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The kinetic equations are solved in the absence of spectral diffusion. The solution describes the propagation of nonequilibrium phonons with $r \sim t^{2/3}$.

This letter concerns the development of a phonon spectroscopy of paramagnetic crystals based on methods for selectively detecting nonequilibrium phonons.

Let us assume that a crystal contains impurity two-level electronic centers with a spectral density $\delta N(\omega)$ of the difference between the populations (per unit volume) of

the ground state ($|1\rangle$) and the excited state ($|2\rangle$). The Hamiltonian of the interaction of a center with acoustic phonons can be written in the form $\mathscr{H} = \Sigma_{\lambda\Gamma} V_\lambda(\Gamma) e_\lambda(\Gamma)$, where the operators $V_\lambda(\Gamma)$ are defined in the space of states of the center, and the quantities $e_\lambda(\Gamma)$ are linear combinations of the components of the dynamic strain tensor $e_{\alpha\beta}$, which transform in accordance with a row λ of an irreducible representation Γ of the point symmetry group of the center. The lifetime of a phonon with a wave vector $\mathbf{q}$ and a frequency $\omega_j(\mathbf{q})$ (j specifies the branch of the vibrational spectrum), which is determined by the resonant absorption by electronic centers, is $\tau_{j\mathbf{q}} = (\delta N^0(\omega_j(\mathbf{q}))W_j(\mathbf{q}))^{-1}$, where δN^0 is the equilibrium spectral density of the population difference, and

$$W_j(\mathbf{q}) = \sum_{\lambda\Gamma} \frac{\pi\omega_j(\mathbf{q})}{\hbar\rho v_{j\mathbf{q}}^2} |\langle 1|V_\lambda(\Gamma)|2\rangle|^2 f^j_{\lambda\Gamma}(\mathbf{q}/q) .$$

Here ρ is the density of the crystal, $v_{j\mathbf{q}}$ is the phonon velocity, and $f^j_{\lambda\Gamma}$ is the square of a bilinear form of the direction cosines of the polarization vector and the wave vector of the phonon. In the case of a low phase-relaxation rate, and with a highly nonuniform broadening of the electronic transition (the linewidth satisfies $\Delta\omega \gg \tau_{j\mathbf{q}}^{-1}$ if the concentration of scattering centers is sufficiently low), the linearized kinetic equations for the deviations of the phonon occupation numbers $\Delta n_{j\mathbf{q}} = n_{j\mathbf{q}} - n^0_{j\mathbf{q}}$ and the spectral density of the excitations of centers, $\Delta N(\omega) = \delta N^0(\omega) - \delta N(\omega)$, from the corresponding equilibrium values are[1]

$$\frac{\partial \Delta n_{j\mathbf{q}}}{\partial t} + \mathbf{v}_{j\mathbf{q}} \nabla(\Delta n_{j\mathbf{q}}) = -\tau_{j\mathbf{q}}^{-1}\left(\Delta n_{j\mathbf{q}} - \frac{\langle\tau(\omega)\rangle}{\tau}\frac{\Delta N(\omega)}{2\rho_0}\right), \tag{1}$$

$$\frac{\partial \Delta N(\omega)}{\partial t} = -\frac{1}{\tau}\Delta N(\omega) + 2\sum_j \int \frac{d^3q}{(2\pi)^3}\delta(\omega - \omega_j(\mathbf{q}))\frac{\Delta n_{j\mathbf{q}}}{\tau_{j\mathbf{q}}}, \tag{2}$$

where $\rho_0 = \Sigma_j \int [d^3q/(2\pi)^3]\delta(\omega - \omega_j(\mathbf{q}))$ is the density of lattice states per unit volume per unit frequency interval, τ is the lifetime of an electronic excitation, and $\langle\tau(\omega)\rangle$ is the expectation value of the lifetime of the phonons with a frequency ω, given by

$$\frac{1}{\langle\tau(\omega)\rangle} = \frac{1}{\rho_0}\sum_j \int \frac{d^3q}{(2\pi)^3}\frac{1}{\tau_{j\mathbf{q}}}\delta(\omega - \omega_j(\mathbf{q})).$$

At times $t \gg \tau_{j\mathbf{q}}$, at which the phonon subsystem reaches a state of equilibrium with the electronic subsystem, the spatial Fourier transforms of the variables $\Delta n_{j\mathbf{q}}(\mathbf{r},t)$, $\Delta N(\omega,\mathbf{r},t)$ are related by the following equation, according to Eq. (1):

$$\Delta n_{j\mathbf{q}}(\mathbf{k},t) = (1 + i\tau_{j\mathbf{q}}\mathbf{k}\,\mathbf{v}_{j\mathbf{q}})^{-1}\frac{\langle\tau(\omega)\rangle}{\tau}\frac{\Delta N(\omega,\mathbf{k},t)}{2\rho_0} . \tag{3}$$

The total number of excitations with a spectral density (per unit volume) $J(\omega) = \frac{1}{2}\Delta N(\omega) + \Sigma_j \int [d^3q/(2\pi)^3]\delta(\omega - \omega_j(\mathbf{q}))\Delta n_{j\mathbf{q}}$ remains constant. Substitution

of expression (3) into the conservation law in differential form yields a transport equation (in the limit $\mathbf{k}\to 0$; the flux density of excitations is determined by the drift of phonons):

$$\frac{\partial \Delta N(\omega, \mathbf{k}, t)}{\partial t} + \left(1 + \frac{\tau}{\langle \tau(\omega)\rangle}\right)^{-1} A(\mathbf{k})\, \Delta N(\omega, \mathbf{k}, t) = 0, \tag{4}$$

where

$$A(\mathbf{k}) = \frac{1}{\rho_0} \sum_j \int \frac{d^3q}{(2\pi)^3}\, \delta(\omega - \omega_j(\mathbf{q}))\, \frac{\tau_{j\mathbf{q}}^{-1} (\mathbf{k}\cdot\mathbf{v}_{j\mathbf{q}})^2}{\tau_{j\mathbf{q}}^{-2} + (\mathbf{k}\cdot\mathbf{v}_{j\mathbf{q}})^2}\,. \tag{5}$$

The behavior of $A(\mathbf{k})$ in the limit $\mathbf{k}\to 0$ (the length scale of interest here is $L \sim 1/k \gg v_{j\mathbf{q}}\tau_{j\mathbf{q}}$) depends strongly on the form of $\tau_{j\mathbf{q}}$, as a function of the direction of the phonon wave vector. Depending on the symmetry of the lattice and the scattering center and also on the nature of the transition, the function $f^j_{\lambda\Gamma}(\mathbf{q}/q)$ (see the discussion above) (a) may be constant, (b) may vanish on a line, or (c) may vanish at individual points on a sphere centered at the origin of coordinates in q space. Only in the first of these cases ($\tau_{j\mathbf{q}}^{-1} > 0$) does the asymptotic expansion of $A(\mathbf{k})$ begin with a quadratic form of the components of the vector $\mathbf{k}$, corresponding to a diffusive regime of the propagation of the nonequilibrium phonons.[1,2] In case c the leading term of the expansion $A(\mathbf{k})$ is proportional to $k^2 \ln k$. The transport process[3] has its fastest propagation regime ("nonlocal") in the most general case, b, in which the expansion of $A(\mathbf{k})$ is represented by a power series in $k^{-1/2}$, and the leading term of the series is $A(\mathbf{k}) \sim F(\theta_k, \varphi_k) k^{3/2}$, where k, θ_k, and φ_k are the spherical coordinates of the vector $\mathbf{k}$. As an example, we write the explicit expression for the function $F(\theta_k, \varphi_k)$ for the resonant scattering of longitudinal phonons (considered in the approximation of an isotropic medium) on a singlet-doublet transition of non-Kramers paramagnetic ions in a crystal field of tetragonal symmetry (transitions are induced by the strains e_{xz} and e_{yz}; z is the symmetry axis):

$$A(\mathbf{k}) \approx \frac{(\Gamma(1/4))^2}{6\sqrt{30\pi}}\, |\sin\theta_k|^{3/2} (v_l k)^{3/2} \langle \tau(\omega)\rangle^{1/2}\,.$$

The length scale of the spatial distribution of nonequilibrium phonons increases over time in accordance with $t^{2/3}$, instead of the $t^{1/2}$ law in the case of ordinary diffusion. The time scale of the process is proportional to the square root of the concentration of scattering centers, while in the case of diffusion there would be a direct proportionality. It may be that the effects which stem from an anisotropy of resonant scattering determine those features of the propagation of nonequilibrium phonons and of the luminescence kinetics in ruby and alexandrite active media as functions of the concentration of excited chromium ions which were reported in Refs. 4 and 5.

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