

Absorption of acoustic waves in complex dielectric crystals with impurities

Yu. V. Gulyaev, A. G. Kozorezov, and M. V. Krasil'nikov

Institute of Radio Engineering and Electronics, USSR Academy of Sciences

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The absorption of acoustic waves in complex dielectric crystals with impurities is investigated. It is shown that under certain conditions, the contribution of optical phonons to the absorption cannot be neglected. An expression is found for the optical-phonon relaxation time in a complex crystal under scattering from impurities. The change in the temperature dependences of the sound absorption coefficient in impure crystals, relative to ideal matrices, is analyzed. It is shown that in intense phonon-impurity scattering, the temperature dependence of the sound absorption coefficient should be stronger than in the ideal matrix or in the model of a simple crystal in which the sound absorption is due to the acoustic phonons. Some criteria are suggested for choosing the characteristics of the substituting and substituted atoms in order to obtain the maximum decrease in the transverse sound absorption coefficient over a broad range of temperatures.

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The absorption of acoustical waves in dielectric crystals with impurities can differ appreciably from the absorption in ideal crystals. A number of theoretical and experimental works have been devoted to the study of sound absorption in impure dielectric crystals (see, for example, Refs. 1–13). The direct interaction of acoustic waves with optical phonons is, as a rule, not analyzed in detail, although a generalization, for example, of the Akhiezer theory to this case is well known.

In addition, there are crystals in which, over a wide range of temperatures, excluding only the very lowest, one cannot neglect the direct interaction of the initial acoustic wave with the optical phonons.⁶ To such crystals we should add complex crystals with a large number of atoms in the elementary cell, for example, crystals of the type of aluminium yttrium garnet $Y_3Al_5O_{12}$ ^{9,10,12}, rutile TiO_2 ¹⁴ and so on. The presence of a large number of low-lying optical branches leads to a sharp increase in the density of vibrational states of the crystal, beginning with energies corresponding to optical phonons. Probably, even at temperatures of liquid nitrogen, the direct effect of the optical phonons on the sound absorption in $Y_3Al_5O_{12}$ should be significant (even if the contribution to the absorption from a single optical mode is much less than the corresponding contribution of the acoustical mode).

The present work has been stimulated by a number of experimental researches into sound absorption in aluminum yttrium garnets.^{9,10,12,13} The aim of the work is the study of the features of the absorption of acoustic waves in complex dielectric crystals with impurities. In contrast to Refs. 1–13, we shall assume from the very beginning that it is just this interaction of the acoustic wave with the optical phonons which is the principal reason for sound absorption over a wide range of temperatures.

We consider a dielectric crystal containing R atoms in the elementary cell. We assume that the crystal contains a large number of impurity atoms, that replace atoms of the matrix at the positions κ_1 (κ enumerates the position of the atoms in the elementary cell). Let an acoustic wave of frequency ω_s and wave vector \mathbf{q} propagate in the crystal in one of the symmetry directions. We shall assume that the tem-

perature is sufficiently high $T \gtrsim T_0$ (at $T = T_0$, the characteristic energy of the phonons is of the order of ω_{min} , where ω_{min} is the minimum frequency in the spectrum of optical phonons). For ordinary crystals in the given range of temperatures, the mean relaxation time of thermal phonons is much less than the period of the sound wave (for all sound frequencies, including the microwave region). Thus the coupling between the perturbation in the phonon system and the deformations produced in the medium by the acoustic wave is local. It was noted in Ref. 2 that in the case of scattering of optical phonons by impurities, the role of the longwave optical phonons (in contrast to the longwave acoustical phonons) in the sound absorption is a small one. This means that the fundamental contribution to the sound absorption will be played by the group of optical phonons belonging to the regions of the space of wave vectors for which the density of states is large.

It is clear from what has been said that we can use the approach of Maris for analysis of the sound absorption in the crystals considered.¹⁵ The approach has been modified somewhat to take into account the dependence of the time of phonon relaxation by the impurities on the wave vector.⁵

It is easy to show that, as in the case of the absorption of acoustic waves in simple crystals, the absorption of longitudinal waves depends weakly on the presence of impurities in the crystal. This result is general, for the absorption of both acoustical and optical phonons. It follows from the basic assumption of Maris, according to which the relaxation energy of the phonons is due to anharmonism, while the scattering of the phonons from impurities is quasi-elastic. We note that the assumption of Maris is well satisfied under those conditions when the phonon-phonon relaxation changes weakly upon injection of impurities, for example, because of a weak renormalization of the elastic moduli and the density of the crystal.¹¹

In what follows, we discuss in detail the problem of the absorption of transverse sound. We consider the most interesting case, in which the scattering of phonons by impurities is dominant (in the opposite case, the sound absorption in the impure crystal will differ little from absorption in an ideal

matrix). As experimental results on sound absorption in $Y_3Al_5O_{12}$ crystals alloyed with ions of rare-earth metals testify,⁹⁻¹³ the effect of the impurities on the absorption of the acoustic waves is significant over a wide range of high temperatures, beginning at 40–50 K. This allows us to assume (in the absence of reliable numerical estimates of the value of the phonon-phonon and phonon-impurity scattering in complex crystals), that the model discussed is entirely realistic.

The expression for the absorption coefficient in the considered case has the usual Akhiezer form:

$$\Gamma_i = \frac{1}{2} \frac{\hbar^2 \omega_s^2}{T \rho V S^3} \sum_{\mathbf{k}j} \gamma_s^2(\mathbf{k}j) \omega^2(\mathbf{k}j) n(\mathbf{k}j) [n(\mathbf{k}j) + 1] \tau_e(\mathbf{k}j). \quad (1)$$

Here ρ and V are the density and volume of the crystal; S is the velocity of the original wave; $\hbar\omega(\mathbf{k}j)$ is the energy of a phonon with polarization j and quasimomentum $\hbar\mathbf{k}$; $n(\mathbf{k}j)$ is the equilibrium Planck distribution function of the phonons; τ_e is the relaxation time in the case of scattering of the phonons from impurities; γ_s is a symmetric combination of Gr \ddot{u} nenstein constants.

However, the problem of the temperature dependence of the sound absorption coefficient is not trivial, nor is the problem of the dependence of the absorption on the type of the substituted atoms and on the characteristics the substituting atoms.

Thus, our basic aim is the investigation of the character of the temperature dependence of the absorption of acoustic waves in complex dielectric crystals with impurities. It is evident that, in order to answer the posed questions, we must obtain and analyze an expression for the relaxation time of the phonons scattering from impurities in the complex crystal. Under the simplest assumptions we can use standard procedures for the derivation of the given expression.^{17,18} We need only take account of the specific features of the complex crystal in the corresponding Dyson equations for the Green's function. We limit ourselves to the study of only the isotopic model of the defect, assuming that the basic qualitative rules for the sound absorption for a number of complex crystals can be described within the framework of this simplified model. We note that for solid solutions of the substitution type $Y_3Al_5O_{12}-Re_3Al_5O_{12}$ (the symbol Re denotes any of the rare-earth atoms of the lanthanide series), upon isomorphic substitution of ions of the rare-earth atoms for the Y ions, by virtue of the closeness of the ionic radii of Y and Re (as data of the experimental studies of the acoustical¹⁹ and optical²⁰ properties of these solutions show), the change in the force constants is not large and does not exceed several per cent. Thus, we can expect that the model we have used for these systems is completely valid.

The Fourier transform of the Green's function of the crystal $G_{\alpha\beta}(\mathbf{k}, \omega)$, averaged over the random distribution of impurity atoms, in the approximation of the averaged T matrix, satisfies the following Dyson equation:¹⁷

$$G_{\alpha\beta}(\mathbf{k}, \omega) = G_{\alpha\beta}^0(\mathbf{k}, \omega) + \sigma(\chi_1) \sum_{\tau} G_{\alpha\tau}^0(\mathbf{k}, \omega) G_{\tau\beta}(\mathbf{k}, \omega), \quad (2)$$

$$\sigma(\chi_1) = c \Delta M_{\chi_1} \omega^2 \left[1 - 1/3 \Delta M_{\chi_1} \omega^2 G_{i_1 i_1}^0(\mathbf{0}, \omega) \right]^{-1}.$$

Here G^0 is the Green's function of the ideal crystal; c is the concentration of impurity atoms; $\omega \equiv \omega(\mathbf{k}j)$; $\Delta M_{\chi_1} = M_{\chi_1} - \bar{M}_{\chi_1}$, where M_{χ_1} and \bar{M}_{χ_1} are the masses of the substituted and substituting (impurity) atoms, respectively.

Equation (2) was obtained with the assumption that $c \ll 1$ and is written down for a cubic crystal under the condition that the location of the substitution has cubic symmetry. In these approximations, the solution of Eq. (2) for G has the form

$$G_{\alpha\beta}(\mathbf{k}, \omega) = \sum_j (M_{\chi_1} M_{\chi_1'})^{-1/2} e_{\alpha}(\chi_1 | j, \mathbf{k}) e_{\beta'}(\chi_1' | j, \mathbf{k}) \times \left[\omega^2 - \omega^2(\mathbf{k}j) - \frac{\sigma(\chi_1)}{M_{\chi_1}} |e(\chi_1 | j, \mathbf{k})|^2 \right]^{-1}, \quad (3)$$

where $e_{\alpha}(\chi_1 | j, \mathbf{k})$ is the α -th component of the vibration polarization vector of the χ_1 -th atom in the mode $\mathbf{k}j$; M_{χ_1} is the mass of this atom in an ideal crystal.

Calculating the imaginary part of the pole of the Green's function, we find the relaxation time of the phonon of the $\mathbf{k}j$ -th mode:

$$\tau_e^{-1}(\mathbf{k}j) = c\pi \frac{\varepsilon^2}{1-\varepsilon} \Phi_{\chi_1}(\omega^2(\mathbf{k}j)) \omega^3(\mathbf{k}j) |e(\chi_1 | j, \mathbf{k})|^2, \quad (4)$$

where $\varepsilon = \Delta M_{\chi_1} / M_{\chi_1}$, and $\Phi_{\chi_1}(\omega^2)$ is the spectral density of the square of the displacement of the impurity atom. It has the form²¹

$$\Phi_{\chi_1}(\omega^2) = (1-\varepsilon) g_{\chi_1}(\omega^2) \left\{ \left[1 - \varepsilon \omega^2 \int_0^{\omega_{0\max}^2} \frac{g_{\chi_1}(\omega_0^2) d\omega_0^2}{\omega^2 - \omega_0^2} \right]^2 + [\pi \varepsilon \omega^2 g_{\chi_1}(\omega^2)]^2 \right\}^{-1}, \quad (5)$$

where $g_{\chi_1}(\omega^2)$ is the spectral density of the square of the displacement of the atom of the χ_1 -th type in an ideal matrix:

$$g_{\chi_1}(\omega^2) = \frac{1}{3} \left(\frac{a}{2\pi} \right)^3 \sum_{j=i}^{3R} \int_{\omega^2=\omega_0^2(\mathbf{k}j)} dS_{\mathbf{k}} \frac{|e(\chi_1 | j, \mathbf{k})|^2}{|\nabla \omega_0^2(\mathbf{k}j)|}. \quad (6)$$

Here $\omega_{0\max}$ is the maximum frequency in the vibrational spectrum of the crystal; a^3 is the volume of the elementary cell; $dS_{\mathbf{k}}$ is the element of the phonon isoenergetic surface.

Both the functions $\Phi_{\chi_1}(\omega^2)$ and $g_{\chi_1}(\omega^2)$ are normalized to unity:

$$\int_0^{\infty} \Phi_{\chi_1}(\omega^2) d\omega^2 = 1, \quad \int_0^{\omega_{0\max}^2} g_{\chi_1}(\omega^2) d\omega^2 = 1.$$

The properties of the function $\Phi_{\chi_1}(\omega^2)$ have been investigated in detail by Iosilevskii and Kagan.²¹ Its form, and consequently, the characteristics of its scattering, depend on the location in the elementary cell at which the impurity atom is located, which is connected with the dependence of (5) on the polarization vectors of the substituted atom.

Along with the functions $\Phi_{\chi_1}(\omega^2)$, the value of the coupling constant is important for the scattering characteristics. In contrast to simple crystals, the value of the effective coupling constant depends not only on the relative change in the mass of the substituted atom (within the framework of the isotopic model), but also because of the factor $|e(\chi_1 | j, \mathbf{k})|^2$ in (4), on the position at which the substitution occurs. Thus, in order to obtain effective phonons scattering (and, consequently, a significant reduction in the absorption coefficient

of transverse acoustic waves in the region of corresponding temperatures), it is necessary to satisfy the following two conditions simultaneously.

- a) to choose the impurity such that the spectrum $\Phi_{x_1}(\omega^2)$ has a maximum in the given range of frequencies;
- b) to guarantee a relatively large value of the effective coupling constant.

It is clear that for realization of the second condition, we must replace atoms which oscillate mainly in the given range of frequencies. As applied to the $Y_3Al_5O_{12}$ crystals, this means, for example, that the maximum decrease in the absorption in the alloying of the crystals with atoms of rare-earth elements in place of Y takes place at temperatures corresponding to the spectral region in which the oscillations of the heavy Y atoms are chiefly localized. The replacement of the Y atoms with much lighter ones, in order to shift the spectrum $\Phi_{x_1}(\omega^2)$ toward higher frequencies, is ineffective, since, along with the shift mentioned, there will be a simultaneous weakening of the coupling constant.

We also note that, in principle, we can obtain a significant decrease of the absorption coefficient of transverse waves in complex crystals, relative to the ideal matrix, over a very wide range of temperatures, beginning with the temperature at which the Akhiezer absorption regime commences. For this, by virtue of what was said above, it is necessary to choose a number of necessary substitution positions in a suitable manner, and correspondingly the substituting impurities.

We now proceed to a detailed consideration of the temperature dependence of the absorption coefficient of the transverse sound in impure dielectric crystals. As before, we shall consider a crystal with a single type of substituting atom. We assume that the substituted atom is "attached" to the acoustic portion of the spectrum, i.e., the maximum of the function $g_{x_1}(\omega^2)$ is localized in a comparatively low-frequency region of the phonon spectrum near the frequency $\omega_c \ll \omega_{0\max}$. We now consider the case in which the substituting atom is heavier than the lattice atom, i.e., $\bar{M}_{x_1} > M_{x_1}$. According to (5) and (6) (see Ref. 21), the function $\Phi_{x_1}(\omega^2)$ is displaced relative to $g_{x_1}(\omega^2)$ in the direction of the lower frequencies. At temperatures $T \gtrsim T_c$ (T_c is the temperature at which the energy of the significant phonons is of the order of $\hbar\omega_c$) the principal role in the sound absorption is played by phonons with frequencies $\omega > \omega_c$, since in the region of the maximum $\Phi_{x_1}(\omega^2)$ the relaxation times of phonons scattered from the impurities are small (we recall that we are assuming that phonon scattering from impurities is dominant over a sufficiently wide range of frequencies). Thus, the phonons that are active in sound absorption are located in the region of the right-hand slope of the spectral line $\Phi_{x_1}(\omega^2)$. If in this region of the spectrum $\Phi_{x_1}(\omega^2)$ falls off according to a power law, then the mean relaxation time of the phonons, which determines the sound absorption coefficient (see Ref. 1) will increase with temperature, also according to a power law. Actually, by virtue of the normalization condition, $\Phi_{x_1}(\omega^2)$ and $|e(x_1|k)|^2$ in the region of the right-hand slope of the spectral line, $\Phi_{x_1}(\omega^2)$ falls off with increase in ω^2 more rapidly than ω^{-2} , while $|e(x_1|k)|^2$ falls off more rapidly than

$j^{-1} \propto \omega^{-1}$. Thus, in the indicated temperature range, the temperature dependence of the transverse sound absorption coefficient in an impure dielectric crystal will be much steeper than in an ideal matrix. At higher temperatures, finally, the scattering of phonons by impurities becomes ineffective, and the sound absorption tends towards the absorption in an ideal matrix. This is connected with the fact that the phonon relaxation time, which determines the sound absorption in the ideal matrix, falls off increase in temperature (we recall that within the framework of the considered model the temperature dependence of the longitudinal sound absorption coefficient is exactly the same as in the ideal matrix).

In the range of temperatures $T < T_c$, the relaxation time of the phonons from the impurities falls off with increase in temperature, while the corresponding temperature dependence of the transverse sound absorption coefficient is weakened. Schematically, such a temperature dependence is shown in Fig. 1 (it is assumed that the range of temperatures $T < T_c$ already corresponds to the Akhiezer regime of absorption).

We note that at $\omega > \omega_c$

$$\int_0^{\omega_{0\max}^2} \frac{g_{x_1}(\omega_0^2) d\omega_0^2}{\omega^2 - \omega_0^2} \approx \frac{1}{\omega^2}$$

and

$$\Phi_{x_1}(\omega^2) \approx (1-\varepsilon) g_{x_1}(\omega^2) \{ (1-\varepsilon)^2 + [\pi\varepsilon\omega^2 g_{x_1}(\omega^2)]^2 \}^{-1}.$$

Substituting this expression in (4), we establish the fact that for the case considered ($|\varepsilon| > 1, \varepsilon < 0$) τ_ε depends weakly on ε . Consequently, if the phonons which make the principal contribution to the sound absorption are in the region of the right slope of the function $\Phi_{x_1}(\omega^2)$ then the variation of the sound absorption coefficient for different impurities will be basically connected with the renormalization of the elastic moduli and the density of the crystal. A more significant dependence on the nature of the alloying impurity can arise at $T \lesssim T_c$, where the absolute value of the decrease in the absorption in the impure crystal will be sensitive to the character of the strain $\Phi_{x_1}(\omega^2)$ toward the side of lower frequencies.

We shall now say a few words regarding the limits of applicability of the reduced T -matrix approximation for calculation of the relaxation time of phonons on impurities in a complex crystal. First of all, we note that the conditions for the appearance of quasilocal oscillations in a complex crystal are very severe.²¹ Therefore, it is most probable that the function $\Phi_{x_1}(\omega^2)$ is quite smooth, so that even for phonons with energies $\sim \hbar\omega_c$, the parameter $\omega_c \tau(\omega_c) \gg 1$. Actually, setting $\Phi = \text{const}$ in the frequency interval $\Delta \sim \omega_c$ near ω_c and $|e(x_1|k)|^2 = \text{const}$ in the frequency range $\Delta_1 \sim \omega_c$, near the same frequency, and using the conditions of normalization of the functions ω_c and $|e|^2$, we obtain the rough estimate

$$\tau^{-1}(\omega_c) \sim c\omega_c^2\omega_D/R\Delta\Delta_1$$

and, correspondingly,

$$\omega_c \tau(\omega_c) \sim (R/c) (\omega_c/\omega_D) \gg 1.$$

Under these conditions, the approximation that we have

used is valid, as usual, outside the immediate vicinities of the ends of the intervals of the continuous spectrum, van Hove singular points, and points of intersection of the phonon dispersion curves, for which the only criterion is the smallness of the quantity c in comparison with unity. If the line $\Phi_{\kappa_1}(\omega^2)$ is very narrow in comparison with $g_{\kappa_1}(\omega^2)$ (i.e., a quasilocal oscillation is generated), then the limits of applicability of the results, as for simple crystals,¹⁸ become much more severe. Without specially analyzing this case, we note that the basic conclusion of the given work on the resonance character of the scattering of phonons by impurities is not connected with the generation of quasilocal oscillations.

Finally, we remark that, within the framework of the considered model, it is still necessary to require that the wavelength of the phonon be greater than the mean distance between impurities, i.e., $\lambda > ac^{-1/3}$ (only in this case can we use the averaged Green's function). This limitation turns out to be unimportant, however, if we consider the more realistic model of substitution at a large number of sites. [For example, in $Y_3Al_5O_{12}$, the substitution can take place at any of 24 Y positions in the elementary cell, so that for the compound $Y_{2.7}Lu_{0.3}Al_5O_{12}$ (i.e., $c = 0.1$) 2.4 impurity atoms fit on average in the elementary cell.] The expression for τ_e in this case turns out to be very complicated. For analysis of τ_e it is necessary to introduce the spectral density matrix of the quadratic combination of displacements of the impurity atom in the lattice. However, it can be expected that if sound absorption by optical phonons plays an important role in the crystal, while phonons are essentially scattered by impurities, then the effects of anisotropy do not lead to a qualitative change in the predicted features in the temperature dependence of the absorption.

We now estimate the contribution of the acoustical and optical modes to the absorption of transverse sound in an impure dielectric crystal. For this purpose, we consider the simplest case of a two-atom cubic crystal and analyze Eq. (4) for this case.

We shall assume that the mass of the impurity atom (m') differs little from the mass of the substituted atoms ($m_1 \approx m'$) so that $\Phi_{\kappa_1}(\omega^2) \approx g_{\kappa_1}(\omega^2)$. Then (4) takes the form

$$\tau_e^{-1}(\mathbf{k}j) = c\pi\epsilon^2\omega^3g(\omega^2)|e(\kappa_1)|_j^k|^4, \quad (7)$$

where $g(\omega^2(\mathbf{k}j))$ is the density of states of the considered branch.

We use the completeness property of the polarization vectors and also the fact that in the long wavelength limit ($\mathbf{k} \rightarrow 0$) we can write for the optical and acoustical oscillations, respectively, of the two-atom crystal:

$$\begin{aligned} m_1^{1/2}e_T(\kappa_1|_j^0) &= -m_2^{1/2}e_T(\kappa_2|_j^0), \\ m_2^{1/2}e_T(\kappa_1|_j^0) &= m_1^{1/2}e_T(\kappa_2|_j^0). \end{aligned}$$

In this case,

$$\tau_{eo}^{-1} \approx c\pi(\Delta\mu/\mu)^2\omega^3g(\omega^2), \quad (8)$$

$$\tau_{ea}^{-1} \approx c\pi(\Delta m/M)^2\omega^3g(\omega^2). \quad (9)$$

Here τ_{eo} and τ_{ea} are the relaxation times of the optical and acoustical phonons, respectively, in scattering from the impurities; $\Delta m = m_1 - m'$, $M = m_1 + m_2$; μ is the reduced mass of the pair of atoms $\Delta\mu = \mu - \mu'$.

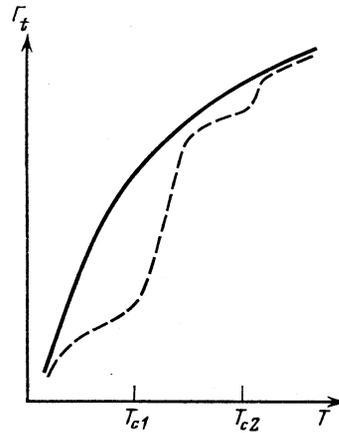


FIG. 1. Temperature dependence of the absorption coefficient of transverse acoustic waves: the solid curve corresponds to an ideal matrix, dashed curve corresponds to a complex dielectric crystal with impurities.

After uncomplicated transformations, we obtain from (1) the following for the contribution to the transverse sound absorption of the optical branch in the case in which the anharmonicity of the crystal can be neglected:

$$\Gamma_t^o = \frac{1}{4\pi^2} \frac{\hbar^2\omega_s^2}{T\rho S^3} \int_0^{k_B} k^2\gamma_s^2\omega^2n(n+1)\tau_{eo}dk. \quad (10)$$

Here k_B is the Brillouin vector. For the estimate, we shall not make a distinction between the tensors of the Grüneisen constants for the optical and acoustical phonons. Substituting (8) in (10) and carrying out the necessary calculations, in the case of high temperatures ($\hbar\omega_{1,2} < T$, where ω_1 corresponds to the frequency of the optical phonon at the center ($k = 0$) and ω_2 at the edge ($k = k_B$) of the Brillouin zone, we obtain

$$\Gamma_t^o \approx \left(\frac{\mu}{\Delta\mu}\right)^2 \frac{\omega_s^2 T \langle \gamma_s^2 \rangle}{\pi c M S^3} \frac{|\Delta\omega|}{\omega_1\omega_2},$$

where $\langle \dots \rangle$ is the usual kinetic averaging; $|\Delta\omega| = |\omega_1 - \omega_2|$. It is seen from (11) that the basic contribution to the sound absorption will be made by the low-lying optical branches with strong dispersion.

Estimates of Γ_t^o and Γ_t^a (Γ_t^a is the contribution to the absorption from a single acoustical mode¹) given for a typical crystal $\Gamma_t^o \sim (1-0.1)\Gamma_t^a$, and thus the contribution of the optical phonons to the sound absorption cannot be neglected.

As is known, in the calculation of the interaction of transverse sound only with acoustical phonons at high temperatures, the functions $\Gamma_t(T)$ have the following form: $\Gamma_t(T) \propto T, T^{2/3}, T^{5/9}$ (see Refs. 1, 2, 3, respectively). For the contribution to the absorption of transverse sound from a single optical branch, we obtained $\Gamma_t^o \propto T$ from (11). It is obvious that in crystals with a large number of atoms in the elementary cell, as a consequence of the increase in the heat capacity of the optical phonons with increase in the temperature, and also because of the temperature dependence $\tau_e(T)$ discussed above, the $\Gamma_t(T)$ dependence should be very steep.

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their experimental work before publication, and for many useful discussions. The authors are deeply grateful to V. L. Gurevich and members of the seminar directed by him for discussion of the research.

¹¹ However, if the effects of the combined phonon-impurity interaction are significant,¹⁶ then the basic assumption of the Maris theory is incorrect. The conclusion that the absorption of the longitudinal waves is relatively insensitive to the impurity content will also be incorrect. This question will be investigated in a separate paper.

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