

# Theory of the lattice absorption of sound in "dirty" crystals

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The kinetic approach is used to develop a theory of the lattice absorption of acoustic waves in "dirty" crystals, which is valid in a wide range of frequencies and which includes the published results and those obtained for pure crystals as special cases. The criteria of validity of the Maris theory of the absorption of acoustic waves in crystals with impurities are established. It is shown that, when the absorption of acoustic waves is dominated by the contribution of phonons of approximately thermal energy, the frequency dependence of the acoustic absorption coefficient of dirty crystals is quadratic at low frequencies, disappears at higher frequencies, and then becomes again quadratic and linear at the highest frequencies.

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The absorption of acoustic waves in ideal insulators is due to the lattice anharmonicity and it is described in different ways depending on the product of the acoustic wave frequency  $\omega_s$  and the reciprocal of the phonon relaxation time  $\tau^{-1}$ . The absorption of low-frequency acoustic waves characterized by  $\omega_s \tau \ll 1$  is governed by the Akhiezer mechanism and the absorption coefficient is  $\Gamma \propto \omega_s^2 \tau$ . In the opposite limit of  $\omega_s \tau \gg 1$  the absorption is dominated by three-phonon processes and the dependences of the absorption coefficient on the acoustic wave frequency and on the temperature of a crystal are governed by the polarization of the original wave, nature of the phonon constant-energy surfaces, and anisotropy of the elastic moduli. In the case of some simple crystals the expression for the absorption coefficient obtained from the theory of Landau and Rumer is  $\Gamma \propto \omega_s T^4$  ( $T$  is the temperature of the crystal).

In the case of insulator crystals with high impurity and defect concentrations the nature of the absorption may be very different from that in pure crystals. In particular, as shown in Refs. 1-6, even when the condition  $\omega_s \tau \ll 1$  is formally obeyed, the absorption in a crystal with impurities may differ considerably from the value predicted by the Akhiezer theory and the absorption coefficient may be much lower than in the case of a pure crystal.

Several authors<sup>1-3</sup> have considered the case of a crystal with a large number of impurity atoms whose role reduces to effective phonon scattering and they have assumed that the corresponding scattering cross section is typical of the Rayleigh mechanism. In view of the very rapid fall of the Rayleigh scattering cross section on increase in the phonon wavelength, the absorption is dominated by a small group of "subthermal" phonons which can be identified by the condition  $\omega_s \tau_d(q_1) = 1$  [ $\tau_d(q)$  is the relaxation time of phonons with a quasimomentum  $q$  when they are scattered by Rayleigh centers] if the anharmonicity effects can be ignored or by the condition  $\tau_d(q_2) = \tau_f(q_2)$  ( $\tau_f$  is the relaxation time in the case of phonon-phonon collisions) in the opposite case.<sup>1)</sup> Clearly, since in both cases we have  $q_{1,2} < q_T$  ( $q_T$  is a typical phonon quasimomentum), the specific heat of the absorption-active modes may be considerably less than in the case of a pure crystal and the absorption coefficient may be small compared with that for the same waves traveling in a pure crystal. Moreover,

the presence of impurities generally alters the frequency and temperature dependences of the absorption coefficient compared with the corresponding dependences in the case of a pure crystal.

In Refs. 4-6 the nature of defects is not specified (in contrast to Refs. 1-3): only the fact that the phonon scattering by defects is near-elastic is regarded as important. Moreover, Maris<sup>4</sup> assumes that the absorption of acoustic waves is still dominated by the thermal phonon group, exactly as in the case of a pure crystal. For this reason the results of Refs. 1-3 cannot be obtained from those of Maris<sup>4</sup> and in fact these two groups of investigations deal with two different special cases of the absorption of acoustic waves in dirty crystals. This shortcoming of the treatment given by Maris<sup>4</sup> was eliminated by Logachev,<sup>5,6</sup> who retained the dependence of the phonon relaxation time on the quasimomentum and showed that under appropriate conditions and neglecting the effects associated with modulation of the phonon temperature (ignored in Refs. 1 and 2) it is possible to reproduce results of Refs. 1 and 2. In this connection it should be pointed out that the representation of the phonon-impurity collision integral used in Refs. 4-6 is based on the introduction of a temperature  $T(\omega)$  for each phonon group associated with a fixed constant-energy surface defined by  $\hbar\omega = \text{const}$ . We shall show that this representation is valid even in the case of an extremely dirty crystal ( $\omega_s \tau_e \rightarrow 0$ , where  $\tau_e$  is the relaxation time of phonons in the case of quasielastic scattering mechanisms). For this reason the results of Refs. 4-6 are of limited validity.

Our aim will be to develop a theory of the absorption of sound in real insulators covering the widest possible range of frequencies and including the main results of Refs. 1-6 as well as the results of the Akhiezer and Landau-Rumer theories for pure crystals as special cases. In contrast to Refs. 4-6, we shall use the explicit form of the phonon-impurity collision integral. This will make it possible to determine, in particular, the criteria of validity of the results obtained in Refs. 4-6 and to consider a much wider range of frequencies where the results of Refs. 1-6 are invalid.

Let us assume that an acoustic wave travels in an insulator crystal containing a large number of impurities. We shall describe the phonon system of a crystal per-

turbed by sound with the aid of the Boltzmann kinetic equation. As shown in Ref. 7 (see also Ref. 8), the absorption of acoustic waves in insulators can be considered within the framework of the kinetic approach for any values of the parameter  $\omega_s \tau$ . The only criterion of validity of this approach is the inequality  $\hbar \omega_s \ll k_B T$ . In the range of acoustic waves and crystal temperatures under consideration here this inequality is satisfied by a large margin.

The absorption coefficient of an acoustic wave considered in the linear approximation with respect to the sound amplitude can be expressed in terms of a sound-induced nonequilibrium correction to the phonon distribution function<sup>7</sup>:

$$\Gamma = \frac{\hbar e_\alpha(\mathbf{q}J) q_\beta}{2\rho V \omega_s v_s(\mathbf{q}J)} \sum_{\mathbf{k}j} \operatorname{Re} \left\{ \frac{\Delta N(\mathbf{k}j)}{u_0} \right\} \omega(\mathbf{k}j) \gamma^{\alpha\beta}(\mathbf{k}j). \quad (1)$$

Here  $\mathbf{q}$  and  $\omega_s$  are the wave vector and frequency of the incident acoustic wave;  $J$  is the wave polarization;  $e_\alpha(\mathbf{q}J)$  are the components of a unit polarization vector;  $\rho$  and  $V$  are the density and volume of the investigated crystal;  $v_s(\mathbf{q}J)$  is the initial wave velocity;  $u_0$  is the wave amplitude;  $\gamma^{\alpha\beta}(\mathbf{k}j)$  is the tensor of the Grüneisen constants for phonons of polarization  $j$  and quasimomentum  $\mathbf{k}$ ;  $\hbar \omega(\mathbf{k}j)$  is the energy of a  $(\mathbf{k}, j)$  phonon;  $\Delta N(\mathbf{k}j)$  is the nonequilibrium correction to the phonon distribution function due to the passage of an acoustic wave.

The problem thus reduces to finding the correction  $\Delta N(\mathbf{k}j)$ . This quantity can be obtained by solving the Boltzmann kinetic equation

$$\frac{\partial N}{\partial t} + \frac{\partial N}{\partial r_i} \frac{\partial H}{\partial k_i} - \frac{\partial N}{\partial k_i} \frac{\partial H}{\partial r_i} = I_{ff}\{N\} + I_{fd}\{N\}, \quad (2)$$

$$H = H_0 [1 + \gamma^{\alpha\beta}(\mathbf{k}j) u_{\alpha\beta}(\mathbf{r}t)].$$

Here,  $N$  is the unknown phonon distribution function;  $H_0$  is the Hamiltonian for a  $(\mathbf{k}, j)$  mode in the unperturbed crystal;  $u_{\alpha\beta}(\mathbf{r}t)$  is the strain vector for the region affected by the wave;  $I_{ff}\{N\}$  is the phonon-phonon collision integral;  $I_{fd}\{N\}$  is the collision integral representing the scattering of phonons by impurities and crystal defects.

We shall select the phonon-phonon collision integral in its standard form<sup>9</sup> and, for simplicity, we shall consider only the  $\tau_U \ll \tau_N$  case ( $\tau_U$  and  $\tau_N$  are the characteristic times for the umklapp and normal processes, respectively):

$$I_{ff}\{N\} = -\frac{\hbar \omega(\mathbf{k}j)}{\tau_i} \frac{1}{k_B T} n(\mathbf{k}j) [n(\mathbf{k}j) + 1] \left[ i q u_\alpha \gamma_\alpha(\mathbf{k}j) - \frac{\Delta T}{T} \right] - \frac{\Delta N}{\tau_j}, \quad (3)$$

where  $n(\mathbf{k}, j)$  is the equilibrium Planck phonon distribution function,

$$\gamma_\alpha(\mathbf{k}j) = \gamma^{\alpha\beta}(\mathbf{k}j) e_\alpha(\mathbf{q}J) q_\beta q^{-1},$$

and  $\Delta T$  is the correction to the crystal temperature, which is linear in respect of the acoustic wave amplitude. The question under what conditions we can introduce a local phonon temperature will be discussed below.

In contrast to Refs. 4–6, the integral describing collisions of phonons with impurities will be used in its

explicit form

$$I_{fd}\{N\} = \sum_{\mathbf{k}'j'} W(\mathbf{k}j; \mathbf{k}'j') [N(\mathbf{k}j) - N(\mathbf{k}'j')] \delta[\omega(\mathbf{k}j) - \omega(\mathbf{k}'j')], \quad (4)$$

but without specifying the scattering mechanism. The expression (4) describes in a sufficiently general form the quasielastic phonon scattering mechanisms which may involve the scattering by impurities, vacancies, dislocations, crystal inhomogeneities, etc.

For the sake of simplicity, we shall assume that the wave polarization is not affected by the scattering on impurities or defects. Clearly, this assumption cannot alter the results qualitatively. We shall find a solution of the kinetic Eq. (2) in the form

$$N(\mathbf{k}j) = n(\mathbf{k}j) + \Delta N(\mathbf{k}j) \exp [i(\mathbf{q}\mathbf{r} - \omega_s t)]. \quad (5)$$

Again for simplicity, we shall consider an isotropic case on the assumption that a crystal is elastically isotropic and, moreover, that  $\tau(\mathbf{k}j)$  and  $\gamma_s(\mathbf{k}j)$  depend only on the modulus of  $\mathbf{k}$ . The problem of absorption of transverse sound in cubic crystals requires a special study. In fact, in the case of cubic crystals the tensor  $\gamma^{\alpha\beta}(\omega) = \langle\langle \gamma^{\alpha\beta}(\mathbf{k}j) \rangle\rangle$  is diagonal. Here,

$$\langle\langle \dots \rangle\rangle = \sum_{\mathbf{k}j} (\dots) \delta[\omega - \omega(\mathbf{k}j)] / \sum_{\mathbf{k}j} \delta[\omega - \omega(\mathbf{k}j)].$$

In the case of shear waves traveling along high-symmetry directions we have  $\gamma_s(\omega) = \langle\langle \gamma_s(\mathbf{k}j) \rangle\rangle = 0$ . We shall return to this case later.

We shall transform the collision integral  $I_{fd}\{N\}$  to (see Ref. 10)

$$I_{fd}\{N\} = -\frac{1}{\tau_0} \left\{ \Delta N(\mathbf{k}j) - \frac{\int d\theta' \sin \theta' d\varphi' S(\theta\varphi; \theta'\varphi') \Delta N(\mathbf{k}j, \theta')}{\int d\theta' \sin \theta' d\varphi' S(\theta\varphi; \theta'\varphi')} \right\}, \quad (6)$$

$$\tau_0^{-1} = \sum_{\mathbf{k}'j'} W(\mathbf{k}j; \mathbf{k}'j') \delta[\omega(\mathbf{k}j) - \omega(\mathbf{k}'j')].$$

Here,  $\tau_0^{-1}$  is the reciprocal of the relaxation time for the scattering of phonons by impurities;  $S(\theta\varphi; \theta'\varphi')$  is the cross section for the scattering of a phonon corresponding to a constant-energy surface with a quasimomentum  $k$  from a state  $(\theta\varphi)$  to a state  $(\theta'\varphi')$ . The system (6) is derived bearing in mind that in the isotropic case the distribution function  $\Delta N(\mathbf{k}j)$  depends only on the cosine of the angle  $\theta$  between the vectors  $\mathbf{k}$  and  $\mathbf{q}$ .<sup>2)</sup>

It is more convenient to introduce a new as yet unknown distribution function  $F_{kj}(z)$  related to the function  $\Delta N(\mathbf{k}j)$  by

$$\Delta N(\mathbf{k}j) = \frac{1}{k_B T} n(\mathbf{k}j) [n(\mathbf{k}j) + 1] [-i q u_\alpha \hbar \omega(\mathbf{k}j) \gamma_\alpha(\mathbf{k}j) + F_{kj}(z)], \quad z = \cos \theta. \quad (7)$$

The kinetic Eq. (2) modified by Eqs. (3)–(7) becomes

$$\left( -i\omega_s + i q v_s(\mathbf{k}j) + \frac{1}{\tau_{j0}} \right) F_{kj}(z) = \frac{1}{2\tau_0} \sum_{n=0}^{\infty} \frac{\bar{S}_n}{S_0} P_n(z) \int_{-1}^1 d\zeta P_n(\zeta) F_{kj}(\zeta) + q u_\alpha \omega_s \hbar \omega(\mathbf{k}j) \gamma_\alpha(\mathbf{k}j) + \frac{\Delta T}{T} \frac{\hbar \omega(\mathbf{k}j)}{\tau_j}. \quad (8)$$

Here,  $\tau_{j0}^{-1} \equiv \tau_j^{-1} + \tau_0^{-1}$ ;  $P_n(\zeta)$  is a Legendre polynomial of order  $n$ ;  $\bar{S}_n$  is the  $n$ -th coefficient in the series expansion of the quantity  $\bar{S}(\cos \vartheta) = S(\theta\varphi; \theta'\varphi')$ ; in terms of Legendre polynomials;  $\vartheta$  is the angle between the vec-

tors  $\mathbf{k}$  and  $\mathbf{k}'$ .

We shall assume that the dispersion law of phonons is linear, i. e., that  $\omega(kj) = v_s(kj)/k$ . We shall introduce the quantity

$$w_{j0} = \left(1 + \frac{i}{\omega_s \tau_{j0}}\right) \frac{1}{\alpha_{j1}},$$

where  $\alpha_{j1} = v_s(j)/v_s(j)$ . The final form of Eq. (8) is then

$$F_{kj}(z) = \lambda \int_{-1}^1 d\xi K(z\xi) F_{kj}(\xi) + M_{kj}(z). \quad (9)$$

The following notation is used above:

$$\lambda = \frac{1}{2i\omega_s \tau_{j0} \alpha_{j1}}, \quad K(z\xi) = - \sum_{n=0}^{\infty} \frac{S_n P_n(z) P_n(\xi)}{w_{j0} - z},$$

$$M_{kj}(z) = \frac{\hbar\omega(kj)}{i\omega_s \tau_{j0} \alpha_{j1}} \frac{1}{w_{j0} - z} + \frac{i q u_s \hbar\omega(kj) \gamma_s(kj)}{\alpha_{j1}} \frac{1}{w_{j0} - z}.$$

Equation (9) for the unknown distribution function  $F_{kj}(z)$  is a linear Fredholm integral equation with an infinite degenerate  $K(z\xi)$ . In order to find the required absorption coefficient, we shall need the quantity

$$F_{kj}^{(0)} = \frac{1}{2} \int_{-1}^1 d\xi F_{kj}(\xi).$$

The procedure for finding a solution of Eq. (9) for the quantity  $F_{kj}^{(0)}$  is fully analogous to the corresponding procedure in the problem of electron absorption of acoustic waves in semiconductors at "intermediate" frequencies.<sup>10</sup> The final expression for  $F_{kj}^{(0)}$  is

$$F_{kj}^{(0)} = \hbar\omega(kj) \left[ - \frac{1}{i\omega_s \tau_j} \frac{\Delta T}{T} + i q u_s \gamma_s(kj) \right] \frac{i\omega_s \tau_0}{1 - i\omega_s \tau_0 \alpha_{j1} B}, \quad (10)$$

where

$$B = -w_{j0} - \frac{1^2}{-3w_{j1} - \frac{2^2}{-5w_{j2} - \frac{3^2}{-7w_{j3} - \dots}}}. \quad (11)$$

As in the problem of electron absorption of acoustic waves in semiconductors at intermediate frequencies, the answer in the present case can also be expressed in terms of a continued fraction  $B$  with a structure in many respects similar to that of the fraction studied earlier. The quantities  $w_{jn}$  occurring in Eq. (11) are described by

$$w_{jn} = \alpha_{j1}^{-1} (1 + i/\omega_s \tau_{jn}), \quad \tau_{jn}^{-1} = \tau_j^{-1} + \tau_n^{-1},$$

and  $\{\tau_n\}$  is a sequence of relaxation times of harmonics of the perturbed phonon distribution function

$$\frac{1}{\tau_n} = \frac{1}{\tau_0} \left(1 - \frac{S_n}{S_0} \frac{1}{2n+1}\right), \quad n=1, 2, 3, \dots$$

We now have to determine  $\Delta T/T$ . Obviously collisions do not alter the total energy of the phonon system, i. e.,

$$\sum_{kj} \hbar\omega(kj) I\{N(kj)\} = 0. \quad (12)$$

Since collisions of phonons with impurities and defects are assumed to be quasielastic, we can rewrite Eq. (12) in the form

$$\sum_{kj} \hbar\omega(kj) I_{II}\{N(kj)\} = 0. \quad (13)$$

Using now Eqs. (3), (5), (7), (10), and (13), we obtain

$$\frac{\Delta T}{T} = i q u_0 \frac{\langle \gamma_s(kj) \tau_j^{-1}(kj) i\omega_s \tau_0(kj) [1 - i\omega_s \tau_0(kj) \alpha_{j1} B(kj)]^{-1} \rangle}{\langle \tau_j^{-1}(kj) \{1 + \tau_0(kj) \tau_j^{-1}(kj) [1 - i\omega_s \tau_0(kj) \alpha_{j1} B(kj)]^{-1}\} \rangle}. \quad (14)$$

The kinetic averaging  $\langle \dots \rangle$  means

$$\langle \dots \rangle = \sum_{kj} \hbar^2 \omega^2(kj) n(kj) [n(kj) + 1] (\dots) / \sum_{kj} \hbar^2 \omega^2(kj) n(kj) [n(kj) + 1].$$

We shall introduce

$$P(kj) = \frac{i\omega_s \tau_0(kj)}{1 - i\omega_s \tau_0(kj) \alpha_{j1} B(kj)}. \quad (15)$$

Using Eqs. (1), (10), (14), and (15), we finally obtain the following expression for the "lattice" acoustic absorption coefficient:

$$\Gamma = \frac{1}{2} \frac{C_v T \omega_s}{\rho v_s^3 (qJ)} \text{Re} \left\{ i \langle \gamma_s^2(kj) P(kj) \rangle - \frac{1}{\omega_s} \frac{\langle \gamma_s(kj) \tau_j^{-1}(kj) P(kj) \rangle^2}{\langle \tau_j^{-1}(kj) [1 + P(kj) / i\omega_s \tau_j(kj)] \rangle} \right\}, \quad (16)$$

where  $C_v$  is the specific heat of a crystal.

We shall now consider a number of special cases.

1. In the case of a pure crystal ( $\tau_0 \gg \tau_j$ ), we have

$$w_{jn} \rightarrow w_j = \alpha_{j1}^{-1} (1 + i/\omega_s \tau_j),$$

and the continued fraction in Eq. (15) is easily summed<sup>11</sup>:

$$P \rightarrow \frac{1}{2\alpha_{j1}} \ln \frac{w_j + 1}{w_j - 1}. \quad (17)$$

Using Eq. (17), we can easily demonstrate that Eq. (16) for the absorption coefficient gives the correct result for the absorption of both low-frequency ( $\omega_s \tau \ll 1$ , the Akhiezer case) and high-frequency ( $\omega_s \tau \gg 1$ , the Landau-Rumer case) acoustic waves in pure insulators.

2. We shall now assume that  $\tau_0 \ll \tau_j$ ; moreover, we shall postulate that  $\omega_s \tau_0 \ll 1$  (strong scattering by impurities). Clearly, the  $\omega_s \tau_0 \gg 1$  case is of no real interest because the influence of impurities on the absorption of acoustic waves in this limit, considered within the framework of the adopted approximations, is solely due to the changes in the elastic moduli and in the density of a crystal. We shall begin by postulating that  $\omega_s \tau_j \gg 1$ . We shall then consider two limiting cases: a)  $\omega_s^2 \tau_0 \tau_j \ll 1$ ; b)  $\omega_s^2 \tau_0 \tau_j \gg 1$ .

a. Let  $\omega_s^2 \tau_0 \tau_j \ll 1$ . Using Eq. (11), we obtain

$$B = -1 - i/\omega_s \tau_{j0} - i/\omega_s \tau_j, \quad (18)$$

$$P = (\omega_s^2 \tau_j^2 - i\omega_s \tau_j) / (1 + \omega_s^2 \tau_j^2).$$

Assuming, for simplicity, that  $\tau_j$  is independent of  $(kj)$ , we obtain

$$\Gamma = \frac{1}{2} \frac{C_v T}{\rho v_s^3 (qJ)} \frac{\omega_s^2 \tau_j}{1 + \omega_s^2 \tau_j^2} (\langle \gamma_s^2 \rangle - \langle \gamma_s \rangle^2), \quad (19)$$

which is identical with the result obtained by Maris<sup>4</sup> (see also Refs. 5 and 6).

We can see that the criterion of validity of the Maris theory is  $\omega_s^2 \tau_0 \tau_j \ll 1$ . When this condition is satisfied there is a local coupling between the response of the phonon system and a perturbation  $[v_s(\tau_0 \tau_j)^{1/2}]$  is the characteristic length in which the phonon energy changes significantly]. Such local coupling clearly occurs if this

length is considerably less than the wavelength of sound. Then at each point in a crystal we can introduce a temperature for each group of phonons associated with a specific constant-energy surface and to use the phonon-impurity collision integral in the form

$$I_{i\alpha}(N) = -\frac{1}{\tau_s} [N(\mathbf{k}j) - N_s(\mathbf{k}j)],$$

$$N_s(\mathbf{k}j) = \left[ \exp \frac{\hbar\omega(\mathbf{k}j; \tau t)}{k_B T(\omega(\mathbf{k}j; \tau t))} - 1 \right]^{-1}.$$

b. In the opposite limiting case of  $\omega_s^2 \tau_0 \tau_f \gg 1$  (although we still have  $\omega_s \tau_0 \ll 1$ ) the results of the theories of Refs. 4–6 are invalid. Under these conditions we have

$$P \approx 1 - 1/3 i \omega_s \tau_1, \quad (20)$$

$$\Gamma = \frac{1}{2} \frac{C_s T}{\rho v_s^3 (qJ)} \omega_s^2 \tau_1 \langle \gamma_s^2 \rangle. \quad (21)$$

It should be pointed out that the concept of a local phonon temperature can be introduced only if  $\omega_s^2 \tau_0 \tau_f \ll 1$ . In the opposite limiting case this concept clearly loses its meaning. Moreover, using Eqs. (18) and (20), we can easily show that  $\Delta T/T$  found from Eq. (14) does not tend to zero on increase in the parameter  $\omega_s^2 \tau_0 \tau_f$ . However, it should be pointed out that if  $\omega_s^2 \tau_0 \tau_f \gg 1$ , the contribution of the term in the nonequilibrium phonon distribution function due to modulation of the phonon temperature [even when  $\Delta T/T$  is given by Eq. (14)] to the rate of entropy generation it is small, when judged on the basis of the parameter  $(\omega_s^2 \tau_0 \tau_f)^{-1} \ll 1$ , compared with the contribution of the term due to modulation of the energy  $\Delta\omega/\omega \propto \gamma_s^2 \mu_{\alpha\beta}$  of phonons in the field of an acoustic wave. In fact, the contributions of both terms to  $\Gamma$  are represented respectively by the second and first terms in the braces in Eq. (16). Using Eqs. (18) and (20), we can easily show that the second term in Eq. (16) is small compared with the first when judged on the basis of the parameter  $(\omega_s^2 \tau_0 \tau_f)^{-1} \ll 1$ . For this reason the expression (16) for the absorption coefficient of an acoustic wave is valid throughout the range of frequencies in which we can use the kinetic approach, with the exception of the direct vicinity of the frequency  $\omega_s = (\tau_0 \tau_f)^{-1/2}$  where Eq. (16) can be regarded simply as an interpolation. In the case of a pure insulator a similar approach (see Refs. 7 and 8) clearly loses its validity in the vicinity of the frequency  $\omega_s = \tau_f^{-1}$ .

We can thus see that the frequency dependence of  $\Gamma$  is quadratic at frequencies  $\omega_s < \tau_f^{-1}$ , is absent in the range  $\tau_f^{-1} < \omega_s < (\tau_0 \tau_f)^{-1/2}$ , again becomes quadratic for  $(\tau_0 \tau_f)^{-1/2} < \omega_s < \tau_0^{-1}$  and changes to linear when  $\omega_s > \tau_0^{-1}$ .

If we ignore small changes in the velocity of sound, in the elastic moduli, and in the phonon-phonon relaxation times of a dirty crystal compared with a pure one, we find that at a fixed temperature the frequency dependence of the absorption coefficient of an acoustic wave has the form shown in Fig. 1. For comparison, Fig. 1 includes the corresponding dependence for a pure crystal (dashed curve).

Therefore, in the range of sufficiently low temperatures  $\tau_0 \ll \tau_f$ , the acoustic absorption coefficient for a nonideal crystal considered at frequencies  $\tau_f^{-1} < \omega_s < \tau_0^{-1}$  is much smaller (for the same waves and at the

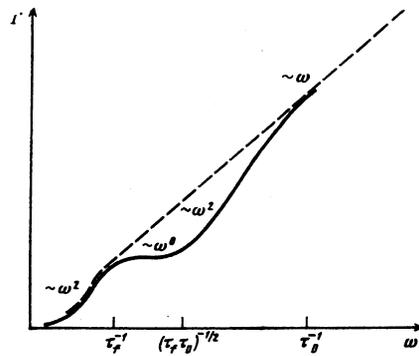


FIG. 1.

same temperature) than the absorption coefficient of a pure crystal. In the case of two crystals with identical times  $\tau_f$  but with different values of  $\tau_0$ , this reduction in the absorption coefficient is more marked for a crystal with a small value of the ratio  $\tau_0/\tau_f$ . Under experimental conditions this situation may be realized in the case of solid solutions with different stoichiometric compositions. Then clearly the ratio in question has a minimum for a specific stoichiometric composition. It follows that the composition dependence of the absorption coefficient of an acoustic wave may be represented qualitatively by a bell-shaped curve with a minimum (naturally, if for at least one solid-solution composition the value of  $\tau_0$  becomes less than  $\tau_f$ ).

3. We shall now consider the absorption of acoustic waves in insulator crystals containing Rayleigh scattering centers. As before, we shall assume that  $\omega_s \tau_0(T) \ll 1$  [ $\tau_0(T)$  is the relaxation time of phonons of energy of the order of  $k_B T$  scattered by impurities] and, moreover, we shall postulate that  $\tau_0(q) \propto q^{-4}$ . Using Eqs. (18) and (20) for the  $q > q_1$  case [ $q_1$  is found from the condition  $\omega_s \tau_0(q_1) = 1$ ] and Eq. (17) for  $q < q_1$  [ $w_f$  in Eq. (17) should then be replaced with  $w_0 = 1 + i/\omega_s \tau_0$ ], we can easily show that—as in Refs. 1 and 2—the acoustic wave absorption is dominated by phonons with quasimomenta close to  $q_1$ . This result is valid if the role of the anharmonicity is small, i. e., if  $\tau_f(q_1) \gg \tau_0(q_1)$ . It should be noted that if  $\tau_f(q_1) \gg \tau_0(q_1)$ , then

$$\omega_s^2 \tau_f(q_1) \tau_0(q_1) \gg 1$$

and modulation of the phonon temperature in the acoustic wave field may be ignored. However, if the anharmonicity effects are important, i. e., if  $\tau_f(q_1) \ll \tau_0(q_1)$ , we find that

$$\omega_s^2 \tau_0(q_1) \tau_f(q_1) \ll 1$$

[and this applies even more strongly when  $\omega_s^2 \tau_0(q_2) \tau_f(q_2) \ll 1$ , where  $q_2$  is found from the condition  $\tau_0(q_2) = \tau_f(q_2)$ —see Refs. 2 and 3] and modulation of the temperature of phonons with quasimomenta in the range  $q \gtrsim q_1$  can no longer be ignored. Under these conditions we find from Eqs. (16) and (18) that the absorption is dominated by phonons with quasimomenta of the order of  $q_T$  and the absorption coefficient is still given by Eq. (19). This was first pointed out by Logachev<sup>5,6</sup> for the specific case of  $\omega_s \tau_f \ll 1$ . If  $\omega_s \tau_f \ll 1$ , this means that in con-

trast to the results of Refs. 2 and 3, impurities have practically no influence on the acoustic wave absorption. We shall show below that the results of Refs. 2 and 3 apply to the absorption of shear waves in high-symmetry crystals.

4. We shall now consider briefly the problem of the absorption of a shear acoustic wave traveling along a high-symmetry axis in a cubic crystal. As shown above, under these conditions we have  $\gamma_s(\omega) = 0$ ; the assumption that  $\gamma_s$  depends only on the modulus of  $\mathbf{k}$  is unjustified and for this reason Eq. (16) for the absorption coefficient of an acoustic wave is invalid.

We shall assume that a shear wave travels in a cubic crystal along the [001] axis. In the case of a wave polarized along the [100] axis the quantity  $\gamma_s(\mathbf{k}j)$  considered in the elastic continuum approximation is given by

$$\gamma_s(\mathbf{k}j) = \frac{1}{2c_{11}} (c_{44} + c_{155}) \sin 2\bar{\theta} \cos \bar{\varphi},$$

where  $\bar{\theta}$  and  $\bar{\varphi}$  are the angles which define a unit vector in the direction  $\mathbf{k}$ ;  $c_{11}$ ,  $c_{44}$ , and  $c_{155}$  are second- and third-order elastic moduli (a crystal is assumed to be elastically isotropic).<sup>12</sup>

The kinetic equation for the function  $F_{kj}(\bar{\theta}, \bar{\varphi})$  given by Eq. (7) (where we must retain the dependence of  $F_{kj}$  on  $\bar{\theta}$  and  $\bar{\varphi}$ ) is

$$F_{kj}(\bar{\theta}\bar{\varphi}) = \lambda \sum_{n=0}^{\infty} S_n \int_0^{\pi} d\bar{\theta}' \sin \bar{\theta}' \int_0^{2\pi} d\bar{\varphi}' P_n(\cos \bar{\theta}) F_{kj}(\bar{\theta}'\bar{\varphi}') + \frac{iq u_0 \hbar \omega(kj)}{\alpha_{j1}(\omega_{j0} - z)} \frac{c_{44} + c_{155}}{c_{11}} \sin \bar{\theta} \cos \bar{\theta} \cos \bar{\varphi} - \frac{\Delta T}{T} \frac{\hbar \omega(kj)}{i\omega_s \tau_j \alpha_{j1} \omega_{j0} - z} \frac{1}{\omega_{j0} - z}. \quad (22)$$

We shall show that under these conditions we have  $\Delta T/T = 0$ . In fact, in order to determine  $\Delta T/T$ , we find from Eq. (13) that we need to know the quantity

$$\frac{1}{4\pi} \int_0^{\pi} d\bar{\theta} \sin \bar{\theta} \int_0^{2\pi} d\bar{\varphi} F_{kj}(\bar{\theta}\bar{\varphi}).$$

Integrating the left- and right-hand parts of Eq. (22) with respect to  $d\bar{\varphi}$  from 0 to  $2\pi$ , and introducing the function

$$f_{kj}(\bar{\theta}) = \frac{1}{2\pi} \int_0^{2\pi} d\bar{\varphi} F_{kj}(\bar{\theta}\bar{\varphi}),$$

we obtain

$$f_{kj}(z) = \lambda \int_{-1}^1 d\zeta K(z\zeta) f_{kj}(\zeta) - \frac{\Delta T}{T} \frac{\hbar \omega(kj)}{i\omega_s \tau_j \alpha_{j1} \omega_{j0} - z} \frac{1}{\omega_{j0} - z}.$$

Using Eq. (10), where the second term in the brackets should be assumed to vanish, and Eq. (13), we can easily show that  $\Delta T/T = 0$ , i. e., that in this case there is no modulation of the phonon temperature. We can also show easily that a solution of Eq. (22) is of the form

$$F_{kj}(\bar{\theta}\bar{\varphi}) = F_{kj}(z) \cos \bar{\varphi}.$$

In fact, assuming that  $\Delta T/T = 0$  and transforming the expression for  $P_n(\cos \bar{\theta})$  in accordance with the addition theorem for spherical harmonics, we find that

$$F_{kj}(\bar{\theta}\bar{\varphi}) = F_{kj}(z) \cos \bar{\varphi},$$

where  $\bar{F}_{kj}(z)$  is found by solving the following kinetic

equation:

$$F_{kj}(z) = \lambda \sum_{n=0}^{\infty} \sum_{m=1}^n \frac{(n-m)!}{(n+m)!} \frac{S_n}{\omega_{j0} - z} \int_{-1}^1 d\zeta P_n^m(z) P_n^m(\zeta) F_{kj}(\zeta) - \frac{iq u_0 \hbar \omega(kj)}{3\alpha_{j1}} \frac{c_{44} + c_{155}}{c_{11}} \frac{1}{\omega_{j0} - z} P_2^1(z), \quad (23)$$

where  $P_n^m(z)$  are associated Legendre functions.

It should be noted that because of  $\Delta T/T = 0$ , the expression for  $\Gamma$  found by solving Eq. (23) applies throughout the range of validity of the kinetic approach when  $\hbar \omega_s \ll k_B T$ . Like the kinetic equation (9), Eq. (23) describes a strongly anisotropic phonon distribution function for the  $\omega_s \tau_0 \approx 1$  case and this function cannot generally be represented in the frequency range under discussion by a finite number of harmonics (relaxation of each harmonic is characterized by a different time constant and these time constants may differ considerably from one another: in particular,  $\tau_0$  and  $\tau_1$  in the sequence  $\{\tau_n\}$  may differ by several orders of magnitude if a small-angle phonon scattering predominates).

Equation (23) can be solved exactly if: a) the cross section for the scattering of phonons by impurities  $\bar{S}(\cos \bar{\theta})$  is isotropic, i. e., if it is independent of  $\bar{\theta}$ ; b) if  $\bar{S}(\cos \bar{\theta})$  can be represented by an expansion in terms of the first two Legendre polynomials, i. e., if

$$\bar{S}(\cos \bar{\theta}) = \bar{S}_0 + \bar{S}_1 \cos \bar{\theta}.$$

We shall not analyze the behavior of these solutions in the region  $\omega_s \tau_0 \approx 1$  when the anisotropy of the phonon distribution function mentioned above is important. We shall simply note that in all cases when  $\omega_s \tau_0 \ll 1$  ( $\tau_0 \ll \tau_j$ ) the absorption coefficient of an acoustic wave is described by a formula similar to the formula obtained by Logachev<sup>5</sup> which in the case of insulators with large numbers of Rayleigh scattering centers gives the results similar to those obtained in Refs. 1 and 2. In particular, if the anharmonicity is significant, the phonon group dominating the absorption is the one which obeys the condition  $\tau_j(q_2) = \tau_0(q_2)$ . The identity of the results obtained by both approaches is to be expected because in this case we have  $\Delta T/T = 0$ .

We point out finally that in crystals with complex unit cells the presence of optical branches (particularly low lying branches of optical phonons) may alter considerably the results of Refs. 1-3. In particular, in the temperature range where optical phonons make a considerable contribution to the absorption of sound, the absorption is dominated (exactly as in a pure crystal) by the thermal phonon groups because the cross sections for the scattering of optical phonons on impurities do not decrease very rapidly on increase in the optical phonon wavelength.

We conclude by considering briefly the reasons for the appearance of the above-mentioned singularities in the frequency dependence of the absorption coefficient of an acoustic wave. As pointed out above, the contribution to the dissipative function is made by modulation of both the energy and temperature of phonons in the field of an acoustic wave.

If there is no modulation of the phonon temperature

(see Sec. 4), relaxation of the acoustically perturbed phonon distribution function to a state of equilibrium in dirty crystals occurs because of collisions of phonons with impurities and also because of diffusion. The absorption coefficient in the  $\omega_s \tau_0 < 1$  case is then clearly described by the Akheizer formula, i. e., by  $\Gamma \sim \omega_s^2 \tau_0$ . The impurities therefore have a considerable influence on the absorption of transverse sound in high-symmetry crystals. The anomalies of the frequency and temperature dependences of  $\Gamma$  obtained for crystals with Rayleigh scattering centers are discussed in detail in Refs. 1-3 and are considered above.

If  $\Delta T/T \neq 0$ , the rate of entropy generation in a system depends strongly on the ratio of the characteristic lengths which are the acoustic wavelength and the lengths describing the relaxation of phonon energy directly because of phonon-phonon collisions and also because of diffusion. If  $\omega_s^2 \tau_f \tau_0 \gg 1$ , then—as pointed out above—the contribution of the term due to modulation of the phonon temperature is small and, as in the  $\Delta T/T = 0$  case, relaxation of the nonequilibrium correction to the phonon distribution function is due to phonon-impurity collisions and diffusion. These collisions suppress a possible anisotropy of the quasimomentum distribution and the diffusion processes are responsible for the equalization of the energy (and of the phonon occupation number) modulated by the wave field. This is the physical cause for the Akheizer dependence of the absorption coefficient of sound in dirty crystals at frequencies  $(\tau_0 \tau_f)^{-1/2} < \omega_s < \tau_0^{-1}$ .

At lower frequencies characterized by  $\omega_s^2 \tau_0 \tau_f \ll 1$  the relaxation of the nonequilibrium correction to the phonon energy distribution function is due to direct phonon-phonon collisions and diffusion processes. Both these processes are important throughout the frequency range  $\omega_s^2 \tau_0 \tau_f \ll 1$ . At the lowest frequencies corresponding to  $\omega_s < \tau_f^{-1}$  the phonon-phonon processes predominate (their contribution  $\tau_f / \tau_0 \gg 1$  times greater than that of the diffusion processes). For this reason the influence of impurities on the absorption of sound is slight under the conditions discussed here. At higher frequencies  $\tau_f^{-1} < \omega_s < (\tau_0 \tau_f)^{-1/2}$  the temporal dispersion makes the phonon-phonon collisions ineffective and the diffusion plays the dominant role, resulting in the restoration of the Akheizer frequency dependence of the absorption coefficient of sound in the range  $\omega_s > (\tau_f \tau_0)^{-1/2}$ . This is the reason for the predicted weakening of the frequency dependence of the absorption coefficient of an acoustic wave in the intermediate range  $\tau_f^{-1} < \omega_s < (\tau_0 \tau_f)^{-1/2}$ .

It should be pointed out that a similar frequency dependence of the absorption coefficient of sound in insulators was obtained by Gurevich and Éfros<sup>13</sup> under conditions such that the characteristic time of the normal processes is short compared with the characteristic time of the umklapp processes:  $\tau_N \ll \tau_U$  (the characteristic time  $\tau_U$  is understood in Ref. 13 to be any time describing the loss of the phonon quasimomentum). In

the case under discussion here we have  $\tau_N \gg \tau_U$  and the phonon-impurity collision time is  $\tau_0 \ll \tau_f$ , so that the physical picture of the absorption is very different. The similarity of the frequency dependences of the absorption coefficient of sound in both cases is therefore related to the generality of the acoustic absorption coefficients<sup>13</sup> although the systems under discussion have long relaxation times (in our case, the energy relaxation time is  $\tau_f$ ).

We shall finally point out that in the frequency range  $\tau_f^{-1} < \omega_s < (\tau_0 \tau_f)^{-1/2}$  where the absorption coefficient is independent of the frequency of an incident acoustic wave, its temperature dependence becomes very strong:  $\Gamma \sim T^4 \tau_f^{-1}(T)$ . Clearly, the size effect in the absorption of hypersound in insulators<sup>14</sup> is due to this circumstance and due to the fact that we can reduce significantly the value of  $\tau_0$  by restricting the transverse dimensions of a sample even in the case of relatively "pure" samples.

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- <sup>1</sup>As shown below, the condition  $\tau_f(q_2) = \tau_d(q_2)$  is important only in the case of transverse waves.
- <sup>2</sup>It should be pointed out that the scattering of phonons by dislocations can be regarded as isotropic in the case of averaging over all the directions of dislocations and when these directions are distributed uniformly in a crystal.

- <sup>1</sup>P. B. Miller, Phys. Rev. **137**, A1937 (1965).
- <sup>2</sup>L. É. Gurevich and B. I. Shklovskii, Zh. Eksp. Teor. Fiz. **53**, 1726 (1967) [Sov. Phys. JETP **26**, 989 (1968)].
- <sup>3</sup>V. D. Kagan, Fiz. Tverd. Tela (Leningrad) **23**, 1551 (1981) [Sov. Phys. Solid State **23**, 909 (1981)].
- <sup>4</sup>H. J. Maris, Phys. Rev. **175**, 1077 (1968).
- <sup>5</sup>Yu. A. Logachev, Fiz. Tverd. Tela (Leningrad) **15**, 3454 (1973) [Sov. Phys. Solid State **15**, 2308 (1974)].
- <sup>6</sup>Yu. A. Logachev, Fiz. Tverd. Tela (Leningrad) **16**, 3109 (1974) [Sov. Phys. Solid State **16**, 2008 (1975)].
- <sup>7</sup>H. J. Maris, in: Physical Acoustics: Principles and Methods (ed. by W. P. Mason and R. N. Thurston), Vol. 8, Academic Press, New York, 1971, p. 279.
- <sup>8</sup>Yu. A. Logachev and B. Ya. Moizhes, Fiz. Tverd. Tela (Leningrad) **15**, 2888 (1973) [Sov. Phys. Solid State **15**, 1931 (1974)].
- <sup>9</sup>T. O. Woodruff and H. Ehrenreich, Phys. Rev. **123**, 1553 (1961).
- <sup>10</sup>A. G. Kozorezov and Yu. V. Gulyaev, Fiz. Tverd. Tela (Leningrad) **18**, 2048 (1976) [Sov. Phys. Solid State **18**, 1191 (1976)].
- <sup>11</sup>A. N. Khovanskii, Prilozhenie tsepykh drobeĭ i ikh obobshcheniĭ k voprosam priblizhennogo analiza (Applications of Continued Fractions and Their Generalizations to Problems in Approximate Analysis), Gostekhizdat, M., 1956.
- <sup>12</sup>K. Brugger, Phys. Rev. **137**, A1826 (1965).
- <sup>13</sup>V. L. Gurevich and A. L. Éfros, Zh. Eksp. Teor. Fiz. **51**, 1693 (1966) [Sov. Phys. JETP **24**, 1146 (1967)].
- <sup>14</sup>Yu. V. Gulyaev, S. N. Ivanov, A. G. Kozorezov, I. M. Kotelyanskii, and V. V. Medved', Pis'ma Zh. Eksp. Teor. Fiz. **31**, 447 (1980) [JETP Lett. **31**, 418 (1980)].

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