Intrinsic dielectric loss in crystals: Low temperatures

V. L. Gurevich and A. K. Tagantsev

A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR, Leningrad
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A detailed theoretical study is made of the frequency and temperature dependence of the intrinsic dielectric loss in crystals of different symmetries for temperatures much lower than the Debye temperature, where the loss is due to the interaction of acoustic phonons with the alternating electric field. This work is essentially an extension of previous theoretical studies which considered the dependence of the loss on the crystallographic system, the degeneracy of the phonon spectrum, and the presence or absence of a center of symmetry in the crystal. It is shown that the detailed behavior of the loss is sensitive to the crystal class. Results are presented and tabulated for all 32 classes. The influence of accidental degeneracy of the phonon spectrum is investigated for all the typical forms of accidental degeneracy, i.e., those which occur with a finite probability. The reasons for the discrepancy between the conclusions of this study and the results of Coombs and Cowley are discussed, and the present results are compared with the existing experimental data.

INTRODUCTION

In this paper we make a theoretical study of the intrinsic dielectric loss in ordinary crystalline dielectrics of different symmetries. We consider the case of low temperatures $T \ll \Theta$ (Θ is the Debye temperature), where the intrinsic loss is due to the interaction of the alternating electric field $\mathbf{E}$ with the acoustic phonon system.

The intrinsic loss in crystals was first studied theoretically by Vinogradov,[1] who obtained an expression for one of the contributions to the loss (the so-called "three-quantum" contribution) and found that for $T \gg \Theta$ this contribution is proportional to T. Stolen and Dransfeld[2] pointed out the existence of another loss contribution (the four-quantum contribution) and showed that for $T \gg \Theta$ it is proportional to T^2. Coombs and Cowley[3] detected still another intrinsic loss mechanism, which is characteristic of noncentrosymmetric crystals and is due to the perturbation of the phonon distribution function; this has since come to be called the quasi-Debye contribution.

In our previous papers[4–7] we showed that the frequency and temperature dependence of the three-quantum loss, like its order of magnitude, are extremely sensitive to the crystal symmetry, by which we mean not only the presence or absence of a center of symmetry in the crystal, but also the presence of symmetry axes and generally of acoustic axes which lead to degeneracy of the phonon spectrum. The importance of lines of degeneracy of the phonon spectrum in the kinetics of phonons has been pointed out by Herring.[8]

The contribution to the dielectric loss from the regions bordering lines of symmetry degeneracy has been discussed by Balagurov, Vaks, and Shklovskiĭ,[9] who considered the loss in cubic displacive ferroelectrics, but they concluded that this is not the dominant contribution in cubic ferroelectrics.[1)]

In Refs. 4–7 we compared the various contributions to the loss and obtained a general picture of the dielectric loss in crystals of different symmetries over a wide range of frequencies ω and temperatures T. This picture is qualitatively different from that proposed by Coombs and Cowley.[3] The origins of this discrepancy are discussed in Sec. 3.

As examples, in Refs. 4 and 5 we discussed in detail the crystal classes C_{3v}, C_{4v}, and C_{6v}. The results of those papers[4,5] also hold for a number of centrosymmetric and noncentrosymmetric classes (see Tables I and II). Generally, as we shall see, it is the crystal class that determines the form of the frequency and temperature dependence of the three-quantum loss for $T \ll \Theta$. Therefore, the need arose for a systematic study of the loss in all classes; for some of the classes we have obtained new data here. In particular, we have considered in detail the question (only briefly touched upon in Refs. 4 and 5) of how the loss is affected by the presence of lines of accidental contact of acoustic branches. As a result, the findings of Refs. 4 and 5 are corrected and supplemented.

We restrict discussion to the case of low frequencies ω of the alternating field:

$$\hbar\omega \ll T. \tag{1.1}$$

For $T \gtrsim \Theta$ the loss is due, by and large, to short-wavelength acoustic and optical phonons, and the situation becomes even more complicated: the answer can depend on the space group of the crystal symmetry. In this paper we discuss only briefly the behavior of the loss of $T \gtrsim \Theta$ (Sec. 4).

2. NONCENTROSYMMETRIC CRYSTALS

For noncentrosymmetric crystals, the loss can be written as the sum of two contributions—the quasi-Debye and three-phonon. The quasi-Debye loss is given by the formula (see p. 221 of Ref. 5):

$$\operatorname{Im}\varepsilon_{ij} = 4\pi T \sum_a \int \frac{d^3k}{(2\pi)^3} \frac{\omega \hat{I}}{\omega^2+\hat{I}^2} N_0(N_0+1) \times \left(\frac{\hbar\Omega}{T}\right)^2 \Lambda^{(i)}\Lambda^{(j)}. \tag{2.1}$$

Here the summation is over three acoustic branches a; I is the linearized operator for normal phonon–phonon colli-

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TABLE I. Noncentrosymmetric crystals.

Symmetry class	Symmetry degeneracy	EPP		Loss for minimum number of acoustic axes required by symmetry		Loss in the presence of additional "optional" acoustic axes	
C_1; C_2	0	1		$\omega^3 T$		$\omega^{2.5}T$, $\omega^2 T^2$	
D_2	0	1		$\omega^{2.5}T$, $\omega^2 T^2$		—	
C_s	0	1		$\omega^3 T$		$\omega^{2.5}T$, $\omega^2 T^2$ or $\omega^2 T^2$	
C_{2v}	0	1		$\omega^{2.5}T$, $\omega^2 T^2$ or $\omega^2 T^2$		$\omega^2 T^2$	
T	—	1		$\omega^2 T$, ωT^3		—	
O	—	1		$\omega^{2.5}T$, $\omega^2 T^2$		—	
T_d	+	1		ωT^3		—	
		∥	⊥	∥	⊥	∥	⊥
C_3	—	ϑ, k	1	$\omega^{2.5}T$, $\omega^2 T^2$	$\omega^{2.5}T$, $\omega^2 T^2$	—	—
D_3	—	ϑ	1	$\omega^{2.5}T$, $\omega^2 T^2$	$\omega^{2.5}T$, $\omega^2 T^2$	—	—
C_{3v}	+	ϑ	1	$\omega^2 T^2$	$\omega^2 T^2$	—	—
C_4	—	ϑ^2, k	ϑ	$\omega^3 T$ (*)	$\omega^{2.5}T$, $\omega^2 T^2$	$\omega^{2.5}T$, $\omega^2 T^2$	—
D_4	—	ϑ^2	ϑ	$\omega^3 T$	$\omega^{2.5}T$, $\omega^2 T^2$	$\omega^{2.5}T$, $\omega^2 T^2$	—
C_{4v}	+	ϑ^2	ϑ	$\omega^3 T$	$\omega^2 T^2$	$\omega^{2.5}T$, $\omega^2 T^2$ or $\omega^2 T^2$	*
S_4, D_{2d}	+	1	ϑ	ωT^3	$\omega^2 T^2$	—	*
C_6	—	k	ϑ	$\omega^3 T$	$\omega^{2.5}T$, $\omega^2 T^2$	*	$\omega^2 T$, ωT^3
D_6	—	$k^2\vartheta^2$	ϑ	$\omega^5 T$	$\omega^{2.5}T$, $\omega^2 T^2$	*	$\omega^2 T$, ωT^3
C_{6v}	+	$k^2\vartheta^4$	ϑ	$\omega^3 T$	$\omega^2 T^2$	$\omega^2 T^5$, $\omega^3 T$	$\omega^2 T \ln\frac{\omega_1}{\omega}$, ωT^3
C_{3h}	+	ϑ, k	1	ωT^5, $\omega^2 T^2$	ωT^3	ωT^5, $\omega^2 T$, ωT^3	*
D_{3h}	+	ϑ	1	$\omega^2 T^2$	ωT^3	$\omega^2 T \ln\frac{\omega_1}{\omega}$, ωT^3	*

sions, $\Omega_q(\mathbf{k})$ is the phonon frequency, N_0 is the Planck function, and $\Lambda(a,\mathbf{k})$ is the electrophonon potential (EPP), which characterizes the change in the phonon frequency in an electric field.

For thermal phonons, we have in order of magnitude

$$\dot{I} \approx \frac{1}{\tau} \approx \frac{T}{Mv^2}\left(\frac{T}{\Theta}\right)^3 \frac{T}{\hbar}, \qquad (2.2)$$

where M is of the order of the average mass of the atoms making up the dielectric, and v is some average sound speed. We shall sometimes find it convenient to write this estimate in a somewhat different form, setting

$$Mv^2 \approx \rho v^5 \hbar^3/\Theta^3,$$

where ρ is the density of the crystal. To determine the tem-

TABLE II. Centrosymmetric crystals.

Symmetry class	Symmetry degeneracy	EPP		Loss for minimum number of acoustic axes required by symmetry		Loss in the presence of additional "optional" acoustic axes	
C_i	0	1		$\omega^5 T$		$\omega^2 T^4$	
T_h	$-(C_2)$	ϑ		$\omega^2 T^4$ (*)		—	
	$+(C_3)$	1					
O_h	+	ϑ		$\omega^2 T^4$		—	
		∥	⊥	∥	⊥	∥	⊥
C_{2h}	0	1	ϑ	$\omega^5 T$	$\omega^5 T$	$\omega^2 T^4$	$\omega^2 T^4$ or $\omega^4 T^2$
D_{2h}	0	1	ϑ	$\omega^2 T^4$	$\omega^4 T^2$	—	$\omega^2 T^4$
S_6	+	1	ϑ	$\omega^2 T^4$	$\omega^2 T^4$	—	—
D_{3d}	+	ϑ^2	ϑ	$\omega^4 T^2$	$\omega^2 T^4$	—	—
C_{4h}	+	1	ϑ	ωT^5	$\omega^2 T^4$	—	—
D_{4h}	+	ϑ^2	ϑ	$\omega^3 T^3$	$\omega^2 T^4$	$\omega^2 T^4$	—
C_{6h}	+	1	ϑ	ωT^5	$\omega^2 T^4$	*	$\omega^2 T^2$, ωT^5
D_{6h}	+	$k^2\vartheta^4$	ϑ	$\omega^5 T^5$	$\omega^2 T^4$	$\omega^2 T^6$, ωT^9	$\omega^2 T^2$, ωT^5

Note. In classes C_{2h} and D_{2h} the angle ϑ is measured from the acoustic axis lying in the symmetry plane. In C_{2h} if there is no such axis, then ϑ should be replaced by 1.

perature and frequency dependence of the quasi-Debye loss, it is necessary to use the estimate for Λ given in Refs. 4 and 5:

$$|\Lambda| \approx 1/\rho^{1/2} v. \tag{2.3}$$

This value of Λ will be called the standard value for noncentrosymmetric crystals and will be denoted by Λ_1. If we assume that Λ has the standard value and that the main contribution to integral (2.1) is from thermal phonons,[2)] i.e., phonons with $\hbar\Omega \approx T$, we obtain the following estimate for the loss:

$$\operatorname{Im}\varepsilon \approx \frac{T^4}{\rho v^5 \hbar^3} \frac{\omega\tau}{1+(\omega\tau)^2} \approx \frac{T}{Mv^2}\left(\frac{T}{\Theta}\right)^3 \frac{\omega\tau}{1+(\omega\tau)^2}. \tag{2.4}$$

This estimate is good for all noncentrosymmetric classes with the sole exception of the longitudinal loss in the class D_6. One can show that for this crystal class the longitudinal component of the electrophonon potential can be estimated as

$$\Lambda_{\parallel} \approx \Lambda_1 (ka)^2. \tag{2.5}$$

It follows that a formula like (2.4) holds for the longitudinal component $\operatorname{Im}\varepsilon_{\parallel}$, but with $(T/\Theta)^3$ replaced by $(T/\Theta)^7$.

At the same time, we do not share the view of Coombs and Cowley,[3] who assume that the electrophonon potential is equal to zero for the class O. Below [see Eq. (2.25)] we write an explicit expression for the invariant in the free energy expansion that gives the standard value of Λ for this class.

Three-quantum loss

Three-quantum processes can be either decay or coalescence processes. For the first we have the estimate[4,5]

$$\operatorname{Im}\varepsilon \approx \frac{T}{Mv^2}\left(\frac{\omega}{\Omega_D}\right)^3 \approx \frac{\omega^3 T}{\rho v^5}, \tag{2.6}$$

where $\Omega_D = \Theta/\hbar$. The longitudinal loss in group D_6, for which $\operatorname{Im}\varepsilon \propto \omega^5 T$, is again an exception.

Three-quantum coalescence processes, in which a quantum of the electric field attaches to an acoustic phonon to form a phonon of another branch, give the following contribution:

$$\operatorname{Im}\varepsilon_{ik} = 4\pi^2 \frac{\hbar^2\omega}{T} \sum_{aa'} \int \frac{d^3k}{(2\pi)^3} \Lambda^{(i)}_{aa'} \Lambda^{(l)*}_{aa'} \Omega_a(\mathbf{k}) \Omega_{a'}(\mathbf{k}) N_0(N_0+1) \times \delta(\omega + \Omega_{a\mathbf{k}} - \Omega_{a'\mathbf{k}}), \tag{2.7}$$

where the superscripts indicate the number of the vector component of the electrophonon potential. The off-diagonal (in the branch index a) components of the electrophonon potential determine the amplitudes of transitions involving two phonons a and a' and the alternating electric field. The amplitude of such a transition is $\hbar\Lambda_{aa'}\mathbf{E}(\Omega_a\Omega_{a'})^{1/2}$.

The decay formula (2.6) gives the minimum possible value of the intrinsic loss. It is important to know whether the coalescence expression (2.7) is larger than this value. This can happen only if the absorption involves phonons whose frequencies Ω are much higher than ω (e.g., thermal phonons).[3)] In this case phonons of two vibrational branches of nearly equal frequency can participate in the coalescence process. In other words, such processes can occur in accordance with the general ideas of Herring near lines (or surfaces) of contact of two acoustic branches. In practice, these branches must be transverse or quasitransverse.

Thus, the problem reduces to one of analyzing how the branches diverge and how the electrophonon potential behaves as one moves away from the point of contact. We have carried out such an analysis for all crystal classes. As an illustration we consider examples of several classes. The results for the other classes are given without derivation. All the results are tabulated.

Classes C_1, C_2, and D_2. According to Herring,[11] lines of contact due to symmetry can occur only along the threefold, fourfold, or sixfold axes. Lines of accidental degeneracy in noncentrosymmetric crystals can lie only in symmetry planes,[4)] of which there are none in the given classes.

However, noncentrosymmetric crystals can have acoustic axes (see, e.g., Ref. 12). These are directions along which the velocity of quasitransverse waves as calculated in the theory of elasticity (the elastic approximation) coincide. In approximations to higher order in ka, the phonon frequencies should diverge for these classes. Therefore, lines of this kind will be called lines of quasidegeneracy.

If there are no such lines, the loss is given by the decay formula (2.6). However, if such lines do exist, they lie along random directions, and so there are no symmetry limitations on the electrophonon potential. Accordingly, its off-diagonal components comply with the standard estimate (2.3). Taking the k_z axis along the line of quasidegeneracy, we can write the k dependence of the divergence of the branches in the form

$$\Delta\Omega \equiv \Omega_2 - \Omega_1 = [(vk_\perp)^2 + v_1^2 a^2 k_z^4]^{1/2}. \tag{2.8}$$

Here v and v_1 are quantities which are of the order of the speed of sound and depend on the azimuthal angle φ. This expression takes two circumstances into account: 1) the transverse divergence of the phonon frequencies in the elastic approximation is a linear function of $k_\perp$; 2) the longitudinal divergence of the phonon frequencies arises in the next order above the elastic approximation and should therefore be proportional to k_z^2. In the spirit of Ref. 5 (see p. 208), we obtain an estimate for the contribution to the loss:

$$\operatorname{Im}\varepsilon \approx \frac{T}{Mv^2}\left(\frac{\omega}{\Omega_D}\right)^{2.5}, \tag{2.9}$$

which is valid for $\omega \ll \omega_1$, where

$$\omega_1 = \Omega_D (T/\Theta)^2. \tag{2.10}$$

The loss in this case is determined by subthermal phonons, specifically, those for which the longitudinal divergence of the branches is less than ω. In all such cases the loss is proportional to T [see Eq. (2.6)].

In the opposite case, when $\omega \gg \omega_1$, the frequency of the *rf* quantum is so high that the longitudinal divergence of the vibrational branches can be neglected, and the contribution to the loss turns out the same as from the line of degeneracy:

$$\operatorname{Im}\varepsilon \approx \frac{T}{Mv^2}\frac{T}{\Theta}\left(\frac{\omega}{\Omega_D}\right)^2 \qquad (2.11)$$

In all the formulas for the three-quantum loss written below, we indicate only the form of the temperature and frequency dependence. To obtain an estimate of the loss, we must make the formulas dimensionless by dividing one power of T by Mv_2, the remaining powers of T (if present) by the Debye temperature Θ, and the frequency ω by the Debye frequency Ω_D.

In Table I the class D_2 is put in a separate row. As was shown by Khatkevich,[12] crystals of the rhombic system always have acoustic axes. We have taken this into account in calculating the loss. Column 4 of Table I gives the results obtained with allowance for the mandatory acoustic axes which must exist by virtue of the crystal symmetry and whose orientation depends on the relationship among the elastic constants and in this sense is random. In column 5 we give the results which are due to the presence of additional acoustic axes which may or may not exist in the crystal depending on the relationships among the elastic constants.

Class C_s has a symmetry plane. Lines of true degeneracy can lie in this plane. If there are no such lines, then the loss in class C_s is the same as in classes C_1 and C_2. If such lines do exist, their contribution to the loss is given by the standard expression (2.11). Unlike the case of centrosymmetric crystals, here such a line does not give rise to anisotropy of the frequency and temperature dependence of the loss, in spite of the fact that when **k** lies in the symmetry plane the electrophonon-potential vector in this plane vanishes.

Class C_{2v} belongs to the rhombic system. In this class there are two mandatory acoustic axes.[12] If these axes do not lie in the symmetry plane, the result is the same as for D_2. However, if they or any of the nonmandatory axes lie in the plane, the result is given by (2.11).

Classes C_3, D_3, *and* C_{3v}. These classes have four mandatory acoustic axes, one of which is along the C_3 axis.[12,13] For the first two classes this is a line of quasidegeneracy, and for the third class it is a line of true degeneracy. For the mandatory lines of accidental (quasi) degeneracy, an estimate of the electrophonon potential is given by the standard expression Λ_1, and the contribution of these lines is given by (2.9) or (2.11). Allowance for the lines of symmetry degeneracy does not alter this estimate.

Classes C_6, D_6, C_{6v}, C_{3h}, *and* D_{3h} belong to the hexagonal system. Accordingly, in the elastic approximation the phonon spectrum is degenerate on the higher-order symmetry axis, and the branches diverge quadratically with distance from this axis. In the first two classes there is quasidegeneracy, and in the other three classes there is true degeneracy. It should be noted that our statements about classes C_{3h} and D_{3h} disagree with the conclusions of Herring.[8] Since the classes C_{3h} and D_{3h} belong to the hexagonal system, the elastic approximation for them should yield a quadratic divergence of the branches rather than the linear divergence given by Herring. The fact that the C_3 axis is a true line of degeneracy in the group C_{3h} (and is not of quasidegeneracy, as Herring has it in Ref. 8), can be seen from another paper by Herring[11] (see Table II of that paper).

The contribution to the loss from a line of symmetry degeneracy is determined by the dependence of the electrophonon potential on k and on the angle ϑ between the line of degeneracy and **k**; The powers of k and ϑ are given in Table I. Also given are the resulting frequency and temperature dependence of the absorption. As an illustration, let us find the functional form of the electrophonon potential for several of the cases indicated in Table I.

By analogy with Refs. 4 and 5, one can obtain the following general expression for the electrophonon potential:

$$\Lambda_{aa'}^{(p)} = \frac{e_i^*(\mathbf{k},a)\,e_m(\mathbf{k},a')}{2\rho\Omega_a(\mathbf{k})\,\Omega_{a'}(\mathbf{k})}(\xi_{p,ilmn}k_lk_n+\dots), \qquad (2.12)$$

where $e_i(k,a)$ are the polarization unit vectors of the acoustic vibrations, ξ is a fifth-rank tensor of a constitutive nature, and the ellipsis denotes terms containing higher powers of k and the corresponding constitutive tensors of higher rank. These terms must be taken into account when the lower-order terms vanish by reasons of symmetry.

The quantity in the numerator of (2.12) is nothing but the average value (over the volume) of the product of two components of the strain tensor due to two acoustic vibrations, a and a'. The problem thus reduces to finding the independent components of the constitutive tensors $\xi,\dots$, or, equivalently, to constructing the invariants of the symmetry transformations of the given crystal classes. These invariants should be linear functions of the electric-field components E_i and bilinear functions of the components of the strain tensor or its spatial derivatives and should not contain total derivatives with respect to the spatial coordinates,[5] since such terms do not contribute to the volume energy.

Let us analyze some concrete examples, starting with class C_{3h}. Two of its longitudinal invariants (i.e., those corresponding to a field **E** directed parallel to the C_3 axis) are given by the real and imaginary parts of the expression

$$E_z\frac{\partial}{\partial z}(u_x+iu_y)\left(\frac{\partial}{\partial x}+i\frac{\partial}{\partial y}\right)(u_x+iu_y), \qquad (2.13)$$

where **u** is the elastic displacement vector. Hence, with allowance for (2.12), we obtain for the longitudinal electrophonon potential the relation

$$\Lambda_\parallel \approx \Lambda_1\vartheta. \qquad (2.14)$$

However, there is another invariant which, as we shall see, is less important. It gives an electrophonon potential which is proportional to a higher power of k but which does not contain ϑ. This invariant is given by

$$\operatorname{Im} E_z\frac{\partial^2}{\partial z^2}(u_x+iu_y)\frac{\partial}{\partial z}(u_x-iu_y) = \frac{\partial^2 u_y}{\partial z^2}\frac{\partial u_x}{\partial z}-\frac{\partial^2 u_x}{\partial z^2}\frac{\partial u_y}{\partial z} \qquad (2.15)$$

and corresponds to

$$\Lambda_\parallel \approx ka\Lambda_1. \qquad (2.16)$$

Using (2.7), we see that the invariant (2.15) gives

$$\operatorname{Im}\varepsilon_\parallel \propto \omega T^5. \qquad (2.17)$$

At larger frequencies, however, the invariants (2.13) play an increasing role. They give the standard dependence

$$\mathrm{Im}\,\varepsilon_{\parallel}\propto\omega^2T^2, \tag{2.18}$$

which is dominant over contribution (2.17) for $\omega\gg\omega_2=\Omega_D(T/\Theta)^3$.

Let us find out whether these results can be altered by the accidental contact of vibrational branches. In the elastic approximation, the dispersion relations have cylindrical symmetry. Therefore, if there is accidental contact of vibrational branches, it occurs along the surface of a cone in **k** space.

In the next order above the elastic approximation, the contact vanishes, i.e., there is actually a surface of quasidegeneracy. For frequencies $\omega\gg\omega_1$ it behaves like a surface of true degeneracy, and we have, with allowance for the invariants (2.13),

$$\mathrm{Im}\varepsilon_{\parallel}\propto\omega T^3. \tag{2.19}$$

At frequencies $\omega\ll\omega_1$, we should use for the frequency splitting an expression of the type

$$\Delta\Omega=[(vk_{\perp})^2+v_1^2a^2k_{\parallel}^4]^{1/2}.$$

Here v and v_1 are quantities of the order of the speed of sound and depend on the azimuthal angle, and $k_{\parallel}$ and $k_{\perp}$ are the components of the vector **k** along the generatrix of the cone and perpendicular to the surface of the cone, respectively. In this case

$$\mathrm{Im}\,\varepsilon_{\parallel}\propto\omega^2T. \tag{2.20}$$

A comparison shows that for frequencies $\omega\gg\Omega_D(T/\Theta)^4$ the contribution from a surface of quasidegeneracy is much larger than the contribution from a line of symmetry degeneracy.

For the transverse invariant let us take

$$(E_x+iE_y)\left[\frac{\partial}{\partial z}(u_x+iu_y)\right]^2. \tag{2.21}$$

This invariant leads to the standard estimate for the values of $\Lambda_{\perp}$, which, moreover, remains finite at $\vartheta=0$. Hence for $\mathrm{Im}\varepsilon_{\perp}$, estimate (2.19) obtains at all frequencies. The presence of accidental degeneracy cannot alter this result, but it can lead to dispersion of the absorption coefficient at frequencies $\omega\approx\omega_1$.

Let us now turn to the group C_{6v}. This group has already been considered by one of the authors,[4,5] so we shall confine ourselves here to a few additional remarks. As was pointed out in Ref. 4, the contribution to the transverse loss from a line of symmetry degeneracy is given by expression (2.11). It was also noted in Ref. 4 that the presence of a surface of accidental degeneracy at frequencies $\omega\gg\omega_1$ leads to a contribution of type (2.19) to the transverse loss. To obtain the loss for $\omega\ll\omega_1$, we must take into account that in the next order above the elastic approximation, a surface of accidental degeneracy "decomposes" into 12 lines lying in the symmetry planes. The frequency splitting near a line of the kind is of the form

$$\Delta\Omega=[(vk_{\perp})^2+v_1^2k_2^2(k_{\parallel}a)^2]^{1/2}, \tag{2.22}$$

where k_2 is the component of the wave vector in the direction perpendicular to the symmetry plane. The contribution from each such line is

$$\mathrm{Im}\,\varepsilon_{\perp}\propto\omega^2T\ln(\omega_1/\omega). \tag{2.23}$$

In evaluating the longitudinal loss we encounter the peculiar situation that the form of the invariant alone does not permit a definite conclusion as to the form of the electrophonon potential. For example, one can easily write invariants which at first glance could lead to off-diagonal components of the electrophonon potential. An example of such an invariant is the product of E_z times any combination appearing in the expression for the elastic energy of a hexagonal crystal. However, for a suitable choice of correct zero-order functions, the contribution from such terms in (2.12) gives zero for $a\neq a'$.

In fact, suppose that the wave vector **k** deviates slightly from the z axis, so that its x component lies in the symmetry plane xz. Then the correct zero-order function would be two transverse vibrations, one of which is polarized along x and the other along y. At the same time, there would be no components of the constitutive tensor ξ [or of any of the higher-order tensors in (2.12)] which have only one index y, since the group C_{6v} includes a symmetry transformation which exchanges y and $-y$. And since $k_y=0$, the corresponding invariant must vanish.

If, however, we consider only the invariants of lowest order in k, they, like the elastic spectrum, have cylindrical symmetry. In this case we can always assume that the xz plane passes through the symmetry axis and the vector **k**, so that the contribution of the lowest-order invariants is zero for any orientation of **k**. To write the invariant one must therefore take into account derivatives of higher order.

The above considerations permit finding the dependence of $\Lambda_{\parallel}$ and $\Lambda_{\perp}$ on k and ϑ and, consequently, the dependence of the loss on ω and T for all noncentrosymmetric classes of the hexagonal and tetragonal systems (the divergence of the branches with distance from the symmetry axis in the latter case is also quadratic). This dependence is given in Table I.

In the elastic approximation, classes T, 0, and T_d of the cubic system have lines of degeneracy parallel to the edges or along the body diagonals of the cube. However, in the higher approximations in k the degeneracy remains only for the class T_d and is lifted for classes T and 0, so that we are actually dealing with quasidegeneracy (see Ref. 8).

Here the class T has no analog among the classes considered above. Along the twofold axis there is quasidegeneracy with a quadratic divergence of the branches and with a nondiagonal electrophonon potential, which is given for any direction by the standard estimate. The correctness of the latter assertion can be seen with the aid of the invariant

$$E_x\frac{\partial u_y}{\partial x}\frac{\partial u_z}{\partial x} \tag{2.24}$$

plus terms obtained by cyclic permutation of the coordinates.

For the class O we will simply give an invariant which yields a diagonal (and, incidentally, a nondiagonal) electrophonon potential. We wish to stress the existence of the first, since its presence contradicts Coombs and Cowley.[3] The invariant is of the form

$$E_z\left(\frac{\partial u_x}{\partial x}\frac{\partial u_x}{\partial y}-\frac{\partial u_y}{\partial x}\frac{\partial u_y}{\partial y}\right) \tag{2.25}$$

+ terms obtained by cyclic permutation of the coordinates.

In this way we have obtained the data of Table I. The first column lists the noncentrosymmetric classes. The second column indicates the presence of degeneracy (+), quasidegeneracy (−), or neither (0). The third column gives the dependence of the electrophonon potential on the angle ϑ (measured from the direction of highest symmetry) and k. A "1" in this column indicates that the electrophonon potential does not depend importantly on either ϑ or k. For cubic crystals the ones mean that there is no such dependence along the degeneracy or quasidegeneracy directions which give the leading contribution to the loss. For the class T these are the C_2 axes, for class O the C_3 axes, and for class T_d the S_4 axes.

The frequency and temperature dependences are given in the order in which they should succeed one other as the frequency is increased in an experiment. For rhombohedral crystals, column 4 also takes into account the contribution from the three mandatory lines of accidental degeneracy discussed above. The dashes in column 5 indicate that the frequency and temperature dependence of the loss do not change when the accidental degeneracy is taken into account. An asterisk means that the coefficient multiplying the powers of ω and T in the corresponding frequency and temperature intervals can exhibit dispersion at the intermediate frequency ω_1.

Overall picture of the loss in noncentrosymmetric crystals

A schematic is shown in Fig. 1a. At low frequencies the quasi-Debye loss is predominant. This loss passes through a maximum at $\omega\approx 1/\tau$ and then starts to fall off. It then gives way to the three-quantum loss, which causes a new increase of the intrinsic loss. A general feature of the three-quantum loss is that extrapolation into the region $\hbar\omega\approx T$ gives a universal value, specifically, the value corresponding to the maximum of the quasi-Debye loss. Here the sum of the exponents of ω and T in the highest frequency region is always equal to 4. Again, the longitudinal loss in class D_6 is an exception to these rules.

3. CENTROSYMMETRIC CRYSTALS

General considerations

In this case the total loss is the sum of the three-quantum and four-quantum losses. In noncentrosymmetric crystals the quasi-Debye loss was known to be dominant for $\omega\tau\lesssim 1$. Therefore, the behavior of the three-quantum loss in this frequency region was not of interest. For centrosymmetric crystals, however, it is important.

For $\omega\tau\gg 1$ the three-quantum loss is described by the general expression (2.7). For $\omega\tau\lesssim 1$, as one of the authors has shown,[14,6] the situation is as follows. If for $\omega\tau\gtrsim 1$ formula (2.7) implies that $\mathrm{Im}\varepsilon\propto\omega^n$ with $n>2$, then for $\omega\tau\lesssim 1$ the concept of a three-quantum loss cannot be introduced in a consistent manner, and the intrinsic loss on the whole is due to the four-quantum contribution (see below). On the other hand, if $n\leqslant 2$, then even for $\omega\tau\lesssim 1$ one can isolate a principal contribution to the loss that turns out to be just as sensitive to the details of the phonon spectrum as was the three-quantum loss for $\omega\tau\gg 1$. Remarkably, this isolation is possible even though the collisional spreading of the phonon frequencies here is greater than ω. We will keep the term "three-quantum" for this contribution, although it is not completely accurate in this frequency region.

We shall use the following prescription to obtain an expression for the three-quantum loss for $\omega\tau\ll 1$ (Refs. 14, 6). If $n<2$, then all but one the powers of ω in the expression obtained from formula (2.7) for $\omega\tau\gtrsim 1$ should be replaced by the same power of $1/\tau$. If $n=2$, an additional factor of $\ln(T\tau/\hbar)$ should also be appended.

The four-quantum loss at low temperature was also considered in Ref. 6. This loss turns out to be insensitive to the symmetry of the crystal and admits the estimate[6)]

$$\mathrm{Im}\,\varepsilon\approx\left(\frac{T}{Mv^2}\right)^2\left(\frac{T}{\Theta}\right)^7\frac{\omega}{\Omega_D}. \tag{3.1}$$

Three-quantum loss for $\omega\tau\gg 1$

In centrosymmetric crystals the diagonal components of the electrophonon potential are identically zero. The off-diagonal components are estimated as[4,5]

$$|\Lambda|\approx ka/\rho^{1/2}v. \tag{3.2}$$

We shall call this the standard value for centrosymmetric crystals and denote it by Λ_2.

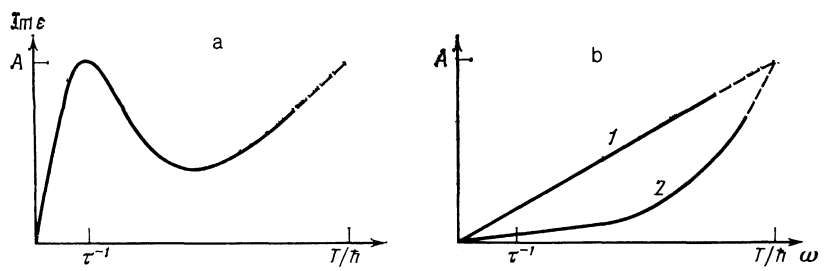

FIG. 1. Schematic of the frequency dependence of the imaginary part of the dielectric constant as a function of the field frequency ω for $\hbar\omega\ll T\ll\Theta$; τ is the characteristic collision time for thermal phonons; $A\approx T^4/Mv^2\Theta^3$; a) noncentrosymmetric crystals, b) centrosymmetric crystals, longitudinal loss; 1) class C_{4h}, 2) class D_{3d}.

As in noncentrosymmetric crystals, the absolute minimum of the loss in centrosymmetric crystals is limited by decay process. The only estimate for these processes is now of the form (except for D_{6h})

$$\mathrm{Im}\,\varepsilon \propto \omega^5 T. \tag{3.3}$$

Let us turn to a discussion of the individual crystal classes. In class C_i there is no symmetry degeneracy, and in the absence of accidental degeneracy the loss is given by (3.3). As Herring[11] has shown, however, in this class and in centrosymmetric classes in general there can be lines of accidental degeneracy. For the acoustic spectrum these are simply acoustic axes, which in this case are true lines of degeneracy (and not lines of quasidegeneracy). If such axes are present, we get

$$\mathrm{Im}\,\varepsilon \propto \omega^2 T^4, \tag{3.4}$$

where we have taken into account that the divergence of the branches with distance from such an axis is linear and have assumed that the electrophonon potential is given by the standard value Λ_2.

In exactly the same way, symmetry degeneracy is absent in the class C_{2h}. Therefore, in the absence of accidental degeneracy decay formula (3.3) holds for this class also. If there are acoustic axes, they will by and large be lines of true degeneracy. Then, according to Herring,[11] they can lie either in the symmetry plane or off it. In contrast to the case of noncentrosymmetric crystals, the contribution of the acoustic axes lying in the symmetry plane gives rise to anisotropy of the frequency and temperature dependence. This property is common to all lines of degeneracy lying in symmetry planes in centrosymmetric crystals.

We take the z axis to be perpendicular to the symmetry plane. In this case the transverse (with respect to the z axis) electrophonon potential is given by the estimate

$$\Lambda_\perp \approx \Lambda_2 k_z/k \tag{3.5}$$

(see Appendix), whereas the longitudinal electrophonon potential is given by the standard estimate Λ_2. The contribution to the longitudinal loss from a line of degeneracy lying in the symmetry plane is given by the standard estimate (3.4), while the contribution to the transverse loss is

$$\mathrm{Im}\,\varepsilon_\perp \propto \omega^4 T^2. \tag{3.6}$$

We see that the contribution to the loss from such a line of degeneracy is sharply anisotropic. At the same time, the contribution from an acoustic axis not in the symmetry plane would not have this anisotropy. Specifically, for the class C_{2h} this means that the contribution of the acoustic axes to the transverse (with respect to the C_2 axis) loss can be proportional either to $\omega^4 T^2$ (in the first case) or to $\omega^2 T^4$ (in the second).

In the class D_{2h} the three mutually perpendicular symmetry planes are not completely equivalent. One of them is known to contain two acoustic axes,[12] and the longitudinal and transverse losses in Table II are defined with respect to the normal to this plane.

For the other classes listed in Table II the lines of symmetry degeneracy were studied by the method of invariants, while the remaining lines were studied on the basis of the considerations discussed above.

We note that for the groups C_{6h} and D_{6h}, the situation in regard to the surface of accidental contact is reminiscent of that which we discussed above for noncentrosymmetric crystals of the hexagonal system. The difference is that now the expression of type (2.22) should contain $(k_\parallel a)^4$ rather than $(k_\parallel a)^2$ under the radical. Accordingly, the role of the characteristic frequency at which the transition from one limiting behavior to the other occurs is now played by $\omega_2 \approx \Omega_D (T/\Theta)^3$ rather than ω_1. The presence of dispersion in the loss at frequencies near ω_2 is denoted by an asterisk in Table II.

As we see from Table II, a characteristic feature of the three-quantum rf loss in centrosymmetric crystals is that the sum of the exponents of ω and T is equal to 6 (except for the longitudinal loss in the class D_{6h}).

General picture of the loss in centrosymmetric crystals

By considering Table II together with formula (3.1) for the four-quantum loss in the light of the above discussion, we can get a general picture of the loss. There is a qualitative difference between the behavior of the loss in noncentrosymmetric and centrosymmetric crystals: in the first case the curve of $\mathrm{Im}\,\varepsilon(\omega)$ goes through a maximum and then a minimum, while in the second case it is a monotonic function of ω.

As an example, let us consider the frequency dependence of the longitudinal loss in crystals of the classes C_{4h} and D_{3d} (Fig. 1b). In the first case the three-quantum contribution to the loss is dominant at all frequencies, while in the second case there is a transition from the four-quantum to the three-quantum loss as the frequency ω increases. The transition by no means occurs at frequencies $\omega \approx 1/\tau$, as was the case in the examples considered in Refs. 4 and 5.

Comparison with Coombs and Cowley[3]

We believe there is a fundamental difference between our results and those of Coombs and Cowley, primarily in regard to the so-called non-Debye (three- and four-quantum) losses. Let us briefly discuss this disagreement.

Coombs and Cowley calculated the loss using a diagram technique. On p. 132 of their paper, in analyzing the denominator of the single-particle phonon Green's function, they write: "The response is identical to that of a classically damped simple harmonic oscillator. There is a single damping constant which is expected to be frequency independent in the low-frequency limit, say for $\omega/2\pi < 1\,\mathrm{THz}$." This statement is equivalent to saying that $\mathrm{Im}\,\varepsilon \propto \omega$ over the entire frequency interval $\hbar\omega \ll T$. However, as can be seen from the present paper as well as from Refs. 4 and 5, the linear frequency dependence of the non-Debye loss (2.7) is actually a rare exception rather than the rule. The powers of the frequency and temperature depend in an essential way on the conservation laws characterizing the interaction of the electromagnetic field with the phonons, and they in turn depend

on the symmetry of the crystal. As a result, the damping coefficient off the mass surface in no way resembles that of a classically damped simple harmonic oscillator, as it has a far-from-trivial frequency and temperature dependence. The study of the frequency and temperature dependence for crystals of different symmetries is the central problem of the theory of dielectric loss.

As to the quasi-Debye loss, the existence of such a contribution, with its specific frequency dependence, was established in Ref. 3. In Refs. 4 and 5 and in the present paper we have studied the temperature distribution of this loss.

4. A FEW REMARKS CONCERNING THE CASE $T \gtrsim \Theta$

In this case the alternating field interacts not only with long-wavelength acoustic phonons but also with optical and short-wavelength acoustic phonons. Using the results presented above, one can estimate a lower bound on the high-temperature three-quantum contribution to the loss. This estimate is based on an extrapolation of the contribution from long-wavelength acoustic phonons into the short-wavelength region. The prescription for this estimate is to leave the frequency dependence as is but to replace all but one power of the temperature by Θ. Here the estimates of the contribution from lines (or surfaces) of quasi-degeneracy remains in force, since they are already proportional to T anyway for $\omega \ll \omega_1$ (or $\omega \ll \omega_2$). As to the region $\omega \gg \omega_1$ (or $\omega \gg \omega_2$), it lies outside the domain of application of the theory when $T \gtrsim \Theta$, since at such temperatures it must be assumed that $\omega_1 \approx \omega_2 \approx \Omega_D$.

For the contribution from the optical branches it can be shown that the presence of true lines of degeneracy does not alter these estimates. However, they can change substantially when allowance is made for lines of quasidegeneracy, points of intersection of the lines of degeneracy with the boundaries of the Brillouin zone, certain singular points on the lines, and surfaces of degeneracy. It can be noted further that the contribution from the neighborhood of the Brillouin zone center should be different for polar and nonpolar vibrations. Analysis of most of these questions requires knowledge not of the crystal class but of the symmetry space group, and so for $T \gtrsim \Theta$ the loss generally is sensitive to the space group.

5. DISCUSSION OF THE EXPERIMENTAL SITUATION

Many papers have been written about the observation of dielectric losses in crystals, but most of the measurements have been made at low frequencies, where it is scarcely possible to be sure that the loss is of an intrinsic nature.

Papers on the observation of the losses at higher frequencies are few in number. We might mention the studies by Stolen and Dransfeld,[2] Owens,[15] and Hadni[16] on alkali halide crystals. These studies dealt mainly with the high-temperature case.

Here, however, we are interested in the low-temperature loss. Let us therefore discuss the recent experiments of Ref. 17 on the loss in leucosapphire[7] (Al_2O_3, class D_{3d} of the rhombohedral system). In these experiments the temperature dependence of the loss was studied at two frequencies: $\omega = 2\pi \cdot 9$ GHz and $\omega = 2\pi \cdot 36$ GHz. In both cases the loss was proportional to T^m, with $m = 4.79 \pm 0.27$ for the first frequency and $m = 4.85 \pm 0.33$ for the second. This dependence was observed at temperatures between 260 and 60 K; as T was decreased further the curve of $\mathrm{Im}\varepsilon(T)$ became parallel to the abscissa. That the loss was of an intrinsic nature in these experiments can be considered established. Since the Debye temperature of leucosapphire is 1054 K, it is natural to use a theory developed for the case $T \ll \Theta$ in interpreting the data of this experiment.

On the other hand, this inequality is satisfied without a very large safety factor. It is therefore necessary to indicate what temperatures can be regarded as sufficiently low that the experimental data can be interpreted in the framework of the present study. It might be thought that a temperature is clearly small from our point of view if the various other kinetic and thermodynamic characteristics of the crystal exhibit the power-law behavior given by the theory for $T \ll \Theta$. As we know, however, both the specific heat and the thermal expansion of leucosapphire actually[18,19] do not obey a T^3 law over anywhere near the entire temperature range 60–260 K. We do not see a contradiction between this circumstance and the fact that the loss obeys a power law over a much wider temperature range. The specific heat and thermal expansion contain contributions from all the vibrational branches, while the intrinsic loss contains contributions only from the transverse branches near lines of contact. There is also a substantial difference in the quantities over which the thermodynamic average is taken: in the calculation of the loss the expression to be averaged includes the square of the electrophonon potential, which has a substantial dependence on k.

However, since we know that the limits of the low-temperature region are exceeded in regard to the specific heat and thermal expansion, we should not expect a very detailed agreement with the predictions of the theory of intrinsic loss as to the power of T. The criteria by which the loss can be judged intrinsic in this case, we believe, are that the loss be proportional to a sufficiently high power of the temperature, that it be of the correct order of magnitude (we consider this criterion to be very important), and that it be independent of the defect concentration.

Let us examine how our theory permits interpretation of the experimental data for $T \ll \Theta$. One possibility is indicated in Ref. 17. It is based on similarity of the elastic properties of leucosapphire to those of a hexagonal crystal with a surface of degeneracy in the elastic spectrum. In this case, according to Refs. 4, 5, and 20, the loss is given in order of magnitude by the estimate

$$\mathrm{Im}\,\varepsilon \approx \eta \frac{\omega T^5}{\rho v^5 \hbar^2 \Theta^2}. \tag{5.1}$$

Here η is a dimensionless coefficient which characterizes the effect of the electric field on the elastic vibrations. The exact value of this coefficient remains unknown, but in order of magnitude it is between 1 and 100. If the values $\omega = 6 \cdot 10^{10}$ sec^{-1}, $T = 100$ K, $\eta = 1$, and the other values for leucosapphire are plugged into (5.1), one gets $\mathrm{Im}\varepsilon \approx 5 \cdot 10^{-9}$. This value would agree satisfactorily with the experimental esti-

mate[17] $\mathrm{Im}\varepsilon \lesssim 5 \cdot 10^{-7}$ if $\eta \approx 10^2$. This may mean that the electrophonon potential is 10 times as large as the value given by estimate (3.2). Dimensionless factors of order 10 can arise in the theory; for example, in many crystals the anharmonic stiffness constants are 10 times as large as the harmonic constants.

As far as one can tell from published data, the elastic properties of leucosapphire are in fact relatively close to those of a hexagonal crystal. According to Ref. 21, the elastic stiffness constants in the temperature range of interest are (in 10^{11} dyn/cm^2): $C_{11} = 49.5$, $C_{33} = 49.7$, $C_{44} = 14.6$, $C_{13} = 11.5$, and $C_{12} = 16.0$. At the same time, the constant C_{14}, which characterizes the nonhexagonality of a rhombohedral crystal, is only $C_{14} = -2.3$. If this "rhombohedral" constant C_{14} is neglected, the remaining constants are such that leucosapphire actually does have a surface of degeneracy.

With allowance for C_{14}, however, this surface "decomposes" into six acoustic axes in accordance with the symmetry of the rhombohedral crystal. In the intermediate directions, i.e., between the acoustic axes, the transverse vibrational frequencies split. Estimates show that for $C_{14} = -2.3$ the relative value of the splitting is less than 0.1. For thermal phonons, however, even a gap of this size is larger than the frequency ω, and therefore at such low frequencies ω leucosapphire cannot be treated as a hexagonal crystal.

Furthermore, as we showed in Sec. 3, estimate (5.1) is only good at $\omega \gg \omega_2$, even for a hexagonal crystal. For example, at 100 K we have $\omega_2 \approx 10^{10}$ for leucosapphire, i.e., a crude estimate puts us at the boundary of the domain of application of (5.1). (As the amount of experimental material accumulates, this estimate will need to be refined, since we do not know the actual values of the coefficients characterizing the dispersion of the sound velocity.)

Another possibility is to explain the experiments[17] in terms of a rhombohedral crystal. According to Refs. 4 and 5 and the present study, the contribution to the loss from the neighborhood of a single point of contact of constant-frequency surfaces in rhombohedral crystals of class D_{3d} differs from (5.1) by a factor of $\hbar\omega/T$, or approximately 10^{-2}. However, this small factor, as we shall presently see, is compensated. Leucosapphire has six additional acoustic axes with a pair of contact points on each. An additional factor of 12 thus appears in the estimate. The estimate was also made on the assumption that the difference of the phase velocities of the transverse acoustic vibrations is of the order of the average velocity of transverse sound. Actually, however, as can be judged directly from the shape of the reciprocal-sound-velocity surfaces near the points of contact,[19] this difference is several times smaller. Accordingly, the loss turns out to be as many times larger.[8)] As a result, a factor of order 10^2 is obtained, so that order-of-magnitude estimates for a rhombohedral crystal also can in principle agree with the experimental data.[17]

It should be kept in mind that this estimate for a rhombohedral crystal holds only for $\omega \gg 1/\tau$, where τ is the characteristic collision time for thermal phonons in regard to both their anharmonic interaction with other phonons and their scattering by lattice defects. For an arbitrary relationship between ω and $1/\tau$ the estimate is of the form[6]

$$\mathrm{Im}\,\varepsilon \approx 10^2 \eta \frac{\omega T^4}{\tau \rho v^5 \hbar \Theta^2}\left[\ln \frac{T^2}{\hbar^2(\omega^2+1/\tau^2)} + \omega\tau \operatorname{arctg} \omega\tau\right]. \tag{5.2}$$

For $\omega\tau \gg 1$ this expression goes over to the estimate discussed above.

There are also intermediate possibilities which arise if, for example, one assumes that the elastic constants stand in definite relationships that are not dictated by the crystal symmetry. For example, a slight refinement of the values of the shear stiffness (and not only C_{14}) from the values given above can lead to a noticeable change in the angle between two neighboring acoustic axes and, in particular, to their practical coincidence. (To be convinced of this one need only compare Figs. 35A.13.1 and 35A.13.2 in Ref. 19, which were constructed for slightly different values of the elastic constants of leucosapphire: in one case there are no optional accidental intersections of the transverse branches, and in the other case there are such intersections.) If the angle between neighboring acoustic axes is so small that the corresponding frequency gap between them is smaller than ω, then this is equivalent to the touching of constant-frequency surfaces which diverge quadratically in the symmetry plane and linearly in the direction perpendicular to this plane. Such an accidental touching would lead to a $\omega^{1.4} T^{4.5}$ law for $\omega\tau \gg 1$. We shall not discuss the other intermediate possibilities here.

Additional data for choosing among the possibilities mentioned above can be obtained by studying the frequency dependence of the intrinsic loss. It must be remembered, however, that the intermediate case corresponding to an intermediate temperature dependence and an intermediate frequency dependence can arise in experiment [e.g., Eq. (5.2) with $\omega\tau \approx 1$].

We are sincerely grateful to E. L. Ivchenko and G. E. Pikus for an extremely fruitful discussion and to V. B. Braginskiĭ for a detailed discussion of the experimental situation.

APPENDIX

Electrophonon potential near lines of accidental degeneracy lying in a symmetry plane

Let us consider two (quasi) transverse acoustic branches having a line of degeneracy in a symmetry plane. One of the branches is even and the other odd with respect to reflection in this plane. We begin with noncentrosymmetric crystals. The longitudinal (with respect to the normal to the plane—the z axis) nondiagonal electrophonon potential is given by the invariant

$$E_z \frac{\partial u_y}{\partial \xi} \frac{\partial u_z}{\partial \xi},$$

where ξ is the direction of the line of degeneracy, and y and z are the directions of the polarization of the two degenerate (quasi) transverse vibrations. The transverse electrophonon

potential is given by the invariants

$$E_x\left(\frac{\partial u_y}{\partial \xi}\right)^2, \quad E_y\left(\frac{\partial u_y}{\partial \xi}\right)^2, \quad E_x\left(\frac{\partial u_z}{\partial \xi}\right)^2, \quad E_y\left(\frac{\partial u_z}{\partial \xi}\right)^2 \tag{A.1}$$

We will be interested in the values of the electrophonon potential in directions close to the line of degeneracy: $\mathbf{k} = \mathbf{k}_0 + \Delta\mathbf{k}$, where $\mathbf{k}_0$ lies on the line of degeneracy. If $\Delta\mathbf{k}$ lies in the symmetry plane, then the invariants (A.1) do not give off-diagonal components of the electrophonon potential, since in this case a vibration, being either even or odd with respect to reflection in this plane, cannot simultaneously contain the y and z components of the polarization vector.

If, however, the vector $\Delta\mathbf{k}$ forms an angle of the order of unity with the plane, then the correct zero-order functions are superpositions of vibrations polarized along y and z. In this case invariants (A1) will give a nondiagonal transverse electrophonon potential having an estimate which, from the standpoint of the contribution to the loss, is practically equivalent to the standard estimate:

$$\Lambda_\perp \approx \Lambda_1 \Delta k_z / |\Delta\mathbf{k}|.$$

The longitudinal component of the electrophonon potential in centrosymmetric crystals can be provided by the invariant

$$E_z\left(\frac{\partial u_y}{\partial \xi}\frac{\partial^2 u_z}{\partial \xi^2} - \frac{\partial u_z}{\partial \xi}\frac{\partial^2 u_y}{\partial \xi^2}\right),$$

which leads to the standard estimate.

Let us turn to the transverse component of the electrophonon potential. Invariants which contain only the spatial derivatives with respect to ξ, i.e., which do not vanish when $\mathbf{k}$ lies on a line of degeneracy, can be constructed only by using u_x and u_y simultaneously. To lowest order, such invariants are of the form

$$E_{x,y}\left(\frac{\partial^2 u_x}{\partial \xi^2}\frac{\partial u_y}{\partial \xi} - \frac{\partial u_x}{\partial \xi}\frac{\partial^2 u_y}{\partial \xi^2}\right). \tag{A.2}$$

For our choice of coordinate axes, if the vector $\mathbf{k}$ comes slightly out of the symmetry plane, then the correct zero-order functions, as before, are polarized in the yz plane. Therefore, the invariant (A.2), unlike (A.1), does not contribute to the transverse nondiagonal electrophonon potential. Allowance for invariants containing derivatives with respect to the coordinate η, directed perpendicular to ξ in the symmetry plane, will not alter this conclusion.

Allowance for invariants containing derivatives with respect to z leads to estimate (3.5) in the text.

[1)] We do not share this view, for reasons which have been discussed in detail in the paper by Tagantsev.[10]

[2)] The peculiarities of those cases in which the main contribution to the quasi-Debye loss if from subthermal phonons have been discussed in detail in the book by Gurevich (Ref. 5, p. 315) and will not be considered here.

[3)] It is easily checked that the coalescence contribution from subthermal phonons with frequencies of the order of ω turns out to be of the same order as the decay contribution (2.6).

[4)] Herring[11] also discussed points of accidental degeneracy, but these should not be present in the long-wavelength part of the phonon spectrum.

[5)] This condition can also be regarded as a consequence of the fact that the Hamiltonian of the lattice vibrations is real. For electronic systems the limitations due to the reality of the Hamiltonian are also linked with the time-reversal symmetry of the wave equation.

[6)] For centrosymmetric crystals the longitudinal loss in the group D_{6h} is an exception.

[7)] We shall not discuss the data[17] on the intrinsic loss in $Y_3Al_5O_{12}$. This material has many atoms in the unit cell and hence an enormous number of optical vibrational branches. Off-hand we would say that the theory will need further development in order to describe the loss in crystals of this kind.

[8)] Such an effect is in principle possible in hexagonal crystals as well if the constant-frequency surfaces intersect at a small angle along lines of accidental contact.

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