Intrinsic dielectric losses in crystals at high temperatures

V. L. Gurevich and A. K. Tagantsev

A. F. Ioffe Physico-Technical Institute
(Submitted 29 November 1989)
Zh. Eksp. Teor. Fiz. **97**, 1335–1345 (April 1990)

We have analyzed theoretically the frequency and temperature dependence of intrinsic dielectric losses in ordinary insulator crystals when the temperature is on the order of the Debye temperature or higher. We investigate crystals with symmorphic symmetry groups which belong to one of the 32 crystal classes. We have taken into account contributions to the losses connected both with symmetry-induced degeneracy of the phonon spectrum and with the most probable types of accidental degeneracy. We show that structure in the high-energy portion of the phonon spectrum (i.e., where the energy is on the order of the Debye temperature) can significantly affect the form of the frequency and temperature dependence of the intrinsic dielectric losses. We establish that the forms of this dependence for crystals with symmorphic groups are determined by the crystal class, but not by the space groups within a given class.

1. INTRODUCTION

In this paper we describe a theoretical investigation of the intrinsic dielectric losses in ordinary crystalline insulators at high temperatures T (compared to the Debye temperature Θ). This paper should be regarded as a continuation of our previous paper Ref. 1, in which we investigated the low-temperature case.[1)] In this paper we will discuss only crystals with symmorphic space groups, and limit ourselves to the case of frequencies ω low enough so that

$$\hbar\omega \ll \Theta$$

(here and in what follows, the temperature will be in energy units).

As we pointed out in Ref. 1 (see also Ref. 2), losses in noncentrosymmetric (NCS) crystals can be represented in the form of a sum of three-phonon and quasi-Debye losses. In contrast, in centrosymmetric (CS) crystals the losses can be cast in the form of a sum of three-phonon and four-phonon losses. The quasi-Debye and four-phonon losses, which were investigated in detail in Ref. 1, are insensitive to the details of the phonon spectrum. The corresponding frequency and temperature dependences can easily by obtained from Eqs. (2.2), (2.4) of Ref. 1 by replacing all powers of T except the first by Θ; in Eq. (3.1) we must retain two powers of T.

Mathematically, this is connected with the fact that integration over the wave vectors of the acoustic phonons now involves an effective cutoff which is no longer due to the Planck function, which cuts off the corresponding integrals at a characteristic value of $k = k_T = T/\hbar v$ (where v is a characteristic value of the sound velocity); instead, the cutoff occurs at $k = k_0$, where k_0 is the limiting value of the phonon wave vector.

The three-phonon losses are sensitive to the shape of the phonon spectrum; this was already apparent in Ref. 1, where we used the distinctive features of the spectrum of long-wavelength acoustic phonons to analyze the low-temperature losses. In Ref. 1 we also showed that by extrapolating the results of Ref. 1 to the high-temperature regime we obtain a minimum of the dielectric loss; this constitutes a lower bound on the latter. This paper will concern itself with exhibiting the distinctive features of the short-wavelength and/or high-frequency part of the phonon spectrum and clarifying how these features affect the losses. In this section and the two which follow we will investigate the contribution of three-phonon losses, as this term is understood "classically," i.e., for $\omega\tau \gg 1$ (τ is a characteristic lifetime of the phonons which interact to produce the losses). The case of lower frequencies is discussed in Sec. 4.

Specifically, we are interested in the so-called associated losses, i.e., processes which couple quanta of the electric field with a phonon to create a phonon on another branch. They give the following contribution to the imaginary part of the dielectric permittivity ε:

$$\operatorname{Im}\varepsilon_{il}=4\pi^2\frac{\hbar^2\omega}{T}\sum_{jj'}\int\frac{d^3k}{(2\pi)^3}\Lambda_{jj'}^{(i)}\Lambda_{jj'}^{(l)*}\Omega_{j\mathbf{k}}\Omega_{j'\mathbf{k}}N_0(N_0+1)$$

$$\times\,\delta(\omega+\Omega_{j\mathbf{k}}-\Omega_{j'\mathbf{k}}), \tag{1.1}$$

where the subscripts jj' indicate the branch of the spectrum while the superscripts label the vector component of the electron-phonon potential (EPP) Λ. If the temperature T is much larger than the characteristic phonon frequencies, we can use the following approximation for the Planck distribution N_0:

$$N_0 = T/\hbar\Omega. \tag{1.2}$$

Substituting (1.2) into (1.1), we find that Im $\varepsilon \propto T$ at high temperatures (this fact was pointed out by Vinogradov in Ref. 3), and (1.1) becomes

$$\operatorname{Im}\varepsilon_{il}=4\pi^2\hbar^2\omega T\sum_{jj'}\int\frac{d^3k}{(2\pi)^3}\Lambda_{jj'}^{(i)}\Lambda_{jj'}^{(l)*}\delta(\omega+\Omega_{j\mathbf{k}}-\Omega_{j'\mathbf{k}}). \tag{1.3}$$

However, the frequency dependence of Eq. (1.3) depends on the behavior of the photon spectrum and the EPP near "contact points," i.e., points in k-space where some pair of phonon frequencies on different branches coincide.

The only kinds of contact points which were studied in the previous article[1] were lines of degeneracy (or quasidegeneracy) of the phonon spectrum near which the "association" processes could occur. Naturally, in the low-tempera-

 0038-5646/90/040753-07$03.00

ture case only the initial (long-wavelength) portions of the acoustic phonon branches play a role. Here, however, we will be interested in lines of degeneracy in the short-wavelength part of the acoustic spectrum, and also the optical branches of the spectrum over their entire range. In this case, the isofrequency surfaces are not even remotely similar. Correspondingly, new objects arise in the theory of losses whose contributions require analysis—points of degeneracy of the spectrum and special points on the lines of degeneracy.

We will apply our results to the special case of a particular crystal class, which we pick to be C_{3v}. For the other classes, we will indicate briefly the characteristic differences in their treatment and trace the origins of these differences; as for remaining details, we limit ourselves to presenting results which will be reduced to tables.

2. AN EXAMPLE: THE CLASS C_{3v}

As we showed in Ref. 1, the low-temperature losses are due to processes which take place near four lines of degeneracy of the long-wavelength acoustic phonon spectrum. In this case one finds Im $\varepsilon \propto \omega^2$.

The transverse losses are determined by a line of symmetry-induced degeneracy. This line goes throughout the whole Brillouin zone along the C_3 axis. For the high-temperature transverse losses there exists a short-wavelength part of this line which, as in Ref. 1, gives Im $\varepsilon_\perp \propto \omega^2$.

The longitudinal losses are given by three "obligatory" lines of accidental degeneracy lying in the symmetry planes. As with the transverse-loss case, these lines must necessarily extend into the short-wavelength part of the spectrum, and therefore they also give a contribution Im $\varepsilon_\parallel \propto \omega^2$.

At high temperatures the optical branches are excited as well. There is also a line of degeneracy along the C_3 axis in the transverse optical branches, which gives a contribution to Im $\varepsilon_\perp$ that, generally speaking, is of the same order as the contribution from the "obligatory" lines of accidental degeneracy of the acoustic branches.

At first glance it appears that the theory of the phenomenon should be exhausted by this, and that we need only verify whether or not there is a special contribution from the ends of the degeneracy lines and (for optical phonons) from the center of the Brillouin zone.

Let us begin with a discussion of the structural features of the spectrum of long-wavelength optical phonons. The degenerate optical vibrations which propagate along the C_3 axis must be polar in general, because the only two-dimensional representation of the group C_{3v} is a vector one (additional degeneracy connected with the reality of the Hamiltonian in the coordinates of the representation does not arise). For small inclinations of the propagation direction from the C_3 axis a splitting of the transverse optical branches appears; in analyzing this splitting we must include the contribution of the macroscopic electric fields which appear as well as spatial dispersion.

Let us investigate two mutually degenerate optical vibrations, which are characterized by a vector **w** proportional to the corresponding relative displacement of the sublattices. Because in an insulator these vibrations take place with a fixed longitudinal component of the displacement **D** (by virtue of the condition div **D** = 0), the role of the potential energy for the optical vibrations is played by the expression

$$\frac{1}{2}\left(\alpha_{ij}+i\lambda_{ijm}k_m+n_{imjp}k_mk_p+\frac{4\pi}{\varepsilon_{ab}^{(\infty)}k_ak_b}\beta_{il}\beta_{jm}k_lk_m\right)w_iw_j$$

$$\equiv H_{ij}w_iw_j. \qquad (2.1)$$

Here the first term describes the increase in energy when there is a uniform relative displacement of the sublattices under the condition that the macroscopic fields vanish; the last term describes the contribution from the macroscopic electric fields (see the paper by Lang and Pashabekova[4] or the book by one of the authors of the present paper[5]). As was pointed out in Ref. 5, the tensor β_{il} is nonsymmetric, generally speaking. The second and third terms are the next terms of an expansion in powers of the small wave vector **k**, i.e., they describe the spatial dispersion; $\varepsilon_{ab}^{(\infty)}$ is the dielectric permittivity calculated without including pairs of branches like those under discussion here.

The splitting of the phonon frequencies is calculated according to the following general expression:

$$\Delta\Omega^2=2\Delta\Omega\Omega=[(H_{11}-H_{22})^2+4|H_{12}|^2]^{1/2}. \qquad (2.2)$$

Let us point out first of all that the second term in (2.1) must be equal to zero. In fact, the only invariant admitted by the symmetry C_{3v}, i.e.,

$$\mathrm{Re}\,(w_x+iw_y)\frac{\partial}{\partial z}(w_x-iw_y)$$

is a total derivative with respect to z, and the integral of such a term reduces to a surface integral.

The invariants corresponding to the third term in (2.1) have the form

1) $\left(\frac{\partial w_x}{\partial z}\right)^2+\left(\frac{\partial w_y}{\partial z}\right)^2$

2) $\left|\left(\frac{\partial}{\partial x}+i\frac{\partial}{\partial y}\right)(w_x+iw_y)\right|^2$,

3) $\mathrm{Re}\left[\left(\frac{\partial}{\partial x}-i\frac{\partial}{\partial y}\right)(w_x+iw_y)\right]^2$,

4) $\mathrm{Re}\frac{\partial}{\partial z}(w_x+iw_y)\left(\frac{\partial}{\partial x}+i\frac{\partial}{\partial y}\right)(w_x+iw_y)$.

The first two of these invariants do not give any splitting, since they correspond to $H_{11}=H_{22}$, $H_{12}=0$. The remaining invariants give a first-order splitting for $k_\parallel \sim k_\perp$, which results in $\Delta\Omega \propto k^2$ for this case. If, however, we have $k_\parallel \gg k_\perp$, then the last invariant turns out to be important, which gives $\Delta\Omega \propto k_\parallel k_\perp$. This splitting turns out to be proportional to the first power of $k_\perp$, in agreement with the general behavior of a line of degeneracies parallel to the C_3 axis.[6]

From this we see that even without including the contribution from macroscopic electric fields the behavior of the splitting as we depart from the point $k=0$ (the Γ point) differs from its behavior away from the line of degeneracy parallel to the C_3 axis. There is a remarkable assertion connected with this, whose validity we will prove below: the contribution to the losses from the vicinity of the point $k=0$ will be parametrically larger than the contribution from the entire line.

In analyzing the fourth term in (2.1) we will take into account the fact that symmetry admits the presence of the following components of the tensor β: $\beta_{xx}=\beta_{yy}$ and β_{zz}. Using the general expression (2.2), we obtain the following equations:

$$\Delta\Omega^2=\Xi^{1/2};\quad \Xi=[Ak_xk_z+B(k_x^2-k_y^2)+F(n_x^2-n_y^2)]^2 +4\left(\frac{A}{2}k_yk_z-Bk_xk_y+Fn_xn_y\right)^2, \tag{2.3}$$

where $n_i=k_i/k$, A and B are proportional to the coefficients of the invariants (4) and (5) respectively, and $F=4\pi\beta_{xx}^2/\varepsilon_{zz}^{(\infty)}$. The final expression for the frequency splitting can be written in the form

$$\Delta\Omega=\Omega_D[c_1n_\perp^4+c_2n_\perp^2k_\perp^4a^4+c_3kk_za^2n_\perp^3\cos 3(\varphi-\varphi_0)]^{1/2}, \tag{2.4}$$

where Ω_D is the Debye frequency, c_i is a dimensionless constant of order unity, a is the mean interatomic spacing, which measures the spatial dispersion, φ is the azimuthal angle, and φ_0 is a constant (in (2.4) we have included the leading terms in the parameter ka).

In what follows, in order to calculate the losses we require an estimate of the nondiagonal components of the EPP, which correspond to transitions between the optical branches of interest to us. The transverse EPP is given by the invariant (**E** is the external electric field):

$$\mathrm{Re}[(E_x+iE_y)(w_x+iw_y)^2], \tag{2.5}$$

clearly this quantity does not depend either on k or on angle in any important way. We obtain the following estimate for it (compare with Ref. 1):

$$|\Lambda^\perp|\sim\Lambda_1. \tag{2.6}$$

Here Λ_1 is the so-called standard value,[1] which in order of magnitude equals

$$\Lambda_1\sim 1/\rho^{1/2}v, \tag{2.7}$$

where ρ is the density of the crystal.

The longitudinal EPP is given by the invariant

$$E_z\,\mathrm{Re}\frac{\partial}{\partial z}(w_x+iw_y)\left(\frac{\partial}{\partial x}+i\frac{\partial}{\partial y}\right)(w_x+iw_y), \tag{2.8}$$

from which

$$|\Lambda^{\parallel}|\sim\Lambda_1kk_\perp a^2. \tag{2.9}$$

Let us begin with a calculation of the transverse losses. Using (1.3), (2.4) and the estimate (2.6), we see that the integral (1.3) contains the small quantities k_z and $k_\perp$; this justifies our use of Eq. (2.4), which was based on an expansion in k, to calculate the losses. Transforming the integration variables $n_\perp\to(\omega/\Omega_D)^{1/2}n_\perp$, $k\to(\omega/\Omega_D)^{1/4}k$, we find that

$$\mathrm{Im}\,\varepsilon_\perp\propto\omega^{7/4}. \tag{2.10}$$

Let us point out that when we use expressions of this type to estimate the losses, we must reduce each power of T to dimensionless form by dividing it by Mv^2 (where M is the mean mass of the atoms which make up the crystal and v^2 is the mean square of the sound velocity) and divide the frequency ω by the Debye frequency Ω_D. The smaller the power of ω, the larger the losses, since the ratio ω/Ω_D is assumed to be small. From this we see that the vicinity of the point $k=0$ gives a contribution larger than the entire line of degeneracy, which leads to the dependence Im $\varepsilon\propto\omega^2$.

An analogous calculation for the contribution of the zone center to the longitudinal losses would give Im $\varepsilon\propto\omega^{15/4}$. This is larger than the contribution from the entire line of symmetry-induced degeneracy. However, in the present case this is unimportant, because the contribution is smaller than that from the "obligatory" line of accidental degeneracy of the acoustic branches.

However, there is a point on the line of accidental degeneracy which gives a contribution even larger than (2.10). This is the point Z with coordinates $(0,0,\pi/a_3)$, where a_3 is the period of the lattice in the direction of the z-axis. The Hamiltonian of the lattice vibrations in the vicinity of the point Z has the same form as the Hamiltonian in the vicinity of the Γ point, except that near Z there is no contribution from the macroscopic electric fields. Actually, at the point Z the group of the wave vector is the same as for the Γ point, and includes the same symmetry element which transforms **k** to $-$ **k**. Therefore all the limitations induced by symmetry on the form of the phonon spectrum at these points are identical. The same applies to the EPP.

The splitting in the neighborhood of the point Z is given by Eq. (2.3) with $F=0$. We have

$$\Delta\Omega=\Omega_Da^2k_\perp[c_1k_z^2+c_2k_\perp^2+c_3k_zk_\perp\cos 3(\varphi-\varphi_0)]^{1/2}, \tag{2.11}$$

where **k** is measured from the point Z; the value of the constant is obviously different from that in Eq. (2.4).

Calculating the transverse losses in the same way as before, we are led to the result

$$\mathrm{Im}\,\varepsilon_\perp\propto\omega^{3/2}. \tag{2.12}$$

From this we see that the contribution from the point Z is even larger than that from the point Γ. The contribution to the longitudinal losses from this point is proportional to $\omega^{7/2}$, which in contrast is smaller than the contribution from the "obligatory" line of accidental degeneracy.

In addition to the points Z and Γ, there may also be yet another type of point which can give a contribution to the losses. This is a so-called "point of accidental tangency"; we will denote such points with the letter Ψ. As we have already mentioned, for small excursions from the C_3 axis the branches diverge linearly; in the group C_{3v} there is only a single invariant which corresponds to this divergence. Actually, the group of the wave vector in our case is also C_{3v}. We will assume that the vectors **w** and **w***, just as for $\mathbf{k}_\perp$, transform according to the two-dimensional representation. Then the cube of this representation contains only the unit representation.

We will identify as a Ψ point that point on the C_3 axis at which the coefficient in this invariant reduces to zero as a function of k_z. At this point the divergence of the branches has the same form as at the point Z. Therefore the contribution from it to the transverse losses is found to be the same. The contribution to the longitudinal losses is found to be proportional to $\omega^{5/2}$; although it is larger than the contribution from the entire line of symmetry-induced degeneracy, it is nevertheless small compared to the contribution from the line of accidental degeneracy.

We now need to discuss the contribution to the losses from other features at the surface of the Brillouin zone besides the point Z. Since we are investigating the symmorphic groups, there cannot be degeneracies over portions of the surface which have finite area (as shown by Herring,[7] such degeneracies can exist in the presence of a screw axis of second order). Let us clarify whether such a line of degeneracy can exist at the surface.

The class C_{3v} belongs to three symmorphic space groups: C_{3v}^1, C_{3v}^2, and C_{3v}^5. The first two have a hexagonal Bravais lattice: correspondingly, the Brillouin zone has the form of a six-sided prism. The vertical edges of the prism are lines at the surface of the zone having a group of the wave vector with the same high symmetry. In C_{3v}^1 this is the group C_3, which is insufficient to give rise to degeneracy of the phonon spectrum. In C_{3v}^2 this is the group C_{3v}; in this case there is degeneracy: the splitting of the branches along the entire edge is linear; the only exception could be special points of type Ψ, which give transverse losses proportional to $\omega^{5/2}$.

The group C_{3v}^5 has a rhombohedral Bravais lattice. In this case it is possible to have two types of Brillouin zones. In one of these, there are no lines of degeneracy at the surface; in the other, the portion which is a longitudinal line of degeneracy along the C_3 axis extends to the surface of the zone. From this we see that in both of these cases no new contributions arise from including the boundaries of the zone.

Taking into account the results of Ref. 1, we now have enough data to discuss the way the losses depend on temperature over the entire interval of temperature variation. We know that the contribution to the losses from the acoustic branches at temperatures much higher than the Debye temperature is proportional to T^2 for the class C_{3v}. As the temperature increases, the temperature dependence shifts from quadratic to linear.

At the same time, the contributions from the points Z and Ψ (for any branches) and Γ (for the optical branches), which are proportional to lower powers of ω, increase exponentially for low temperatures, and for higher temperatures they necessarily dominate. This implies that these contributions must be comparable to the contributions from the acoustic branches even at some intermediate temperature.

3. REMARKS CONCERNING THE OTHER CRYSTAL CLASSES

In investigating the other classes, just as in investigating the class C_{3v}, we encounter two types of features: those which are analogous to features we discussed previously for the low-temperature case, and those which are fundamentally new. We will not discuss features of the first type, because the necessary analysis is completely analogous to that presented in the previous section and the section of Ref. 1 entitled "Certain Remarks Concerning the Case $T \gtrsim \Theta$"; the corresponding results will be presented below in Table I.

In this section we will limit ourselves to describing those new features which may be encountered in the theory. Let us begin with those which are determined by questions of symmetry. In the previous section certain features—the ends of the symmetry-induced lines of degeneracy—were

TABLE I.

Symmetry class	Losses for the smallest number of lines and points of degeneracy admitted by symmetry		Losses in the presence of additional lines and points of accidental degeneracy	
C_1, C_2	3		2,5	
D_2, O	2,5		–	
C_s	3		2; 2,5	
C_{2v}	2; 2,5		2	
T	2		–	
T_d	1		–	
C_i	5		2; 2,5[C]	
T_h	2		1,5[A]	
O_h	2		–	
	∥	⊥	∥	⊥
C_{2h}	5	5	2; 2,5[C]; 3,5[B]	2; 2,5[B]; 4
D_{2h}	2	4	–	2
S_6	2	2	1,5[A]	–
D_{3d}	4	2	2,5[C]; 3,5[A]	–
C_{4h}	1	2	–	–
D_{4h}	3	2	2; 2,5[C]	–
C_{6h}	1	2	–	–
D_{6h}	5	2	2; 2,5[C]; 3[D]; 3,5[A]	1,5[D]
C_3, D_3	2,5	2[A]	–	–
C_{3v}	2	1,5[A]	–	–
C_4	3	2,5	2[B]; 2,5	–
D_4	3	2,5	2,5	–
C_{4v}	3	2	2; 2,5	–
S_4, D_{2d}	1	2	–	–
C_6	3	2,5	2[B]	2
D_6	5	2,5	3[C]; 4[B]	2
C_{3h}	1	1	–	–
D_{3h}	2	1	1,5[D]	–
C_{6v}	3	1,5[A]	2; 2,5[A]	–

found to be important. However, they will be important only if the divergence of the branches is linear along the entire length of a line and quadratic at the points where it ends; this occurs along a threefold axis (i.e., when the group of the wave vector is C_{3v}). There is such a threefold axis in the Brillouin zone for the class C_{6v}: the lateral edge of the six-sided prism. The behavior of the spectrum and EPP at the ends of this edge is the same as at the point Z in the group C_{3v}. Altogether, we obtain Im $\varepsilon_\perp \propto \omega^{3/2}$.

The contribution from the point Z is of interest even when there is no degeneracy on the axis of symmetry, especially for the classes C_3 and D_3. The frequency splitting in this case has the form

$$\Delta\Omega=\Omega_D[c_1(k_z a)^2+c_2(k_\perp a)^4]^{1/2}$$

(here we include the principal terms which give rise to the effect). From this we find Im $\varepsilon_\perp \propto \omega^2$.

Let us turn to an analysis of accidental degeneracy. Here the following structures are possible: the points of accidental tangency introduced in the previous section; special points on a line of "quadratic" degeneracy, i.e., a degeneracy which is lifted quadratically as we deviate from it, at which the rate at which the degeneracy is lifted follows a power higher than the second (we will call such points points of accidental "hypertangency"); points of accidental degeneracy on a symmetry-induced direction which does not contain a line of degeneracy; and a point of degeneracy at an asymmetric position in k-space (see also Refs. 5,8).

The contributions of Ψ-points turn out to be important for the groups S_6, D_{3d}, T_h, D_{6h}, and C_{6v}. In a number of cases, as with C_{3v}, the contribution from these points varies with frequency $\propto \omega^{3/2}$; however, it is clear from Table I given below that the contribution from such points can involve higher powers of ω (due to vanishing of the electron-phonon potential at such points).

The contribution from points of accidental hypertangency are found to be important for the groups D_{6h} and D_{3h}. Near such points the frequency splitting on a sixfold axis has the form

$$\Delta\Omega=\Omega_D[c_1(ak_\perp)^8+c_2 a^6 k_\perp^4 k_z^2]^{1/2}$$

(here we include the principal terms which give rise to the effect). Calculations based on (1.3) including the corresponding estimates of the EPP from Ref. 1 give the following contributions from such points: for D_{3h},

$$\text{Im}\,\varepsilon_\perp \propto \omega, \quad \text{Im}\,\varepsilon_\parallel \propto \omega^{3/2} \tag{3.1}$$

and for D_{6h},

$$\text{Im}\,\varepsilon_\perp \propto \omega^{3/2}, \quad \text{Im}\,\varepsilon_\parallel \propto \omega^3. \tag{3.2}$$

The contributions of points of accidental degeneracy on axes of higher order are found to be important for the groups C_4, C_6, and D_6. In the vicinity of such points the frequency splitting has the form

$$\Delta\Omega=\Omega_D[c_1(ak_\perp)^4+c_2 k_z^2 a^2]^{1/2}$$

(including the important terms). Taking into account the estimates of the EPP from Ref. 1, in sum we obtain for C_4 and C_6

$$\text{Im}\,\varepsilon_\perp \propto \omega^3, \quad \text{Im}\,\varepsilon_\parallel \propto \omega^2 \tag{3.3}$$

and for D_6

$$\text{Im}\,\varepsilon_\perp \propto \omega^3, \quad \text{Im}\,\varepsilon_\parallel \propto \omega^4. \tag{3.4}$$

A contribution from points of degeneracy on a twofold axis is present for the group C_{2h}. In this case the divergence function and estimates of the corresponding nondiagonal components of the EPP have the form

$$\Delta\Omega=\Omega_D[c_1(k_\perp a)^2+c_2(k_z a)^4]^{1/2},$$
$$|\Lambda_\perp| \sim \Lambda_1, \quad |\Lambda_\parallel| \sim \Lambda_1 k_\perp a,$$

which leads to

$$\text{Im}\,\varepsilon_\perp \propto \omega^{5/2}, \quad \text{Im}\,\varepsilon_\parallel \propto \omega^{7/2}. \tag{3.5}$$

The contribution from isolated points of accidental degeneracy at asymmetric points in k-space in a centrosymmetric crystal coincide with the contribution to Im $\varepsilon_\perp$ for the previous case; in a noncentrosymmetric crystal we have Im $\varepsilon \propto \omega^3$. This contribution can turn out to be important for the groups C_i, C_{2h}, D_{3d}, D_{4h}, D_{6h}, and D_6.

We will not stop to calculate the frequency dependences of the three-phonon losses, as the results of this calculation are presented in Ref. 1.

4. BASIC RESULTS

From what was said above it follows that the most important contribution to the intrinsic dielectric losses (three-phonon, quasi-Debye, and four-phonon) have the following temperature and frequency dependence for $T \gtrsim \Theta$: for the quasi-Debye losses,

$$\text{Im}\,\varepsilon \propto T \frac{\omega\tau}{1+(\omega\tau)^2}. \tag{4.1}$$

(Let us recall that τ is the characteristic lifetime of the phonons which interact to produce the absorption). For the four-phonon losses,

$$\text{Im}\,\varepsilon \propto \omega T^2. \tag{4.2}$$

For the three-phonon case, for $\omega\tau \gg 1$ the loss has the form

$$\text{Im}\,\varepsilon \propto \omega^n T, \tag{4.3}$$

where the exponent n was calculated in the previous sections for a number of examples. The results of the calculation for the crystals in the other symmorphic space groups are summarized in Table I.

In Table I we use the symbols $\parallel$ and $\perp$ to denote the values of Im ε for fields parallel and perpendicular to the maximum symmetry direction, respectively.[2] If a given case necessarily corresponds to several contributions with different values of n, then the table lists the smallest value. If different combinations of contributions are possible, then for each combination the smallest value of n is given. The underlining emphasizes the contributions from those distinctive features of the phonon spectrum which arise or can arise only in the short-wavelength portion of the spectrum. The letters indicate the type of singularity involved: superscript A denotes a point of quadratic frequency splitting on a line of symmetry-induced degeneracy with linear splitting behavior; superscript B denotes a point of accidental degeneracy on a symmetry axis of the Brillouin zone; superscript C denotes a point of accidental degeneracy at a nonsymmetric position in the Briliouin zone; superscript D denotes a point

on a line of degeneracy with quadratic frequency splitting of the branches in which the functional dependence of the splitting has a power higher than the second (i.e., points of hypertangency).

The underlining in the columns of Table I imply that the inclusion of additional features of degeneracy do not give rise to contributions having a smaller value of n than exist without including these features. However, this does not altogether imply the nonexistence of additional accidental degeneracy—even for those cases which are underlined, the presence of accidental degeneracy can lead to an additional contribution having the same temperature and frequency dependence. This will significantly influence the value of the numerical coefficient in front of $T\omega^n$.

Eqs. (4.1) and (4.3), including the data in Table I, completely determine the possible temperature and frequency dependences of the intrinsic losses in noncentrosymmetric crystals: for $\omega\tau \gg 1$ they are determined by the sum of the quasi-Debye and three-phonon contributions; for $\omega\tau \ll 1$ the losses are determined by the quasi-Debye contribution.

The results obtained here also allow us to describe the case of centrosymmetric crystals. Without repeating the corresponding discussions, which are given in our previous papers[9] (see also Ref. 1), let us simply state the results. For $n > 2$ and $\omega\tau \gg 1$ the losses are determined by the sum of three-phonon and four-phonon contributions, while the first of these always determines the high-frequency asymptotic form; for $\omega\tau \lesssim 1$ the four-phonon contribution determines the lossses. For the cases $n \leqslant 2$ and $\omega\tau \gg 1$ the three-phonon contribution determines the losses.

It is important that for $\omega\tau < 1$ we have, as previously, succeeded in isolating a contribution to the losses which is very sensitive to features of the phonon spectrum. We have preserved the name "three-phonon" for this contribution (i.e., it is the low-frequency generalization of the three-phonon contribution); its temperature and frequency dependences can be determined based on the frequency dependence of the three-phonon contribution for $\omega\tau \gg 1$. The variation of the temperature and frequency dependences of the intrinsic losses in centrosymmetric crystals as the frequency decreases are shown in Table II.

Note that for $n \leqslant 2$ a change in the function occurs for frequencies of the field ω on the order of $1/\tau$, while for $n > 2$ the change takes place at frequencies considerably larger than $1/\tau$.

5. COMPARISON WITH EXPERIMENT

A comparison of the qualitative predictions of the theory—i.e., the frequency and temperature dependences, and even better the character of the change in the temperature dependence with frequency—with experiment would be most informative. This possibility is presented to us in the experimental data on microwave absorption of alkali-halide crystals.[10,11] While they do not always coincide in their details, these papers illustrate the most important features of the microwave absorption. At relatively low frequencies Im ε behaves as ωT^2; as the frequency increases the frequency dependence becomes more rapid than ω, while a noticeable linear term appears in the temperature dependence. Furthermore, in their paper Stolen and Dransfeld[11] have noted the presence of a characteristic wavelength λ_c such that the term in the absorption which is linear in temperature becomes significant for $\lambda < \lambda_c$: for LiF $\lambda_c = 0.2$ mm, for KI $\lambda_c = 0.35$ mm, for KBr $\lambda_c = 0.7$ mm, and for NaCl $\lambda_c = 2$ mm.

Let us compare these data with theory. Taking into account that the alkali-halide crystals belong to the symmorphic group O_h^5 of the crystal class O_h and using Table I and II, we see that as the frequency increases the intrinsic losses in these crystals must undergo the following change in their frequency and temperature dependences:

$$T^2\omega \ln \frac{Mv^2}{T} \to T\omega^2$$

at frequencies on the order of $1/\tau$, the damping rate of thermal phonons. The quantity $1/\tau$, estimated on the basis of data from Ref. 12 for alkali-halide crystals, is found to be on the order of 10 cm^{-1}, which corresponds to a wavelength on the order of 1 mm. From this we see that the theory reproduces the most important qualitative features of the experimental data rather well. The absolute value of the observed losses also can be matched with the theoretical estimates.[13] However, a serious comparison of the theoretical and experimental absolute values of the losses is not possible at the present time in view of the insufficiency of available information on the details of the phonon spectrum and the parameters of the anharmonic interactions in the crystals under discussion.

In Ref. 1 we showed how results concerning microwave absorption in leucosapphire for $T \ll \Theta$ could be interpreted within the framework of our theory. As shown by Il'chenko,[14] the absorption in leucosapphire at high temperatures also can be interpreted within the same framework.

6. CONCLUSION

In this paper we have thoroughly analyzed the temperature and frequency dependences of intrinsic losses in insulators for temperatures larger than or on the order of the Debye temperature Θ in crystals belonging to the symmorphic groups.

We have shown that the temperature and frequency dependences of the losses for crystals belonging to symmorphic

TABLE II.

Value of n	Change in the function Im ε as the frequency decreases
$n>2$	$T\omega^n \to T^2\omega$
$n=2$	$T\omega^2 \to T^2\omega \ln \frac{Mv^2}{T}$
$n<2$	$T\omega^n \to T^n\omega$

groups are determined by the crystal classes and not by the space groups.[3] Symmetry influences the losses in two ways: it determines the required set of distinctive features of the phonon spectrum for a given class, and also limits the largest possible set of such features. Table I, which contains the basic results of this paper, is in agreement with this principle.

To each of these temperature and frequency dependences there corresponds an estimate of the order of magnitude of the losses (see Section 2), e.g., Im $\varepsilon \propto T^{\alpha}\omega^{\beta}$ implies that

$$\operatorname{Im}\varepsilon \sim \left(\frac{T}{Mv^2}\right)^{\alpha}\left(\frac{\omega}{\Omega_D}\right)^{\beta}.$$

This estimate is very rough, and is most likely too low. As analysis of a number of specific examples show,[1] usually in this kind of estimate there is an additional numerical factor of order 100. The most informative prediction of the theory is its conclusions about the character of the temperature and frequency dependences with varying frequency.

[1)]In Ref. 1 a short review is given of previous papers on the theory of intrinsic losses in insulators.

[2)]As in Ref. 1, for the group D_{4h} the orientation of the field is indicated with respect to the normal to the symmetry plane, which necessarily contains the acoustic axis in this case.

[3)]Certain quantitative differences are possible among the different symmorphic groups of a single class, e.g., the presence or absence of Ψ-points in the class C_{3v} (see Section 2) and the number of such points.

[1]V. L. Gurevich and A. K. Tagantsev, Zh. Eksp. Teor. Fiz. **91**, 245 (1986) [Sov. Phys. JETP **64**, 142 (1986)].
[2]A. K. Tagantsev, Zh. Eksp. Teor. Fiz. **86**, 2215 (1984) [Sov. Phys. JETP **59**, 1290 (1984)].
[3]V. S. Vinogradov, Fiz. Tverd. Tela (Leningrad) **4**, 712 (1962) [Sov. Phys. Solid State **4**, 519 (1962)].
[4]I. G. Lang and U. S. Pashabekova, Fiz. Tverd. Tela (Leningrad) **6**, 3640 (1964) [Sov. Phys. Solid State **6**, 2913 (1965)].
[5]V. L. Gurevich, *Transport in Phonon Systems* (Amsterdam, North-Holland, 1986), p. 27.
[6]C. Herring, Phys. Rev. **95**, 954 (1954).
[7]C. Herring, Phys. Rev. **52**, 361 (1937).
[8]V. L. Gurevich, Fiz. Tverd. Tela (Leningrad) **21**, 3453 (1979) [Sov. Phys. Solid State **21**, 1993 (1979)].
[9]A. K. Tagantsev, Zh. Eksp. Teor. Fiz. **80**, 1087 (1981) [Sov. Phys. JETP **53**, 555 (1981)].
[10]R. Stolen and K. Dransfeld, Phys. Rev. **139**, 1295 (1965).
[11]J. C. Owens, Phys. Rev. **181**, 1228 (1969).
[12]A. Hadni, *Far Infrared Properties of Solids*, S. S. Mitra (ed.), Plenum, New York (1970), p. 561.
[13]A. K. Tagantsev, Dissertation for Doctorate in Phys. and Math. Sci., I. M. Ioffe Physicotech. Inst., USSR Acad. Sci., 1982.
[14]V. S. Il'chenko, Fiz. Tverd. Tela (Leningrad) **31**, 135 (1989) [Sov. Phys. Solid State **31**(7), 1175 (1989)].

Translated by Frank J. Crowne