Low-frequency dielectric relaxation in centrosymmetric crystals

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(Submitted 30 June 1980)
Zh. Eksp. Teor. Fiz. **53**, 1087–1098 (March 1981)

Lattice losses in an ideal centrosymmetric crystal are considered for electric field frequencies below or of the order of the inverse lifetime of the thermal phonons. It is shown that the rate of dissipation of the electric field energy into heat can be expressed in terms of the nonequilibrium contribution to the off-diagonal (with respect to the spectrum branches) components of the single-particle density matrix, for which an equation is derived. The criteria of applicability of the results are discussed. Some order-of-magntiude estimates are given.

PACS numbers: 77.40. + i

1. INTRODUCTION

The problem of the dissipation of energy of a homogeneous electric field into heat in ideal dielectric crystals has been considered by a number of authors.[1-5] The most complete and detailed study has been that made by Gurevich[4] (see also his book[6]). As applied to centrosymmetric crystals, the thrust of his research is as follows: an electric field of frequency ω, much less than the thermal frequency of the phonons $\bar{\omega}$, i.e.,

$$\omega\ll\bar{\omega},\quad \bar{\omega}=\begin{cases} T/\hbar, & T\ll\Theta \\ \Theta/\hbar, & T\gg\Theta \end{cases} \tag{1}$$

(T is the temperature in energy units, Θ is the Debye temperature) produces transitions between phonon states of the crystal. The thermal phonons are assumed to be well defined, i.e., the inequality

$$\Gamma\ll\bar{\omega} \tag{2}$$

holds for their attenuation.

By virtue of the centrosymmetric nature of the crystals, the indicated transitions are possible only between different branches of the spectrum. The inequality (1) leads to the result that only thermal phonons from the immediate vicinity of the set of degeneracy points of the spectrum can take part in processes of the lowest order in perturbation theory. The contribution of such processes to the imaginary part of the permittivity is determined by the dimensionality of this set and the dependence of the energy gap and of the matrix element of the electric field on the distance in k space (wave vectors) to this set (for small values of this distance). The symmetric character of these properties permitted Gurevich to separate the different types of sets of degeneracy points, to determine the frequency and temperature dependences of the imaginary part of the permittivity corresponding to them, and to elucidate the possibility of realization of these types as a function of the symmetry of the crystal. The formulas obtained by him make it possible in principle to calculate the dielectric losses for $\omega\gg\Gamma$.

Up to the present time there is no theory which allows us to calculate the lattice losses in centrosymmetric crystals at $\omega\lesssim\Gamma$. There is only a recipe[4,5] for obtaining an order-of-magnitude estimate of the imaginary part of the permittivity from its value at $\omega\gg\Gamma$. The present work has as its aim the filling of this gap. It will be shown in the paper that the rate of energy dissipation of an electric field into heat in centrosymmetric crystals is expressed in terms of the nonequilibrium contribution to the off-diagonal (with respect to the spectrum branches) components of the single-particle density matrix, for which an equation is derived. When the equation is applicable, the estimates of Refs. 4 and 5 are valid to no worse than logarithmic accuracy. In the opposite case, at $\omega\lesssim\Gamma$, the dielectric losses are determined by the fourth order of ordinary single-particle perturbation theory.

2. THE HAMILTONIAN

We shall describe the system of phonons interacting with the electric field by the following Hamiltonian:

$$H=H_0+H_{int}+H_E;$$

here H_0 is the anharmonic part of the phonon Hamiltonian,

$$H_{int}=\frac{1}{3!}\sum_{ijk}V^{ijk}(\mathbf{q}_1,\mathbf{q}_2,\mathbf{q}_3)\hat{\xi}_{\mathbf{q}_1}^{i}\hat{\xi}_{\mathbf{q}_2}^{j}\hat{\xi}_{\mathbf{q}_3}^{k},\quad \mathbf{q}_1+\mathbf{q}_2+\mathbf{q}_3=\mathbf{b},$$

where $\xi_q^i=a_{qi}+a_{-qi}^+$ (a_{qi}^+ and a_{qi} are the creation and annihilation operators, respectively, of a phonon of branch i and wave vector $\mathbf{q}$), $\mathbf{b}$ is the vector of the inverse lattice, and $V^{ijk}(\mathbf{q}_1,\mathbf{q}_2,\mathbf{q}_3)$ is the potential of triple interaction of phonons.

For the description of the interaction of phonons with the field, we use the electrophonon-potential vector $\Lambda^{ij}(\mathbf{q})$ introduced by Gurevich;[4] then the part of the Hamiltonian that describes this interaction has the form[1)]

$$H_E=\frac{\hbar}{2}\sum_{i,j,\mathbf{q}}\mathbf{E}\Lambda^{ij}(\mathbf{q})\,(\Omega_i(\mathbf{q})\Omega_j(\mathbf{q}))^{1/2}\hat{\xi}_{\mathbf{q}}^{i}\hat{\xi}_{-\mathbf{q}}^{j}.$$

Here $\mathbf{E}$ is the electric field vector, $\Omega_i(\mathbf{q})$ is the dispersion law of phonons of branch i. For brevity below, we shall use the notation

$$\mathbf{B}^{ij}(\mathbf{q})=\hbar(\Omega_i(\mathbf{q})\Omega_j(\mathbf{q}))^{1/2}\Lambda^{ij}(\mathbf{q}).$$

It is well known that the power $T\dot{S}$ dissipated into heat in a unit volume of the dielectric ($\dot{S}$ is the rate of increase of the entropy density) is expressed in terms of the imaginary part of the permittivity tensor $\eta_{\alpha\beta}(\omega)$:

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$$TS=\frac{\omega}{8\pi}\eta_{\alpha\beta}(\omega)E_\alpha^* E_\beta, \tag{3}$$

$\eta_{\alpha\beta}(\omega)$ can be calculated from the formula of Kubo (see Ref. 7, p. 367):

$$\eta_{\alpha\beta}(\omega)=\frac{4\pi}{v}\,\mathrm{Im}\left\{\frac{i}{\hbar}\int_0^\infty\langle[\hat C_\alpha(t)\hat C_\beta(0)]\rangle e^{i\omega t}dt\right\},$$
$$\hat C_\alpha(t)=\frac{1}{2}\sum_{i,j,\mathbf q}B_\alpha^{ij}(\mathbf q)\hat\xi_{\mathbf q}^{i}(t)\hat\xi_{-\mathbf q}^{j}(t), \tag{4}$$

where $[\ldots]$ denotes the commutator, while $\langle\ldots\rangle$ is the statistical average over the equilibrium distribution with the Hamiltonian H_0+H_{int}, and v is the volume of the crystal.

For calculation of $\eta_{\alpha\beta}(\omega)$ by formulas (4), we shall apply the standard temperature technique (see Refs. 2 and 7, p. 367). We define the bare temperature propagator in the following normalization:

$$G_{ij}^0(\mathbf q,i\omega_n)=\frac{T}{\hbar}\frac{2\Omega_j(\mathbf q)}{\Omega_j^2(\mathbf q)+\omega_n^2}\delta_{ij}. \tag{5}$$

The interaction H_{int} generates three-point vertices. To the right corner of each diagram there corresponds $B^{*ij}(\mathbf q)$, and to the left, $B^{ij}(q)$.

3. SELECTION OF DIAGRAMS AND DERIVATION OF THE EQUATION

In lowest order in anharmonism, $\eta_{\alpha\beta}(\omega)$, which is determined by the imaginary part of the graph a (Fig. 1), is equal to

$$\eta_{\alpha\beta}(\omega)=\frac{2\pi}{T}\,\mathrm{Im}\sum_{i\neq j}\int\frac{d^3q}{(2\pi)^3}B_\alpha^{ij}(\mathbf q)B_\beta^{*ij}(\mathbf q)$$
$$\times G_{ii}^0(\mathbf q,i\omega_{n'})G_{jj}^0(\mathbf q,i\omega_{n''}),\quad -i\omega_{n'}-i\omega_{n''}=i\omega_n\to\omega+i\delta. \tag{6}$$

It is not difficult to obtain from formula (6) the expressions for $\eta_{\alpha\beta}(\omega)$ that are contained in the work of Gurevich.[4]

We now consider the higher orders. As was noted by Leggett and ter Haar,[8] they all contain diverging diagrams. The recipe for calculation of the contribution from such diagrams appeared simultaneously with the formulation of the Feynman technique. According to it, we must sum the most strongly divergent diagrams in each order in the coupling constant. We now make a selection of such diagrams:

1. We shall consider only irreducible diagrams, since it is easy to see that the sum of reducible diagrams of type b (Fig. 1) gives only an anharmonically small correction to the sum of the irreducible diagrams.

2. Among irreducible diagrams of a given order, we shall keep only those containing the maximum number of free sections (sections that intersect only the lines of the principal phonon loop). According to the rule of sections,[9] the imaginary part of the diagram contains a contribution in which the factor $[\Omega_i\pm\Omega_j\pm\omega]^{-1}$ corresponds to each free section. It is just these factors, which vanish near the set of degeneracy points, that guarantee the divergence of the diagram; therefore the most strongly diverging diagram should be sought in the class mentioned above. We note that this class consists of diagrams, in which the propagators are renormalized by not more than a simple polarization loop.

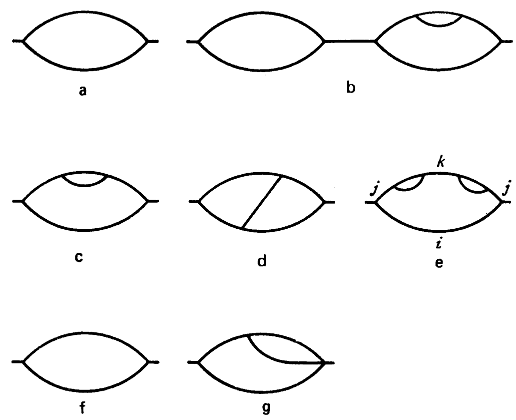

FIG. 1. The Latin letters number the branches of the spectrum corresponding to the propagators (the two vertices in diagram *e* should be connected by one more line.

3. In this class, we can neglect diagrams of the type d (see Fig. 1), which contain corrections to the vertex, in comparison with diagrams of type *c*, which contain corrections to the propagator, since in the first case there is an extra integration at the same rate of vanishing of the denominator. This assertion remains valid in the calculation of diagrams with exact propagators for any relation between ω and Γ. Physically, this selection criterion connected with the fact that only the group of phonons from the immediate neighborhood of the line of degeneracy, where[2)] $|\Omega_i(\mathbf q)-\Omega_j(\mathbf q)|\lesssim\max(\omega_j,\Gamma)$, interacts effectively with the external field.

4. A further narrowing of the class of diagrams considered is possible: to dress the propagator, we shall use only the diagonal part of the simple polarization loop. With diagram e (Fig. 1) as the example, this means that we keep only diagrams with $k=j$. Actually, at $k\neq j$ and $k\neq i$ both near the set of degeneracy points of the branches i and j, and also of i and k, the divergence of the diagram is weaker than in the case $k=j$. Such an argument holds also for more complicated diagrams. Diagrams with $k=i$ can also be neglected; the proof of this fact is given in the Appendix, in view of its cumbersome nature.

Thus, the summation of the most singular terms of all orders of perturbation theory leads to an expression for $\eta_{\alpha\beta}(\omega)$ which differs from (6) only by the replacement of the bare temperature propagators $G_{ii}^0(\mathbf q,i\omega_n)$ by the renormalized $G_{ii}(\mathbf q,i\omega_n)$. Here $G_{ii}(\mathbf q,i\omega_n)$ must be found from the Dyson equation, in which the diagonal part of the simple loop enters as the mass operator.

For the calculations, we make use of the Lehmann representation for the temperature Green's function:

$$G_{ij}(\mathbf q,i\omega_n)=\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{\mathrm{Im}\,G_{ij}^R(\mathbf q,u)}{u-i\omega_n}du. \tag{7}$$

We obtain the retarded Green's function $G_{ih}^R(\mathbf q,u)$ by analytic continuation in frequency of the solution of the

Dyson equation shown above:

$$G_{ij}^{R}(\mathbf{q},\omega)=\delta_{ij}\frac{T}{\hbar}\frac{2\Omega_j(\mathbf{q})}{\Omega_j^2(\mathbf{q})-\omega^2-2i\Omega_j(\mathbf{q})\Gamma_{\mathbf{q}}^{j}(\omega)}. \quad (8)$$

Here $\Gamma_q^j(\omega)$ is the imaginary part of the mass operator after analytic continuation in the frequency; the real part is neglected in writing down (8).

It is now not difficult to obtain the following expression for $T\dot{S}$ by using (3) and (6) with the substitution $G_{ij}^0\to G_{ij}$, and also (7) and (8):

$$T\dot{S}=\frac{\pi\omega}{4\hbar}\sum_{i\neq j}\int\frac{d^3q}{(2\pi)^3}|\mathbf{B}^{ij}(\mathbf{q})\mathbf{E}|^2$$

$$\times\int_{-\infty}^{\infty}[N(x)-N(x-\omega)]A_{\mathbf{q}}^{i}(x)A_{\mathbf{q}}^{j}(\omega-x)\,dx; \quad (9)$$

$$N(x)=\left[\exp\left(\frac{\hbar x}{T}\right)-1\right]^{-1},\quad A_{\mathbf{q}}^{i}(x)=\frac{\hbar}{\pi T}\operatorname{Im}G_{ii}^{R}(\mathbf{q},x).$$

Since (9) was obtained by summation of the diagrams that diverge near the set of degeneracy points of the spectrum, it is natural to expect that the fundamental contribution to (9) will also accumulate near this set, i.e., at q for which $\Delta^{ij}=\Omega_i(\mathbf{q})-\Omega_j(\mathbf{q})\ll\Omega_i(\mathbf{q})+\Omega_j(\mathbf{q})$.

Under this assumption, it is not difficult to obtain[3)]

$$A_{\mathbf{q}}^{i}(x)A_{\mathbf{q}}^{j}(\omega-x)=\frac{1}{\pi}\operatorname{Re}\frac{\delta[x-\tilde{\Omega}(\mathbf{q})]-\delta[x+\tilde{\Omega}(\mathbf{q})]}{-i\omega x/\tilde{\Omega}(\mathbf{q})+i\Delta^{ij}+\Gamma_{\mathbf{q}}^{i}(x)+\Gamma_{\mathbf{q}}^{j}(x)}, \quad (10)$$

where $\tilde{\Omega}(\mathbf{q})$ is the frequency of the loop of the spectrum at the point of degeneracy closest to q.

Substituting (10) in (9) and after simple transformations, we represent the answer in the form

$$T\dot{S}=\frac{T}{2}\sum_{i\neq j}\int\frac{d^3q}{(2\pi)^3}\chi_{\mathbf{q}}^{ij}\frac{I_{\mathbf{q}}^{i}+I_{\mathbf{q}}^{j}}{2}\chi_{\mathbf{q}}^{*ij}, \quad (11)$$

where χ_q^{ij} is determined from the equation

$$[-i\omega+i\Delta^{ij}]\chi_{\mathbf{q}}^{ij}+i\omega\frac{\mathbf{E}\mathbf{B}^{ij}(\mathbf{q})}{T}=-\frac{I_{\mathbf{q}}^{i}+I_{\mathbf{q}}^{j}}{2\tilde{N}(\tilde{N}+1)}\chi_{\mathbf{q}}^{ij}. \quad (12)$$

Here $\tilde{N}=N[\tilde{\Omega}(\mathbf{q})]$, and

$$I_{\mathbf{q}}^{i}=2\Gamma_{\mathbf{q}}^{i}[\Omega_i(\mathbf{q})]N[\Omega_i(\mathbf{q})]\{N[\Omega_i(\mathbf{q})]+1\}$$

is the integrated portion of the linearized collision operator. In writing (11) and (12), we have used the fact that those q are important to us for which $\Delta^{ij}\ll\tilde{\Omega}(\mathbf{q})$.

To identify the quantity χ_q^{ij} we transform (11) with the help of (12) to the form

$$T\dot{S}=\frac{1}{2}\operatorname{Re}\sum_{i\neq j}\int\frac{d^3q}{(2\pi)^3}\tilde{N}(\tilde{N}+1)\chi_{\mathbf{q}}^{ij}D^{*ij}(\mathbf{q}), \quad (13)$$

$$D^{ij}(\mathbf{q})=\frac{\partial}{\partial t}(\mathbf{E}\mathbf{B}^{ij}(\mathbf{q})).$$

On the other hand, the power dissipated into heat per unit volume is the average power obtained by the phonon gas from the electric field $\bar{\dot{U}}$, for which we can write

$$\bar{\dot{U}}=\operatorname{Sp}(\overline{\dot{H}\rho}),$$

where $\hat{\rho}$ is the density matrix, and the bar denotes averaging over the period. In lowest order in the anharmonism the value of the electric field $\bar{\dot{U}}$ can be expressed in terms of the single-particle density matrix $P_{\mathbf{q}}^{ij}=\langle a_{\mathbf{q}i}^{+}a_{\mathbf{q}j}\rangle$. After averaging over the period, we have

$$T\dot{S}=\bar{\dot{U}}=\frac{1}{2}\operatorname{Re}\sum_{i,j}\int\frac{d^3q}{(2\pi)^3}P_{\mathbf{q}}^{ij}D^{*ij}(\mathbf{q}). \quad (14)$$

We see from a comparison of (13) and (14) that $\chi_q^{ij}\tilde{N}(\tilde{N}+1)$ are the nonequilibrium increments to the off-diagonal (with respect to the spectrum branches) components of the single-particle density matrix. We note that in our approximation there is no contribution from the diagonal components to the losses in centrosymmetric crystals since $B^{ii}(\mathbf{q})=0$ in such crystals (see Ref. 4).

4. LIMITS OF APPLICABILITY OF THE EQUATION AND DIELECTRIC LOSSES IN SPECIFIC CRYSTALS

In order that the result obtained in Sec. 3 describe the dielectric losses in a centrosymmetric crystal, we must satisfy two conditions: first, the fundamental contribution to (11) should be made by the small vicinity of the set of degeneracy points[4)] (this was used in fact in the derivation of (11) and (12); second, this contribution should be greater than the contribution from the nondiverging diagrams of fourth order. We now check the applicability of these conditions for all types of sets of degeneracy points selected by Gurevich, thereby obtaining expressions for the imaginary part of the permittivity in the case $\omega\lesssim\Gamma$.

The contribution to the imaginary part of the permittivity from the vicinity of the set of degeneracy points, according to (11), (12) and (3), is

$$\eta_{\alpha\beta}^{s}(\omega)=\frac{4\pi\omega}{T}\sum_{i,j}\int\frac{d^3q}{(2\pi)^3}B_{\alpha}^{ij}(\mathbf{q})B_{\beta}^{*ij}(\mathbf{q})\frac{2\Gamma\tilde{N}(\tilde{N}+1)}{(\omega+\Delta^{ij})^2+4\Gamma^2}; \quad (15)$$

$$\Gamma={}^1/_2\{\Gamma_{\mathbf{q}}^{i}[\Omega_i(\mathbf{q})]+\Gamma_{\mathbf{q}}^{j}[\Omega_j(\mathbf{q})]\}.$$

In the estimating formulas below, we shall understand by Γ the damping of the thermal phonons. We must compare this contribution with the nondiverging contribution of the fourth-order diagrams c and d (see Fig. 1). Such a contribution is made by diagrams for which the phonon spectra, which form the lines of the principal phonon loops, do not have any mutual points of degeneracy. Since these contributions are of the same order, we limit ourselves for comparison with (15), to the regular part of the diagram (Fig. 1):

$$\eta_{\alpha\beta}^{R}(\omega)=\frac{4\pi\omega}{T}\sum_{i,j}\int\frac{d^3q}{(2\pi)^3}B_{\alpha}^{ij}(\mathbf{q})B_{\beta}^{*ij}(\mathbf{q})$$

$$\times N^j(N^j+1)\frac{4\Omega_i^2(\mathbf{q})\Gamma_{\mathbf{q}}^{i}[\Omega_j(\mathbf{q})]}{[\Omega_i^2(\mathbf{q})-\Omega_j^2(\mathbf{q})]^2}. \quad (16)$$

Here $N^j=N[\Omega_j(\mathbf{q})]$.

For brevity in what follows, we group the classification of types of sets of degeneracy points obtained by Gurevich[4] in a table.

The third and fourth columns of this table show the dependence of the gap between the branches Δ^{ij} and the matrix element of interaction with the field B^{ij} on the distance Δq to the set of points of degeneracy as $\Delta q\to 0$. According to Gurevich, the indicated types can be realized in centrosymmetric crystals in the following situations. Type 1—on the boundary of a Brillouin zone in crystals containing second order screw axes. Type

2—on lines of symmetry degeneracy, parallel to the C_3 axis, or on lines of random degeneracy. On the lines of degeneracy parallel to the C_4 or C_6 axes, type 3 is realized for a measuring field orthogonal to the axis, and type 4, for a parallel field. Types 5 and 6 are realized at points of random degeneracy of the spectrum, type 7 at the center of a Brillouin zone.

We begin with the case of a surface (type 1). For calculation of (15), we break up the integration, over d^3q into an integral over the degeneracy surface and over the normal to it $q_\perp$. Since the integral builds up mainly at small values of $q_\perp$, we take into account the dependence on $q_\perp$ only in Δ^{ij} and extend the limits of integration to infinity. Then the integral with respect to $dq_\perp$ of the fraction in (15) is equal to

$$\int_{-\infty}^{\infty} \frac{dq_\perp 2\Gamma}{(\omega+q_\perp \partial\Delta^{ij}/\partial q_\perp)^2+4\Gamma^2} = \frac{\pi}{|\partial\Delta^{ij}/\partial q_\perp|}. \tag{17}$$

Further calculation leads to the result of lowest order perturbation theory. Comparison with $\eta^R_{\alpha\beta}(\omega)$ gives

$$\eta^R/\eta^s \approx \Gamma/\bar{\omega}.$$

Thus, in the given case, the formulas (11) and (12) describe the dielectric losses with power-law accuracy. As a result, $\eta^s_{\alpha\beta}(\omega)$ is given in lowest order perturbation theory for any relation between ω and Γ.

Considering type 2, we shall assume for simplicity the degeneracy line to be a straight line passing through the center of the Brillouin zone. Taking it into account that the basic contribution is made by the **q** which make small angles ϑ with the line, we replace $\sin\vartheta d\vartheta$ by $\vartheta d\vartheta$, and retain the dependence on ϑ only in Δ^{ij}. For the integral of the sum of the two fractions from (15), which differ in the order of the symbols i and j, we have

$$\int_0^\pi \left[\frac{2\Gamma}{(\omega+\vartheta\partial\Delta^{ij}/\partial\vartheta)^2+4\Gamma^2} + \frac{2\Gamma}{(\omega-\vartheta\partial\Delta^{ij}/\partial\vartheta)^2+4\Gamma^2}\right]\vartheta d\vartheta$$
$$= \frac{2\Gamma}{|\partial\Delta^{ij}/\partial\vartheta|^2}\left[\ln\frac{\pi|\partial\Delta^{ij}/\partial\vartheta|^2}{\omega^2+4\Gamma^2} + \frac{\omega}{\Gamma}\operatorname{arctg}\frac{\omega}{2\Gamma}\right].$$

For $\omega \gg \Gamma$, further calculation gives the result of lowest order perturbation theory

$$\eta_{\alpha\beta}{}^s(\omega) = \frac{4\pi^2\omega^2}{T}\sum_{i>j}\int\frac{q^2dqd\varphi}{(2\pi)^3}B_\alpha{}^{ij}(\mathbf{q})B_\beta^{*ij}(\mathbf{q})\frac{\tilde{N}(\tilde{N}+1)}{|\partial\Delta^{ij}/\partial\vartheta|^2}.$$

However, the accuracy is already worse than before:

$$\eta^R/\eta^s \approx \Gamma/\omega.$$

For $\omega \lesssim \Gamma$ we have

$$\eta_{\alpha\beta}{}^s(\omega) = \frac{16\pi\omega}{T}\sum_{i>j}\int\frac{q^2dqd\varphi}{(2\pi)^3}B_\alpha{}^{ij}(\mathbf{q})B_\beta^{*ij}(\mathbf{q})$$
$$\times\frac{\tilde{N}(\tilde{N}+1)}{|\partial\Delta^{ij}/\partial\vartheta|^2}\Gamma\ln\frac{\pi|\partial\Delta^{ij}/\partial\vartheta|}{2\Gamma}. \tag{18}$$

Comparison of (18) and (16) gives

$$\eta^R/\eta^s \approx 1/\ln\frac{\bar{\omega}}{\Gamma}.$$

This means that the diagrams chosen for (11) and (12) are, in the given case only logarithmically large in comparison with the remainder.

From the viewpoint of contributions to the loss, the lines of type 3 are equivalent to lines of type 2, while the lines of type 4 are equivalent points of type 5. Without pausing to prove these assertions, we proceed to the analysis of the points of type 5. It is simple to see that in this case the integral accumulates principally in the immediate vicinity of the degeneracy point only in the case $\omega \gg \Gamma$. In the limit $\Gamma/\omega \to 0$, we get the result of lowest order perturbation theory from (15):

TABLE I

Type	Dimensionality of set	$\Delta^{ij}\sim$	$B^{ij}\sim$
1	surface	Δq	const
2	line	Δq	const
3	line	Δq^2	Δq
4	line	Δq^2	Δq^2
5	point	Δq	const
6	point	Δq, Δq^2	const
7	point	Δq	Δq

$$\eta_{\alpha\beta}{}^s(\omega) = \frac{4\pi^2\omega^3}{T}\int\frac{d^3q}{(2\pi)^3}\sum_{i,j}\delta(\mathbf{q}-\mathbf{q}_{ij})B_\alpha{}^{ij}(\mathbf{q})B_\beta^{*ij}(\mathbf{q})\frac{\tilde{N}(\tilde{N}+1)}{|\partial\Delta^{ij}/\partial\mathbf{q}|^3}. \tag{19}$$

Here q_{ij} are the coordinates of the points of degeneracy. Comparison of (19) and (16) gives

$$\eta^R/\eta^s \approx \Gamma\bar{\omega}/\omega^2.$$

Thus, it is no longer sufficient here that we have the inequality $\omega \gg \Gamma$, as is usual of lowest order perturbation theory predominates, but the following stricter condition is required:

$$\omega \gg (\Gamma\bar{\omega})^{1/2}.$$

At $\omega \lesssim \Gamma$, the vicinity of degeneracy points cannot assure the dissipation of the energy, and formulas (11) and (12) are not applicable. The imaginary part of the permittivity is determined by the regular contribution of the fourth order diagrams.

Consideration of points of types 6 and 7 leads to results only slightly different from those obtained in the previous case: the integral (15) accumulates as before in the vicinity of the point only at $\omega \gg \Gamma$. The second (lower) order in the anharmonism determines the losses for type 6 at

$$\omega \gg (\Gamma^2\bar{\omega})^{1/3},$$

for type 7 at

$$\omega \gg (\Gamma\bar{\omega}^3)^{1/4}.$$

In the case of nonsatisfaction of these criteria, the regular contribution of fourth order diagrams is operative.

We note that in the calculation of the regular contribution of fourth order diagrams, one should take into account not only c and d, but also the diagrams f and g[5] (see Fig. 1). The latter contains vertices that describe the interaction of the electromagnetic field with three phonons. It is easy to see that any graphs containing such vertices drop out of the sequence selected in Sec. 3. Therefore, in order not to distract attention, we have not written out the term in H_E that generates this vertex. The same applies to the four-phonon interaction.

5. CONCLUSION

We have considered the dissipation of energy of a homogeneous electric field, due to interaction with the phonon system of a centrosymmetric crystal. A characteristic feature of this system is the absence of a linear term in the expansion of the phonon frequency in ters of the applied electric field. Therefore, with the help of the kinetic equation (the Akhiezer mechanism), it is possible to describe nonlinear losses only. In the description of the linear losses, we can distinguish two cases: 1) the energy dissipation takes place mainly in a small fraction of the excited volume of k space; 2) the dissipation takes place uniformly throughout this volume.

In the first case, the role of the kinetic equation is played by the algebraic equation (12) obtained in Sec. 3 for the nonequilibrium increments to the off-diagonal components of the density matrix.[6] The fact that the equation, unlike the kinetic equation, does not contain an integral part, is a manifestation of the narrowness of the group of phonons taking part in the absorption. This equation allows us to calculate the dielectric losses at $\omega \lesssim \Gamma$ and to establish strict criteria for the applicability of the results of lowest order perturbation theory. A comparison of the results for $\omega \lesssim \Gamma$ with the estimates obtained in Refs. 4 and 5 shows the validity of the latter with accuracy to $\ln(\bar{\omega}/\Gamma)$.

In the second case, the rate of energy dissipation is no longer dictated by the equation describing the collective motion of the phonons, but is determined by the fourth-order single-particle perturbation theory (see Footnote 5).

As was shown in Sec. 4, the first case is realized at $\omega \lesssim \Gamma$ for the first three types of sets of degeneracy points of the phonon spectrum of the crystal (see the table). We obtain estimating formulas for the contribution to the imaginary part of the permittivity for these types at $\omega \lesssim \Gamma$. In obtaining these estimates, we shall assume that the basic contribution is made by thermal phonons, i.e., we shall take the thermal values of the integrand quantities outside the integral sign. According to Gurevich[4], the thermal value of the electro-phonon potential vector is

$$|\Lambda^{ij}| \approx \hbar\bar{\omega}/\rho^{1/2} w\Theta, \tag{20}$$

where ρ is the density of the crystal and w is the mean sound speed. For the contribution from the degeneracy surface of type 1, using (15), (17) and (20), we have

$$\eta'' \approx \frac{\omega T \Theta^2}{\rho w^5 \hbar^2}, \quad (T \gg \Theta). \tag{21}$$

It has been assumed here that near the degeneracy surface

$$|\partial\Delta^{ij}/\partial q_\perp| \approx w.$$

Considering the contribution from the line of type 2, we shall assume that Δ^{ij} is not anomalously small. For thermal phonons traveling almost to such a line, this leads to the estimate

$$|\partial\Delta^{ij}/\partial\vartheta| \approx \bar{\omega}. \tag{22}$$

Assuming that the contributions from all lines of type 2 are of the same order, we obtain for their total contribution, using (18), (20) and (22),

$$\eta'' \approx n \frac{\omega T \Theta}{\rho w^5 \hbar} \left(\frac{\hbar\bar{\omega}}{\Theta}\right)^3 \Gamma \ln\frac{\bar{\omega}}{\Gamma}, \tag{23}$$

where n is the number of lines of type 2. As has been shown above, the contributions from the line of type 2 and the line of type 3 are of the same order. Thus, formula (23) gives the total contribution from lines of both types if by n we mean their total number.

Gurevich[4] has obtained still another estimate—the estimate for the contribution from the random degeneracy surface in a hexagonal crystal. The surface existed only in the approximation of elasticity theory, i.e., for thermal phonons its place is taken in fact by a gap of order $\hbar\bar{\omega}^2/\Theta$. In obtaining this estimate,[4] the gap has been neglected. It is seen from the analysis of (15), that this is permissible only in the case

$$\Gamma \gg \hbar\bar{\omega}^2/\Theta. \tag{24}$$

It is not possible to satisfy (24) without invoking additional sources of scattering and, generally speaking, we must take the gap into account. Allowance for the gap leads to vanishing of the localized contribution in the case of complete lifting of the degeneracy. If degeneracy lines from the surface now remain, then, as it is not difficult to obtain, their contribution is $\Theta/\hbar\bar{\omega}$ times greater than that given by formula (23), in which by n is now meant the number of these lines.

For formula (23), which describes the most widespread situations, we present a numberical estimate for $T \approx \Theta \approx wa \approx 300$ K, $\omega \approx \Gamma \approx T\Theta/\hbar M w^2$ (M is the average atomic mass, a is the lattice constant):

$$\eta'' \approx n \left(\frac{\Theta}{Mw^2}\right)^3 \ln\frac{Mw^2}{\Theta} \approx (10^{-6}-10^{-7})\, n. \tag{25}$$

In obtaining (25), it was assumed that $Mw^2 \approx 10^4 - 10^5$ K. We note that in high-symmetry crystals, n (the number of degeneracy lines belonging to the excited part of the spectrum and participating in the absorption) can reach values of 20–30 at $T \geq \Theta$.

All the estimates above refer to crystals with permittivity ε of the order of several units. The consideration for ferroelectrics of the displacive type, where ε can reach values of a thousand and higher, leads to a quantitatively different result. Thus, in the work of the author[5], the contribution to losses from a line of degeneracy of type 2 for $SrTiO_3$ was estimated at $T = 90$ K, $\omega = 2.2 \times 10^{10}$ Hz, $\Gamma \approx 10^{11}$ Hz:

$$\Delta\, \mathrm{tg}\, \delta \approx \eta''/\varepsilon \approx 1.4 \cdot 10^{-5}.$$

At this temperature in $SrTiO_3$, there are in operation at least 12 symmetric lines of degeneracy of branches of the soft mode.[5] Taking this into account and also the factor $\ln(\bar{\omega}/\Gamma)$ omitted previously[5] we have for the total contribution to the tangent of the angle of dielectric losses:

$$\mathrm{tg}\, \delta \sim 0.5 \cdot 10^{-3},$$

which agrees in order of magnitude with the experimentally found $\tan \delta = 1.6 \times 10^{-3}$.[12]

We thus see that the contribution to dielectric losses from energy dissipation processes taking place near the set of points of degeneracy of the spectrum can be amenable to experimental observation even at $\omega \lesssim \Gamma$. Estimates for $SrTiO_3$ show that this contribution can in a number of cases turn out to be quite substantial.

In conclusion, we note that, together with the contributions that we have considered, there exists a contribution from four-quantum processes, which differs parametrically from the contribution (23) only by the absence of the factor $\ln(\bar{\omega}/\Gamma)$ (see Sec. 4). It is evident that it is not possible experimentally to detect such a difference. A method for distinguishing these contributions from one another has been proposed by the author in a previous paper,[5] in which it was proposed to carry out measurements of $\tan\delta$ in the presence of a constant electric field, and to distinguish the contributions according to their field dependences.

The author is grateful to B. D. Laĭchtman for a useful discussion, to Yu. M. Galperin and V. L. Gurevich for reading the manuscript and for a number of valuable comments.

APPENDIX

The purpose of this Appendix is to show that diagrams containing even a single free section with identical phonon lines are not subject to selection. For proof, we make use of the fact that potentials of a centrosymmetric crystal are odd:

$$V^{ijk}(\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3) = -V^{ijk}(-\mathbf{q}_1, -\mathbf{q}_2, \mathbf{q}_3), \ \mathbf{B}^{ij}(\mathbf{q}) = -\mathbf{B}^{ij}(-\mathbf{q}).$$

We prove this by rewriting H_{int} in terms of the displacement of the atoms R_n and the anharmonic tensor $D^{\gamma_1\gamma_2\gamma_3}_{\mathbf{n}_1\mathbf{n}_2\mathbf{n}_3}$ (γ enumerates the cartesian coordinates of the displacements and the atoms in the cell, $\mathbf{n}$ numbers the cells):

$$H_{int} = \frac{1}{3!}\sum_{\gamma,\mathbf{n}} D^{\gamma_1\gamma_2\gamma_3}_{\mathbf{n}_1\mathbf{n}_2\mathbf{n}_3} R^{\gamma_1}_{\mathbf{n}_1} R^{\gamma_2}_{\mathbf{n}_2} R^{\gamma_3}_{\mathbf{n}_3}.$$

The invariance of H_{int} relative to spatial inversion makes directly anharmonic tensor odd:

$$D^{\gamma_1\gamma_2\gamma_3}_{\mathbf{n}_1\mathbf{n}_2\mathbf{n}_3} = -D^{\gamma_1\gamma_2\gamma_3}_{-\mathbf{n}_1-\mathbf{n}_2-\mathbf{n}_3}. \qquad (A.1)$$

We now express $V^{ijk}(\mathbf{q}_1\mathbf{q}_2\mathbf{q}_3)$ in terms of the anharmonic tensor and the eigenvectors of the dynamic matrix $e^j_\gamma(\mathbf{q})$, which, thanks to the centrosymmetry of the crystal, can be chosen to be even in $\mathbf{q}$:

$$V^{ijk}(\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3) = \sum_{n,\gamma} D^{\gamma_1\gamma_2\gamma_3}_{\mathbf{n}_1\mathbf{n}_2\mathbf{n}_3} e_{\gamma_1}{}^i(\mathbf{q}_1) e_{\gamma_2}{}^j(\mathbf{q}_2)$$

$$\times e_{\gamma_3}(\mathbf{q}_3)\exp[-i(\mathbf{n}_1\mathbf{q}_1 + \mathbf{n}_2\mathbf{q}_2 + \mathbf{n}_3\mathbf{q}_3)]. \qquad (A.2)$$

It is easy to see that (A.2), with account of (A.1), defines an odd potential. The oddness of the electro-phonon potential is proved in similar fashion.

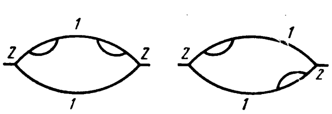

FIG. 2. The numbers denote the branches of the spectrum corresponding to the propagators. These diagrams differ by a relative turn of the lateral blocks.

We proceed directly to the proof. Let us consider a diagram of order $2m$, satisfying the previously formulated selection principles and containing a single free section with idential phonon lines. It is not difficult to establish the fact (by using the section rule of Ref. 9) that its imaginary part is actually of order $2m-2$, but also contains the factor Γ/ω. At $\omega \ll T$, such a diagram would be subject to selection, since, for a diagram of order $2m-2$ it contains the maximum number of divergences. However, let us consider a diagram which differs from the first by the relative turn of the lateral blocks (Fig. 2). At $\omega = 0$, the inverted block differs in sign from the uninverted one, since it contains an odd number of vertices that are odd in the momentum, while the temperature propagators are even both in momentum and in frequency. Consequently, the sum of the block and its inversion is equal to zero. It is not difficult to show that at $\omega \ll T/\hbar$ this sum has the smallness of $\hbar\omega/T$. Thus, the sum of diagrams that differ in the mutual inversion of the blocks, has the order $2m-2$ and the factor $\hbar\omega/T$, i.e., is of order $2m$. Consequently, such diagrams are not subject to selection, since for order $2m$ they do not contain the maximum number of divergences. The proof of the smallness of the sum of diagrams containing several free sections with identical phonon lines is similar, but it requires repeated iteration of the procedure of inversion of the blocks.

1) It will be shown below that allowance in H_E for terms describing the interaction of the field with three and more phonons leads to some quantitative changes.

2) We note that upon absorption of longitudinal sound of frequency ω by longitudinal acoustic phonons at $\omega \gg \Gamma$ only a narrow group of phonons is removed from equilibrium, which premitted Shklovskiĭ[9] to neglect also the corrections to the vertex at $\omega \gg \Gamma$.

3) We shall not linger on the derivation of (10), since an analogous formula has been obtained by Holstein [10] for $A_q{}^i(x)A_{q'}{}^i(\omega - x)$.

4) All the considerations that follow pertain to the neighboring regions that belong to the excited part of the spectrum.

5) The contribution of diagrams of this order has been considered in detail by Balagurov, Vaks, and Shlovksiĭ.[3]

6) We note that an equation similar to (12) was obtained earlier by Gurevich in the description of the growth of fluctuations in a semiconductor with acousto-electric instability.[11]

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Translated by R. T. Beyer