cesses 2⇄1. These mechanisms can compete, because the former has the anharmonicity as a small factor, and the second (for $k_T > k^{(2)}$) has a small phase volume.

5)The exact expression for $b(q_1, q_2, q_3)$ is cumbersome, and depends not only on the wave vectors but also on the frequency arguments $\omega_1, \omega_2, \omega_3$. It is derived in the Appendix.

6)The last term in the expression (20) differs from the analogous term given in Refs. 19 and 20; the latter term contains a matrix element $b(\mathbf{k}, ck; \mathbf{k}', ck'; -\mathbf{k}-\mathbf{k}', ck+ck')$, which does not coincide with the term in (20) even when $\mathbf{k}$ and $\mathbf{k}'$ are parallel. The difference is due to the fact that when one constructs the diagram technique for helium II with one type of bare vertices and Green's functions the bare vertices must depend on the frequency arguments as well as on the momenta; this was not taken into account in Refs. 19 and 20.

7)It is interesting to note that a spectrum of precisely the same type is obtained not only in hydrodynamics but also in the direct study of a weakly nonideal Bose gas with short-range forces.[21] Kemoklidze and Pitaevskiĭ[22] and Feenberg[23] have shown that inclusion of the long-range van der Waals forces can lead to the appearance of a k^4 term.

8)We call attention to the fact that in this respect the situation is different from that which is usual for solids. In them, the quantization is usually carried out in a Lagrangian (comoving) coordinate system, and therefore the anharmonic terms come only from the potential energy of the lattice vibrations. On the other hand, the quantization of the phonons in helium II is done in an Eulerian (laboratory) system,[27] and therefore the anharmonic terms contain contributions from the kinetic energy also.

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Effect of a weak electric field on the dielectric losses in centrosymmetric ferroelectrics of the displacement type

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Two mechanisms whereby a constant homogeneous electric field affects the losses due to lattice anharmonicity of an ideal crystal are considered. These are: 1) partial suppression of the already present processes of absorption of a measuring-field quantum with participation of two phonons from different modes, and 2) the appearance of new processes that cause absorption of this quantum, with participation of two phonons from the same mode. The influence of the finite phonon damping is discussed. A threshold is predicted for the field dependence of the first mechanism. The values of the threshold fields are estimated and their frequency dependences are obtained.

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1. INTRODUCTION

The question of dielectric losses in ferroelectrics of the displacement type was considered by a number of workers.[1–8] A detailed analysis of the losses in a centrosymmetric cubic ferroelectric in a homogeneous alternating electric field, based on the papers of Balagurov *et al.*,[4,5] is contained in Vaks's monograph.[9] The results for noncentrosymmetric cyrystals in inhomogeneous electric fields were obtainedby Balagurov and Vaks.[6,7] All the authors considered the contribution made to the losses by the lattice anharmonicity

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of the ideal crystal. The same contribution to the losses in centrosymmetric crystals is of interest to us, but in the presence of a weak constant homogeneous electric bias field E.

By weak bias field we mean an electric field that causes a small relative change of the frequencies in the phonon spectrum of the ferroelectric. A small relative change $\Delta\omega_0/\omega_0$ of the limiting frequency ω_0 of the "soft" optical mode will correspond to a small relative change of the dielectric constant, i.e., we consider fields that do not lead to noticeable dielectric nonlinearity of the ferroelectric. We consider dielectric losses in a homogeneous measuring field at frequencies $\omega \ll \omega_0$.

The gist of the influence of the field on the losses can be explained with a simple model. We assume that the ferroelectric has the phonon spectrum shown in Fig. 1a. We assume also that the substance is elastically isotropic, so that a point of intersection of the modes in Fig. 1a can exist for any direction of the wave vector **k**, since the longitudinal mode will not interact with the transverse one.

The dielectric loss is determined by the extent to which the lattice vibrations with such a spectrum can absorb the quanta of the measuring-field with energy $\hbar\omega$ and with wave vector equal to zero. It is easily seen that in our model only the following absorption processes are possible:

$$\begin{aligned} &\hbar\omega+\hbar\omega_l\to\hbar\omega_c, \\ &\hbar\omega+\hbar\omega_c\to\hbar\omega_l. \end{aligned} \quad (1)$$

Only these processes contribute in fact to the loss. We note, getting ahead of ourselves, that allowance for the damping of the modes should not change substantially the contribution of these processes.

We now apply the bias field. The inversion center of the system will vanish, an induced piezoelectric effect will appear, and the modes ω_c and ω_l will begin to interact and move apart. The spectrum takes the form shown in Fig. 1b. It is easily seen that the shortest distance between the ω_s branches is proportional to the bias field. In fact, when the degeneracy is lifted the energy gap is proportional to the modulus of the matrix element of the perturbation, while the matrix

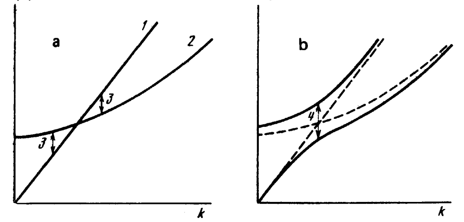

FIG. 1. a) Isotropic spectra of longitudinal acoustic $\omega_l(k)$ (1) and soft transverse optical $\omega_c(k)$ (2) phonon modes of a ferroelectric in the presence of a sphere of their random degeneracy. b) The same spectra after applying a bias electric field. The arrows (3) mark the frequency ω of the measuring field. The arrow (4) marks the minimum distances between the ω_s modes that were pushed apart. The drawing is not to scale.

element of the first power of the field differs from zero, inasmuch as the transitions (1) are possible. Use the term "opening" for the bias field at which $\overline{\omega}_s \approx \max(\omega, \Gamma)$. Here $\overline{\omega}_s$ is the value of ω_s averaged over the directions of the wave vector and Γ is the average value of the half-width of the modes. It is clear that in fields exceeding the opening value the processes (1) cannot take place and cannot contribute to the losses. More accurately speaking, the contribution to the losses will be made only by those transitions in which the wave vectors of the phonons are almost parallel to the bias field (the piezoelectric coupling vanishes for phonons with wave vectors parallel to the field), whereas in the absence of the field the contribution is made by phonons with arbitrary orientation of the wave vector.

The action of the electric field is not limited to distorting the spectrum: other processes with participation of a quantum $\hbar\omega$ become allowed in the system, for example

$$\hbar\omega+\hbar\omega_c\to\hbar\omega_c. \quad (2)$$

Obviously, such a process is possible only when account is taken of the widths of the modes. It can have a phase volume much larger than the previously considered processes. For example, at $\Gamma \gtrsim \omega$ this process can occur on optical modes at all values of the wave vector **k**, whereas processes of type (1) are possible only near mode-intersection points. We note that the contribution made to the losses by the process (2) in weak fields should be proportional to $\mathbf{E}^2$ by virtue of the presence of an inversion center in the initial crystal.

Figure 2 shows schematically the dependence of the contributions of processes (1) and (2) to the losses on $\overline{\omega}_s \sim E$. From an examination of our simple model it is seen that the dependence of the losses on the bias field can be nonmonotonic. Furthermore, if $\Gamma \ll \omega$, the nonmonotonicity will be observed at values of E for which $\overline{\omega}_s(E) \approx \omega$.

Generally speaking, one can expect this nonmonotonicity to depend on the mutual orientation of the fields. If they are parallel, the same interaction ensures absorption of the quantum $\hbar\omega$ and mutual repulsion of the modes. On the other hand if these fields are perpendicular, these interactions are different and the second of them may generally speaking not occur. Thus, one can expect a larger nonmonotonicity when the bias and measuring fields are parallel.

The purpose of the present study is to trace, using a more or less realistic spectrum of the ferroelectric, those features of the phenomenon in question which were noted in the symbol model, namely:

a) the bias field decreases the contribution made by processes of type (1) to the losses;

b) the contribution to the losses from processes of type (2) in weak bias fields can be appreciable;

c) a bias field parallel to the measuring field acts more strongly on the contribution from processes of

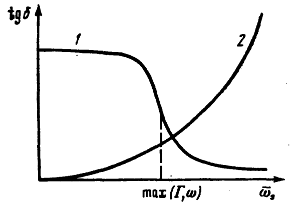

FIG. 2. Curve 1 shows the schematic dependence of the contribution of the processes of type (1) to the tangent of the dielectric loss angle on $\bar{\omega}_s \sim E$, while curve 2 shows the same for processes of type (2).

type (1) than a perpendicular field.

We note that although we use essentially such a feature of displacement-type ferroelectrics as the soft mode, it is nevertheless easy to verify that all the qualitative conclusions remain valid for all centrosymmetric polar dielectrics.

2. TRANSITIONS BETWEEN DIFFERENT MODES

For a measuring-field quantum to be able to cause a transition between different modes, the energy difference between the modes at some point of $\mathbf{k}$-space must be anomalously low. This can occur with a nonzero probability only near the point of intersection of these modes. In the example considered in the introcuction, in the absence of the bias field the modes intersected on a sphere in $\mathbf{k}$-space, and in the presence of a field only at two points. The resultant change of the phase volume of the process was in fact responsible for the decrease of its contribution to the losses. In a real anisotropic crystal the degeneracy of the modes is possible in isolated points and along a certain line of the Brillouin zone.[9,10] The bias field lifts this degeneracy in part, and it is this which leads to a decrease of the contribution to the losses from transitions between different modes.

We consider for simplicity a cubic ferroelectric in the paraphase at a temperature T lower than the Debye temperature Θ. This enables us to consider only the long-wave section of five low-lying modes of the spectrum: three acoustic and two soft transverse optical ones.[9] We confine ourselves to the case when the bias field is directed along a fourfold axis. Then the dielectric tensor has cylindrical symmetry. This allows us to consider only two mutual orientation of the fields, parallel and perpendicular. Thus, we need the long-wave spectrum of the low-lying modes of a cubic ferroelectric in a constant electric field directed along a fourfold axis. The required spectrum will coincide with the spectrum of a cubic ferroelectric in the tetragonal ferroelectric phase (which was analyzed in detail in Secs. 35 and 36 of Ref. 9[1)]), if we take in the latter the polarization to mean the spontaneous polarization and not the one induced by the bias field, and if the limiting frequency of the soft mode is taken at a temperature T.

In view of the unwieldy form of the dispersion equation, we shall not write it out and present only the results of its analysis. In this spectrum, an isolated degeneracy point is present only at the center of the Brillouin zone. The contribution of the attachment processes, as well as the decay of the quantum $\hbar\omega$ in the phonons, processes which occur near this point, can be neglected according to Vaks[9] relative to the parameter ω/ω_0 of $\hbar\omega/T$. It remains to consider the degeneracy lines.

a) Directions of the [100] type. In the absence of a field along these directions, the transverse acoustic and optical modes are pairwise degenerate. The frequency differences between the modes depend quadratically on the distance to the degeneracy line, while the matrix element of the interaction, which ensures absorption of the quantum $\hbar\omega$, vanishes in these directions.[9] Such an interaction can be only the anisotropic part of the electrostriction potential, inasmuch, as shown by Vaks,[9] the triple anharmonicity of the optical displacements makes no contribution to the losses in a centrosymmetric crystal, and the isotopic electrostriction invariant cannot connect two different transverse vibrations and a vector.[2)] The angle part of the matrix element of the anisotropic electrostriction potential is of the form[9]

$$\sum_{\sigma=1}^{3} (\mathbf{v}_0\mathbf{e}_\sigma)(\mathbf{v}_\mathbf{k}^o\mathbf{e}_\sigma)(\mathbf{v}_{-\mathbf{k}}^a\mathbf{e}_\sigma)(\mathbf{k}\mathbf{e}_\sigma), \tag{3}$$

where $\mathbf{v}_0$ is a unit vector in the direction of the measuring field, $\mathbf{v}_\mathbf{k}^o$ is a unit vector of the optical component of the phonon participating in the absorption, $\mathbf{v}_\mathbf{k}^a$ is the unit vector of the acoustic component of the other phonon participating in the absorption, $\mathbf{k}$ is the wave vector of the phonons, and $\mathbf{e}_\sigma$ are the unit vectors of the principal axes of the crystal.

Let the measuring field be directed along [100]. Then we can conclude from an examination of (3) that the main contribution to the losses is made by processes in which the phonon wave vectors are close in direction to [010] and [001]. The point is that for these directions the matrix element is proportional to the sine of the angle between the direction and the wave vector, while for [100] it is proportional to the square of the sine, since $\mathbf{v}_\mathbf{k}^o$ and $\mathbf{v}_{-\mathbf{k}}^a$ are almost transverse. Therefore the contribution from the [010] and [001] degeneracy lines contains, according to Vaks,[9] a small factor ω/ω_0 or $\hbar\omega/T$, whereas the contribution from the [100] line contains the square of this factor.

We apply the bias field parallel to the measuring field. The degeneracy in the directions [010] and [001] is lifted, and the energy gap is proportional to the square of the bias field (the matrix element of the first power of the field is zero for these directions). When this gap becomes much less than ω, the processes stop (the damping of the branches is disregarded for the time being). In the [100] direction the degeneracy still remains, but as already indicated the contribution from this direction is small. Let now the bias field be directed along [010]. This eliminates now only the direction [001], and the contribution to the losses in a field exceeding the "opening" value decreases by one-half.

b) Directions of [111] type. Along these directions there is also degeneracy of the transverse branches. The frequency difference now depends linearly on the distance to the degeneracy line, but the matrix element that ensures the absorption no longer vanishes. The contribution to the losses will contain, according to Vaks,[9] the same small quantity ω/ω_0 or $\hbar\omega/T$ as before. All these lines make equal contributions, and a bias field in any direction always lifts the degeneracy. The gap is now proportional to the first power of the field, since the above-indicated matrix element does not vanish on these directions.

c) Lines lying in phanes of the (110) type. In planes of the (110) type are possible lines of accidental degeneracy longitudinal and optical modes (the unit vector of the displacements of the optical branch is normal to this plane). Arguments similar to those considered in items (a) and (b) can determine the lines that make the principal contribution to the losses in the absence of a bias field in various directions. The results are listed in the table.

d) Lines lying in planes of the type (100). In (100)-type planes are possible lines of random degeneracy similar to the lines in (110). The identification of the lines that make the principal contribution, and the dependence of this contribution of the bias field, can be obtained as in the preceding items. The results are also given in the table.

3. INFLUENCE OF DAMPING OF THE BRANCHES ON THE CONTRIBUTION MADE TO THE LOSSES BY TRANSITIONS BETWEEN DIFFERENT BRANCHES

We have analyzed above transitions in the lowest order in the anharmonicity between the undamped phonon states. How does allowance for higher orders in anharmonicity or, in other words, damping of the branches, affect the result? We consider it advisable to consider this question in the less rigorous but more instructive language of phonon branches of finite width. We stipulate here that we are considering only well defined phonon states, i.e., mode widths Γ much smaller than the average distance between the modes of the long-wave region of the spectrum $\overline{\omega} \approx \omega_0$.

TABLE I.

	Bias field			
	Absent	Directed along [100]	Directed along [010]	Type of degeneracy
a	[010] [001]	No	[010]	Symmetry
б	[111] $[\bar{1}11]$ $[1\bar{1}1]$ $[11\bar{1}]$	No	No	Symmetry
в	(110) $(1\bar{1}0)$ (101) $(10\bar{1})$	No	(101) $(10\bar{1})$	Random
г	(100)	No	(100)	Random

Note. The different degeneracy lines, from the point of view of contribution to the losses, are not equivalent. The table gives the directions (or planes) to which belong the degeneracy lines corresponding to the largest contributions of processes of type (1) to the losses, for measuring fields parallel to [100]. Lines a, b, c, and d correspond to the subsections of Sec. 2. In the second and third column, the bias field exceeds the opening field.

We begin the analysis with the case when the electric-field matrix element does not vanish in almost the entire set of mode degeneracy points [Sec. 2(a) is excluded from consideration]. Then the dependence of the imaginary part of the dielectric constant on Γ is determined by the dependence of the phase volume of the processes on Γ. The following statement can be made concerning the phase volume of the process, namely that it depends on Γ at $\Gamma \gg \omega$ in the same manner as it depends on ω at $\omega \gg \Gamma$. This can be easily verified by calculating the quantity L, which is proportional to the phase volume of the process $\hbar\omega_{\mathbf{k}j} + \hbar\omega \to \hbar\omega_{\mathbf{k}i}$ (i and j designate different branches):

$$L=\int_{-\infty}^{\infty} dx_1 \int_{-\infty}^{\infty} dx_2 \int d^3k\, P_\Gamma(x_1-\omega_{ki}) P_\Gamma(x_2-\omega_{kj})\, \delta(x_1-x_2-\omega)$$
$$=\int_{-\infty}^{\infty} dx_2 \int d^3k\, P_\Gamma(x_2-\omega_{ki}+\omega) P_\Gamma(x_2-\omega_{kj}), \tag{4}$$

where $P_\Gamma(x) = P_0(x/\Gamma)/\Gamma$, and $P_0(x)$ is bell-shaped with a symmetry axis $x=0$ and a width of the order of unity; P_Γ and P_0 are normalized as follows.

$$\int_{-\infty}^{\infty} P_\Gamma(x)\,dx = \int_{-\infty}^{\infty} P_0(x)\,dx = 1. \tag{5}$$

In the isotropic case, when $\omega_{\mathbf{k}j} = \omega_j(k)$, we have

$$L \approx 4\pi \int_{-\infty}^{\infty} dx_2 \int_0^{\infty} k^2\,dk\, P_\Gamma\left[x_2-\omega_i(k_0)-\frac{\partial\omega_i(k)}{\partial k}\bigg|_{k=k_0}(k-k_0)+\omega\right]$$
$$\times P_\Gamma\left[x_2-\omega_j(k_0)-\frac{\partial\omega_j(k)}{\partial k}\bigg|_{k=k_0}(k-k_0)\right]$$
$$=4\pi k_0^2 \int_{-\infty}^{\infty} dr \int_{-\infty}^{\infty} dt\, P_0(r)P_0(t) \Big/ \left|\frac{\partial\omega_i(k)}{\partial k}-\frac{\partial\omega_j(k)}{\partial k}\right|_{k=k_0}$$
$$=4\pi k_0^2 \Big/ \left|\frac{\partial\omega_i(k)}{\partial k}-\frac{\partial\omega_j(k)}{\partial k}\right|_{k=k_0}, \tag{6}$$

since $\omega, \Gamma \ll \overline{\omega}$, $\omega_i(k)$, and $\omega_j(k)$ were expanded in a series about k_0 determined from the equation $\omega_i(k_0) = \omega_j(k_0)$. In the calculation of (6) we made the following changes of variables:

$$t=\frac{1}{\Gamma}\left[x_2-\omega_j(k_0)-\frac{\partial\omega_j(k)}{\partial k}\bigg|_{k=k_0}(k-k_0)\right],$$
$$r=\frac{1}{\Gamma}\left[x_2-\omega_i(k_0)-\frac{\partial\omega_i(k)}{\partial k}\bigg|_{k=k_0}(k-k_0)+\omega\right]$$

and took into account the rapid decrease of $P(x)$ as $x \to \infty$. As expected, L turned out to be proportional to the surface of the k-space sphere near which the attachment process takes place.

For the anisotropic case we carry out the estimate using Sec. 2(c) as an example. The mode frequency difference just as in Secs. 2(b) and 2(d), depends here linearly on the distance to the degeneracy line. Let the degeneracy line be closed; then the surface on which the process takes place can be approximated by a torus in k-space, with a major radius of the order of ω_0/w, where w is the speed of sound, and the minor radius of the order of $\max(\omega, \Gamma)/w$. It is no longer possible to expand $\omega_{\mathbf{k}j}$ around the degeneracy line as before, in a Taylor series, since $\omega_{\mathbf{k}j}$ is not differentiable there with respect to $\mathbf{k}$; however, knowing that the mode frequency difference depends linearly on the distance to the degeneracy line, we can write, in order of magnitude,

$$\omega_{\mathbf{k}j} \approx \omega_0 + A'|\mathbf{k}-\mathbf{k}_0|, \quad \omega_{\mathbf{k}j} \approx \omega_0 + A''|\mathbf{k}-\mathbf{k}_0|, \tag{7}$$

where $A', A'' \approx w, k_0 = \omega_0/w$; $\mathbf{k}_0$ lies in the same plane, perpendicular to the plane of the torus, as $\mathbf{k}$.

Calculations similar to (6) and the use of (7) yields for L a result that differs from (6) in that the surface area of the sphere is replaced by the surface area of the torus:

$$L \approx \frac{1}{w} 4\pi^2 k_0 \begin{cases} \Gamma/w, & \omega/\Gamma \ll 1 \\ \omega/w, & \omega/\Gamma \gg 1. \end{cases} \tag{8}$$

It is easy to verify that the estimate remains valid for any degeneracy line of two spectrum modes whose frequency difference increases linearly with distance from this line [the possible difference consists only in a replacement of $\hbar k_0$ in (8) by the thermal momentum of the phonon].

We note that in addition to the possible dependence of the imaginary part of the dielectric constant ε'' on ω via the phase volume of the processes, ε'' always contains a factor proportional to ω, and when account is taken of the difference between the equilibrium populations of the states $(\mathbf{k}, i)$ and $(\mathbf{k}, j)$, this factor yields

$$N(\omega_{\mathbf{k}j}) - N(\omega_{\mathbf{k}i}) \approx \frac{\partial N_i}{\partial \omega_{\mathbf{k}i}} \omega.$$

Taking this into account, the result obtained above for the phase volume can be reformulated as follows: $\varepsilon''(\omega)/\omega$ depends on Γ at $\omega/\Gamma \ll 1$ in the same manner as $\varepsilon'(\omega)/\omega$ depends on ω at $\omega/\Gamma \gg 1$. The statement so formulated remains valid for the case when the frequency difference between the modes depends quadratically on the distance to the degeneracy line, but the matrix element of the interaction of the field vanishes on this line [Sec. 2(a)].

To demonstrate this, it is no longer sufficient to consider only the phase volume of the process, and it is necessary to write down an expression, for example, for the contribution made to the dielectric-loss tangent $\tan\delta$ by three-quantum process of type (1), with addition of the phenomenological allowance for the mode width. Using Eq. (38.4) of Ref. 9, we write

$$\operatorname{tg}\delta = \frac{\pi}{4\omega_0^2} \sum_{i \neq j} \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} dy \int \frac{d^3k}{(2\pi)^3} |V^{oij}(0, \mathbf{k}, -\mathbf{k})|^2 \times \frac{N(x) - N(y)}{xy} \delta(x - y + \omega) P_\Gamma(x - \omega_{\mathbf{k}i}) P_\Gamma(y - \omega_{\mathbf{k}i}). \tag{9}$$

Here and below $N(x) = (e^{\hbar x/T} - 1)^{-1}$; $\omega_{\mathbf{k}j}$ is the frequency of the phonon branch j for the wave vector $\mathbf{k}$; $P_\Gamma(x)$ is defined in (4); $V^{oij}(0, k, -k)$ is the matrix element of the potential contained in the three-phonon anharmonicity:

$$H_{int}^{(3)} = \frac{1}{2} \sum_{\substack{\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3=0 \\ i,j}} V^{oij}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3) \xi_{\mathbf{k}_1}^o \xi_{\mathbf{k}_2}^i \xi_{\mathbf{k}_3}^j, \tag{10}$$

where ξ_k^i are the normal phonon coordinates of the mode i, and ξ_k^o is the same for the soft optical mode of the spectrum.

Calculations using (9) yield for $\varepsilon''(\omega)/\omega$ a result proportional to (8), thus demonstrating the validity of the statement made concerning $\varepsilon''(\omega)/\omega$ for this case, too. The proportionality indicated above is a consequence of the cancellation of two effects: the increase of the surface of the torus by a factor $[k_0 w/\max(\omega, \Gamma)]^{1/2}$ as a result of the quadratic law according to which the modes move apart, and the decrease of the contribution from the processes occuring on this surface as a result of the vanishing of the matrix element on the degeneracy line.

We can now draw the following conclusions concerning the influence of Γ on the contribution to the losses by processes of type (1).

a) Isotropic case. At $\omega/\Gamma \gg 1$ and in arbitrary fields, allowance for the damping leads to small corrections. At $\omega/\Gamma \ll 1$ in non-opening fields, $\varepsilon''(\omega)/\omega$ is independent of ω and consequently allowance for the damping Γ is inessential. In opening fields, as noted in the introduction, the processes cluster about two points of $\mathbf{k}$-space and consequently the phase volume of the processes begin to depend on ω at $\omega/\Gamma \gg 1$ and the quantity Γ begins to determine the value of $\varepsilon''(\omega)/\omega$ for $\omega/\Gamma \ll 1$. In the language of perturbation theory constructed with even powers of the anharmonicity, this means that in the latter case we cannot confine ourselves to second-order terms and it is necessary to take into account higher-order terms (multiquantum processes), which now become decisive.

b) Anisotropic case. At $\omega/\Gamma \gg 1$ there is obviously no need to take Γ into account in arbitrary bias fields. At $\omega/\Gamma \ll 1$ the value of Γ determines the value of the phase volume of the process. Therefore in arbitrary field the multiquantum processes are decisive. And whereas in the isotropic case the strong three-quantum processes were suppressed by the field against the background of the weak multiquantum processes, now the dependence of the losses on the bias fields are determined by the dependence of the contribution of the multiquantum processes on this field.

Whereas up to now we were interested in the influence of the field on the losses via a slight distortion of the spectrum, we proceed now to examine processes which have become possible because of the lowering of the symmetry by the application of the field.

4. TRANSITIONS "WITHIN" ONE MODE

In the Introduction we have advanced arguments showing that the contribution from processes of type (2) can be appreciable even in weak bias fields. The influence of processes of type (2) on the crystal dynamics was quantitatively considered by a temperature technique in Refs. 4, 6, 7, and 11. We note some of the features of the calculation: a) in contrast to the case of processes of type (1), the actual expansion parameter of the perturbation-theory series turned out to be not $\Gamma/\overline{\omega}$ but Γ/ω; b) therefore the lowest non-vanishing order of perturbation theory can be used only at $\Gamma/\omega \ll 1$; c) at $\Gamma/\omega > 1$ the perturbation-theory series diverges and its summation leads to a solution of the linearized Boltzmann equation for the nonequilibrium growth rate to the phonon distribution function while $\tan\delta$ is expressed in term of this growth rate; we note that the result obtained for the losses can be found directly with the aid of the kinetic equation; d) the equation obtained for $\Gamma/\omega \gg 1$ yields at $\Gamma/\omega \ll 1$ a value of $\tan\delta$ that coincides with the result obtained in the lowest order of perturbation theory.

We now rewrite the result of Balagurov and Vaks[7] for $\tan\delta$ in a homogeneous measuring field:

$$\operatorname{tg}\delta=-\frac{1}{2T\omega_0^2}\sum_i\int\frac{d^3k}{(2\pi)^3}\frac{V^{oii}(0,\mathbf{k},-\mathbf{k})}{2\omega_{\mathbf{k},i}}N_{\mathbf{k},i}(N_{\mathbf{k},i}+1)\operatorname{Im}(f_{\mathbf{k},i}+f_{-\mathbf{k},i}), \tag{11}$$

where the notation is the same as in (9), $N_{\mathbf{k},i}=N(\omega_{\mathbf{k},i})$, and $f_{\mathbf{k},i}$ is determined from the equation

$$i\omega f_{\mathbf{k},i}=\frac{i\omega}{2\omega_{\mathbf{k},i}}V^{oii}(0,-\mathbf{k},\mathbf{k})+\frac{\hat{J}(f)}{N_{\mathbf{k},i}(N_{\mathbf{k},i}+1)}, \tag{12}$$

where the linearized collision operator $J(f)$ takes the usual form (see Refs. 12, 6, and 7).

The approximate solution (12) obtained by Balagurov and Vaks[7] will not do for us, since it yields identically zero for $\tan\delta$. We consider the refinement of this solution to be difficult, and use therefore an estimated solution of the kinetic equation (12), replacing, in order of magnitude, $\hat{J}(f)$ by $N_{\mathbf{k},i}(N_{\mathbf{k},i}+1)\Gamma f_{\mathbf{k},i}$. Now, solving (12) without difficulty and substituting the result in (11), we obtain for the increment[3)] $\tan\delta^{(2)}$ to $\tan\delta$

$$\operatorname{tg}\delta^{(2)}\approx\frac{T}{\hbar\omega_0}\frac{\omega\Gamma}{\omega^2+\Gamma^2}\frac{\omega_E^2}{\omega_0^2}K_\beta^2,\quad \hbar\omega_0<T<\Theta. \tag{13}$$

To obtain (13), $V^{oii}(0,\mathbf{k},-\mathbf{k})$ was expressed in terms of the material constants and other parameters of the problem, namely the shift $\omega_e^2=\omega_0^2(\mathbf{E})-\omega_0^2(\mathbf{E}=0)$ of the square of the limiting frequency of the soft mode under the influence of the electric field $\mathbf{E}$, and the dimensionless constant $K_\beta^2=\hbar b\lambda^2/64\pi^3w^3$ of the fourfold anharmonicity of the optical phonons; here b is the coefficient of dielectric nonlinearity, $\lambda^2=z^2/vM$ has the meaning of the square of the plasma frequency of the soft mode, where z is the effective charge and M is the reduced mass of the oscillation, while v is the volume of the unit cell of the crystal.

It remains to ascertain when $\tan\delta^{(2)}$ is larger than the corrections to $\tan\delta$ in terms of the parameter ω_E^2/ω_0^2, corrections in which we were not interested in this work, for example the field dependence of the four-quantum processes or the implicit dependence of $\tan\delta$ on E^2 because of the dependence of ω_0 on E^2. We explain this first for the isotropic spectrum. The losses in this case, in the absence of a bias field, can be easily obtained from Eq. (9) ($P_\Gamma(x)$ can be replaced by a δ function). We use a result obtained by Vaks[9] from Eq. (9):

$$\operatorname{tg}\delta^{(1)}\approx\frac{T}{\hbar\omega_0}\frac{\omega}{\omega_0}K_q^2,\quad \hbar\omega_0<T, \tag{14}$$

where $K_q^2=\hbar q^2\lambda^2/64\pi^3\rho w^5$ is the dimensionless constant of the electrostriction interaction; here q is the electrostriction modulus and ρ is the density. We note incidentally that $\mu^2\equiv K_\beta^2/K_q^2=b\rho w^2/q^2$ characterizes the influence of the electrostriction on the dielectric nonlinearity of the ferroelectric.

We now find, that $\tan\delta^{(2)}$ exceeds the discarded corrections in order of magnitude when

$$\operatorname{tg}\delta^{(2)}\Big/\frac{\omega_E^2}{\omega_0^2}\operatorname{tg}\delta^{(1)}\gg1\quad\text{or}\quad\frac{\omega}{\mu(\Gamma\omega_0)^{1/2}}\ll1. \tag{15}$$

If we now estimate the losses in the anisotropic case by using the order-of-magnitude value at $\omega/\Gamma \gg 1$

$$\operatorname{tg}\delta^{an}\approx\operatorname{tg}\delta^{(1)}\frac{\omega}{\omega_0}=\frac{T}{\hbar\omega_0}\left(\frac{\omega}{\omega_0}\right)^2K_q^2 \tag{16}$$

and at $\omega/\Gamma \ll 1$

$$\operatorname{tg}\delta^{an}\approx\operatorname{tg}\delta^{(1)}\frac{\Gamma}{\omega_0}=\frac{T}{\hbar\omega_0}\frac{\Gamma\omega}{\omega_0^2}K_q^2, \tag{17}$$

then we find that $\tan\delta^{(2)}$ exceeds the discarded corrections at

$$\omega/(\Gamma\omega_0^2\mu^2)^{1/3}\ll1 \tag{18}$$

if $\omega/\Gamma \gg 1$ and at

$$\Gamma/\mu\omega_0\ll1 \tag{19}$$

if $\omega\Gamma \ll 1$.

5. CONCLUSION

The influence of a weak electric field on the dielectric losses in centrosymmetric ferroelectrics manifests itself in two ways. First, in its effect on the absorption processes that already exist in the absence of a bias field. The analysis in Sec. 2 was carried out for a cubic ferroelectric, and the result of this section can be generally stated as follows: if the phonon spectrum of the ferroelectric, has degeneracy lines near which three-quantum absorption of a measuring-field quantum can take place, then the bias field will generally speaking lift this degeneracy partially, and in the case of an "opening" field this leads to a strong decrease of the contribution of these processes to the losses.

We now estimate the "opening" values of the field.[4)] Here, as above, we confine ourselves to temperatures $\hbar\omega_0 \lesssim T < \Theta$ (at $T \ll \hbar\omega_0$ the soft mode is frozen out and the excited spectrum of the crystal loses its ferroelectric features). At these temperatures, by virtue of the small contribution of the acoustic branches relative to the parameter T/Θ, we are interested only in soft optical branches.

We begin with a gap quadratic in the field [Sec. 2(a)]. Its relative magnitude is of the order of ω_E^2/ω_c^2, which in turn is proportional to the relative change $\Delta\varepsilon/\varepsilon$ of the dielectric constant:

$$\omega_E^2/\omega_c^2=-(\Delta\varepsilon/\varepsilon)(\omega_0^2/\omega_c^2),$$

since $\varepsilon\sim\omega_0^{-2}$. Recognizing that the main contribution to the losses is made by $\omega_c(\mathbf{k})\approx T/\hbar$, we find that this degeneracy line is no longer effective when

$$\frac{\Delta\varepsilon}{\varepsilon}\frac{\omega_0^2}{\omega_c^2}\approx\frac{\max(\omega,\Gamma)}{\omega_c}\qquad\frac{\Delta\varepsilon}{\varepsilon}\approx\frac{T\max(\omega,\Gamma)}{\hbar\omega_0^2}. \tag{20}$$

For the optical intermode gap linear in the field we obtain in place of (20)

$$\left(\frac{\Delta\varepsilon}{\varepsilon}\frac{\omega_0^2}{\omega_c^2}\right)^{1/2}\approx\frac{\max(\omega,\Gamma)}{\omega_c}\qquad\left(\frac{\Delta\varepsilon}{\varepsilon}\right)^{1/2}\approx\frac{\max(\omega,\Gamma)}{\omega_0}. \tag{21}$$

For the gap produced at the location of the random degeneracy of the optical and acoustic modes [Secs. 2(c) and 2(d)] there are differences: the main contribution to the losses is made by $\omega_c(\mathbf{k})\approx\omega_0$; from an examination of the dispersion equation for five low-lying modes near the intersection of the longitudinal acoustic and optical modes it is easy to find that the sought gap $\omega_s\approx\mu^{-1}\omega_E$ (this is easily understood, since the change of ω_E is due to the dielectric nonlinearity, while the change of ω_s is due to electrostriction). In this case we have for $\Delta\varepsilon/\varepsilon$, which characterizes the opening field,

$$\omega_s\approx\max(\omega,\Gamma),\qquad\left(\frac{\Delta\varepsilon}{\varepsilon}\right)^{1/2}\approx\mu\frac{\max(\omega,\Gamma)}{\omega_0}. \tag{22}$$

Second, if (18) and (19) are satisfied, the processes that are allowed in the presence of a bias field cause additional losses in accord with (13), and the losses increase fastest with the field at $\omega\approx\Gamma$. In the opposite case processes of type (2) no longer determine the addition increment quadratic in the field to $\tan\delta$, whose sign and magnitude must be obtained from a detailed analysis of the spectrum.

What can we deduce from measurements of the dependence of $\tan\delta$ on the weak bias field E and on the frequency of the measuring field ω?

a) Since the first of these mechanisms yields a threshold-type dependence of $\tan\delta$ and E (decrease), and the second a purely quadratic dependence (increase), the contribution of the former can be separated against the background of the latter.

b) At $\omega/\Gamma\gg1$ the plot of $\tan\delta(\mathbf{E})$, after separation of the contribution of the second mechanism, should have steps of the order of $\tan\delta(\mathbf{E}=0)$, corresponding to turning off the processes near the next degeneracy line. The fields at which they are observed should have a linear or square-root dependence on ω. One can expect larger numbers of steps when measuring and bias fields are parallel. On the other hand if $\omega/\Gamma\ll1$, the steps will be determined by the contribution of the four-quantum processes. The presence of other mechanisms of dielectric relaxation, which are not connected with lattice anharmonicity, will lead to a decrease of the relative sizes of the steps.

c) The increment quadratic in E to $\tan\delta$ should be positive if conditions (18) and (19) are satisfied; the maximum growth rate of the increment with increasing field corresponds to $\omega\approx\Gamma$.

Thus, by measuring the dependence of $\tan\delta$ on E and ω we can assess the role of three-quantum processes in the formation of the dielectric losses in a ferroelectric. By measuring the constant coefficient in the $\tan\delta$ increment quadratic in E and by finding the maximum of its dependence on ω, we can estimate the value of Γ, which corresponds in (13) to the value of the damping of the soft optical phonons.

In conclusion, let us dwell on the applicability of our results to real ferroelectrics. It is known that ferroelectrics of the displacement type are characterized by large values of the relative damping Γ/ω_0 of the soft mode. For example for a number of perovskites, according to Table 6 on p. 239 of Ref. 9, the ratio Γ/ω_0 takes on values from several hundredths and higher. This makes it difficult to satisfy, with sufficient margin, the inequalities $\omega<\omega_0$ and $\Gamma<\omega$, thus making the observation of frequency-dependent steps difficult. Numerical estimates of the effects described in this paper yield for $SrTiO_3$ at $T=90$ K and $\omega=2.2\times10^{10}$ Hz for the magnitude of the steps

$$\Delta\,\mathrm{tg}\,\delta\approx\frac{T}{\hbar\omega_0}\frac{\omega\Gamma}{\omega_0^2}K_q^2\approx1.4\cdot10^{-5} \tag{23}$$

and for the contribution of the processes of type (2), calculated from (13),

$$\mathrm{tg}\,\delta^{(2)}\approx5\cdot10^{-2}\frac{\Delta\varepsilon}{\varepsilon}. \tag{24}$$

We used for the estimates the numerical values from Ref. 9: $\omega_0=1.3\cdot10^{12}$ Hz, $\Gamma=10^{11}$ Hz, $K_q^2=0.7\cdot10^{-2}$, $K_g^2=0.15$, $\mu^2=20$. For comparison we present the experimental value of $\tan\delta(\mathbf{E}=0)$ from the same source:

$$\mathrm{tg}\,\delta^{\exp}=1.6\cdot10^{-3}. \tag{25}$$

Comparing (23) and (25) we see that the relative magnitude of the predicted steps is exceedingly small. As expected [the inequality (19) is satisfied], $\tan\delta^{(2)}$ is larger than the trivial increment to $\tan\delta(\mathbf{E}=0)=\tan\delta^{\exp}$ of the order of $\tan\delta^{\exp}\Delta\varepsilon/\varepsilon$.

It follows from the estimates that to observe the steps $\tan\delta$ must be measured with rather high accuracy. On the other hand, the contribution of processes of type (2) is probably easy to observe in spite of the requirement $\Delta\varepsilon/\varepsilon\ll1$.

We recall that all the results were obtained under the assumption that there exist well defined phonon states. This limits the temperatures and the class of substances to which they can be applied. For example, our results cannot be applied to the region of developed critical fluctuations. At present there are no known experimental studies of the details of the dependence of $\tan\delta$ on E and ω in weak bias fields.

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[1)]We point out a misprint in Ref. 9; the columns in (35.15) should be interchanged.

[2)]It can be shown that since the longitudinal branch is separated for the considered directions, an admixture of longitudinal acoustic oscillations makes a contribution that is small compared with those considered below.

[3)]More accurately speaking, we have written out here the contribution of the absorption processes within the soft optical mode, since the contribution from such processes within the acoustic modes is less relative to the parameter T/Θ at $T < \Theta$. Here and below, when speaking of the increment to $\tan\delta$, we mean the increment to $\tan\delta(\mathbf{E}=0)$.

[4)]We estimate below $\Delta\varepsilon/\varepsilon$, which is equivalent to an estimate of E, but is more universal.

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Investigation of antiferromagnetic resonance and two-magnon absorption in the weak ferromagnet $CoCO_3$

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The high-frequency AFMR branch and two-magnon absorption are detected and investigated in $CoCO_3$. A frequency shift of the electric-dipole two-magnon absorption in a magnetic field is observed for the first time. The behavior of the low-frequency branch of the spectrum is investigated in magnetic fields comparable with the crystal exchange field. The experimental results are interpreted with the aid of a theory that takes into account the large value of the uniaxial anistropy and the appreciable Dzyaloshinskiĭ interaction. It is shown that the two-magnon absorption in $CoCO_3$ is due to simultaneous excitation of two magnons from the high-frequency branch of the spin-wave spectrum.

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1. INTRODUCTION

Spectroscopy in the longwave infrared (LIR) range of the spectrum is an extremely effective experimental method that has been used to investigate the high-frequency (HF) properties of many antiferromagnetic dielectrics (see, for example, the reviews of Foner[1] and of Richards[2]). The present paper reports the detection and investigation by this method of a HF branch of the spin-wave spectrum and of two-magnon absorption in the weak ferromagnet cobalt carbonate. It also reports an investigation of the low-frequency (LF) branch of the spectrum in magnetic fields comparable with the crystal exchange field. Preliminary results of these investigations were reported by us earlier.[3-5]

The magnetic properties of $CoCO_3$ (space group D_{3d}^6) have been studied in considerable detail. It was discovered by Borovik-Romanov and Ozhogin[6,7] that $CoCO_3$ changes to a magnetically ordered state at $T_N = 18.1$ K. The magnetic moments of the sublattices are canted, and the resultant weak ferromagnetic moment lies in the basal plane of the crystal. Subsequent neutron-diffraction data[8] indicate that the magnetic moments of the sublattices also lie in the (111) plane. In a phenomenological theory of magnetism, the appearance of a weak ferromagnetic moment is due to the existence of a Dzyaloshinskiĭ effective field H_D, whose value for $CoCO_3$, according to the data of Ref. 7, is $H_D = 28$ kOe. This value of H_D was later confirmed by investigations of the susceptibility of $CoCO_3$ in weak magnetic fields.[9] By "weak" fields are understood fields $H \ll H_E$, where H_E is the crystal exchange field. The magnetization of $CoCO_3$ in the basal plane has been investigated by use of pulsed magnetic fields of intensities up to 340 kOe.[10] Signs of an approach of the magnetization to saturation were detected; the saturation value was close to the pure-spin value $2g\mu_B SN = 16\,750$ cgs emu/mol, where $g = 2$, $S = \frac{3}{2}$, and $2N$ is Avogardro's number. The value determined for the exchange field was $2H_E = 300$ kOe.

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