Second sound in ferroelectrics

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It is shown that second-sound temperature waves, which are very difficult to observe by the method of light scattering in ordinary dielectrics, are much easier to observe in a displacive ferroelectric with weakly damped soft mode. The point is that the second-sound waves are formed in ferroelectrics via participation of soft-mode phonons that interact relatively strongly with one another and with acoustic phonons. In the upshot, the frequency interval in which second sound exists turns out to be much broader than in ordinary dielectrics at the same temperatures. This is attributed to the relatively large damping of the soft-mode phonons in ferroelectrics. The second-sound wavelengths in ferroelectrics can be of the order of the wavelength of visible light. The conditions for propagation and damping of second sound in a ferroelectric are quantitatively analyzed for temperatures on the order of the phonon soft-mode energy. The second-sound velocity turns out to be of the order of that of ordinary sound, and the scattering intensity can be 100 times smaller than the Mandel' shtam-Brillouin intensity at the same temperature. Contributing to the experimental separation of light scattering by second sound can be the dependence of the differential scattering intensity on the external field, due to the field dependence of the soft-mode frequency.

1. INTRODUCTION

Second sound, or wavelike propagation of the temperature of the phonon gas of a crystal, is a classical photon-kinetic phenomenon. It was extensively investigated theoretically (see, e.g., Refs. 1 and 2). Its experimental study, however, encountered substantial difficulties: the only "classical" dielectric in which second sound could be observed was crystalline NaF.[3,4] It is therefore vital to search for new physical situations in which weakly damped temperature waves can be observed. The purpose of the present paper is to show that such a situation can be realized in a ferroelectric of the displacive type with a weakly damped soft mode. We shall show that by experimentally investigating the propagation and damping of second sound in ferroelectrics one can determine important thermodynamic, and particularly kinetic, characteristics of phonons. This pertains above all to soft-mode phonons.

The best conditions for experimentally observing this phenomenon, as they seem to us now, will be discussed in detail in Sec. 6.

It must be stated that additional sound-like excitations were found by Schneider and Stoll[5] and by Sakhnenko and Timonin[6] in theoretical studies of various models with structural phase transitions. The results of our present paper are compared in Sec. 7 with those of Refs. 5 and 6.

2. "FREQUENCY WINDOW" OF SECOND SOUND IN A FERROELECTRIC

It is known (see, e.g., Refs. 1 and 2) that the frequency of a weakly damped phonon-gas temperature wave should satisfy the inequality

$$\tau_\varkappa^{-1} \ll \omega \ll \tau_N^{-1}, \tag{1}$$

where $\tau_\varkappa$ and τ_N are the characteristic relaxation times of the quasimomentum and of the energy in the phonon gas. Neglecting phonon scattering by impurities and defects, $\tau_\varkappa^{-1} = \tau_U^{-1}$ is the frequency of the umklapp processes and τ_N^{-1} is the frequency of the normal collisions. Let us show how different the conditions (1) are in ordinary dielectrics and in phonon-soft-mode displacive ferroelectrics.

In an ordinary dielectric,[1] at temperatures T (here and below, in energy units) higher than or of the order of the Debye temperature Θ, we have

$$\tau_U^{-1} \approx \tau_N^{-1} \approx \Omega_D T / M w^2$$

($\hbar\Omega_D \equiv \Theta$, Mw^2, is the characteristic atomic energy in the crystal, and m and w are the mean values of the atom mass and of the speed of sound in the crystal), and the condition (1) for the existence of second sound cannot be met. At $T \ll \Theta$ both frequencies decrease rapidly as the temperature is lowered:

$$\tau_U^{-1} \propto e^{-\Theta/T}, \quad \tau_N^{-1} \propto T^5.$$

The difference between the rates of their decrease comes into play only at sufficiently low temperatures. Both quantities become in this case substantially lower than their values at $T \gtrsim \Theta$. This is manifested by the fact that the obtained second-sound window is of rather low frequency. It is just the low frequency which makes the very existence of the window most vulnerable from the standpoint of impurity scattering. The point is that impurity scattering, together with umklapp processes, ensures relaxation of the phonon-gas quasimomentum density. If $\tau_d^{-1} > \tau_U^{-1}$ (τ_d^{-1} is the characteristic frequency of the phonon-impurity collisions) we have $\tau_\varkappa^{-1} \approx \tau_d^{-1}$, so that owing to the smallness of τ_N^{-1} even low densities of impurities and defects can cause narrowing of the window (1), and if $\tau_d^{-1} \approx \tau_N^{-1}$, which is also easily attainable because τ_N^{-1} is small, they can cause it to vanish completely.

The phonon spectrum of a displacive ferroelectric is known[7] to differ from that of an ordinary dielectric in that the frequency of one of the optical modes at the center of the

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Brillouin zone is anomalously low. The presence of such a mode, called soft, in the ferroelectric lattice-vibration spectrum uncovers new possibilities for the existence of weakly damped temperature waves. The point is that it is possible to separate in such a phonon spectrum a group of soft-mode long-wave phonons and relatively long-wave acoustic phonons, for which the energy and quasimomentum relaxation is effected at any temperature by different processes with different characteristic collision frequencies. The condition (1) for the existence of second sound can therefore be met for this group of phonons at arbitrary temperatures. By arbitrary temperature we mean an arbitrary ratio of T and Θ, but assume, naturally, that at the temperatures considered the soft mode remains soft ($\Omega_0 \ll \Omega_D$) and weakly damped ($\Omega_0 \gg \Gamma$), where Ω_0 and Γ are the frequency and damping of the soft mode at the center of the Brillouin zone. Since the phonon kinetics was investigated in detail only for cubic ferroelectrics, we shall pay attention to just these crystals, although the general reasoning can be applied to other compounds with structural transitions that are accompanied by softening of the phonon soft mode.

As shown by Balagurov, Vaks, and Shklovskiĭ,[8] the damping of the phonon soft mode of a cubic ferroelectric is determined by the interaction between low-energy phonons of the long-wave part of the spectrum (soft-mode phonons with long-wave acoustic ones).

It must be assumed in the framework of the theory of a weakly anharmonic crystal that the main contribution to the damping Γ of the soft mode is made by three-quantum processes,[9] so that the following estimate is valid[8,9]

$$\Gamma \approx T\Omega_D/Mw^2 \quad (T \gtrsim \hbar\Omega_0). \tag{2}$$

This estimate corresponds to the contribution of the electrostriction interaction. It is therefore obvious that this interaction should ensure the same damping of acoustic phonons having frequencies on the order of Ω_0. The contribution (2) is also predominant for these phonons. This can be verified by comparing (2) with the contributions made to the damping by the interaction between the considered phonons and the thermal ones. These contributions, Γ_T, can be estimated from the known equations for the damping of high-frequency sound[1,2]

$$\Gamma_T \approx \begin{cases} \Omega_0 \dfrac{T}{Mw^2}, & T \gtrsim \Theta & (3) \\ \Omega_0 \dfrac{T}{Mw^2}\left(\dfrac{T}{\Theta}\right)^3, & \hbar\Omega_0 \lesssim T \ll \Theta\,. & (4) \end{cases}$$

We see thus that there exists a group of long-wave phonons (soft-mode phonons and acoustic phonons with frequencies of order Ω_0) whose damping builds up in the main through mutual scattering. Since these are long-wave phonons, the quasimomentum is conserved in all these processes.[1)] The intrinsic temperature of this group of phonons can set in after substantially shorter times ($\tau_N = 1/\Gamma$) than the relaxation of their quasimomentum. This means that the condition for existence of second sound is met for the intrinsic temperature of this group.

To determine the size of the "frequency window" for second sound in a ferroelectric, we must estimate the characteristic quasimomentum-relaxation times for the indicated group of phonons.

Consider the contribution of the umklapp processes to the quasimomentum relaxation. Of basic imporance to us is the question of the minimum size of the "window." In our estimate of the characteristic times of the umklapp process we shall see to it only that they are not undervalued. Accordingly, any process in which a short-wave phonon participates will be regarded as umklapp. The contribution to the damping of the considered group from the interaction with the short-wave phonons yields the upper bound of τ_U^{-1}.

For the characteristic frequency of the umklapp processes with participation of an acoustic phonon having a frequency on the order of Ω_0 at $T \gtrsim \Theta$ we can, in accordance with the foregoing, use the estimate (3) for Γ_T, i.e.,

$$\tau_U^{-1} \approx \Omega_0 T/Mw^2. \tag{5}$$

A similar calculation for the characteristic frequency of umklapp processes with participation of a soft-mode long-wave phonon also leads to the estimate (5).

At $\hbar\Omega_0 \lesssim T \ll \Theta$ it is necessary to take into account in the estimate (5) the additional factor $\exp(-\Theta/T)$, which reflects the decrease of the number of thermally excited short-wave phonons on cooling, and we have in place of (5)

$$\tau_U^{-1} \approx \Omega_0 \frac{T}{Mw^2} e^{-\Theta/T}. \tag{6}$$

Comparing (2) with (5) and (6), we see that at $T \gtrsim \Theta$ second sound can exist in a band in which its frequency can change by $\tau_U/\tau_N \approx \Omega_D/\Omega_0 \approx \chi^{1/2}$ times (χ is the dielectric susceptibility of the ferroelectric. For $\Theta \gg T \gg \hbar\Omega_0$ this change can be even larger, by $\chi^{1/2}\exp(\Theta/T)$ times. Thus, the "frequency window" of the second sound in a ferroelectric is situated at rather high frequencies (in an ordinary dielectric, the upper limit of the "window" is of the order of $\Omega_D T/Mw^2 (T/\Theta)^4$ and can be quite broad, at any rate for a pure dielectric, even near the Debye temperature Θ.

3. EQUATIONS OF PHONON HYDRODYNAMICS

For a quantitative description of second sound in a ferroelectric one can set apart two situations that call for different approaches. These are the cases $T \approx \hbar\Omega_0$ and $\hbar\Omega_0 \ll T, \Theta$. In the first case the second sound is a temperature wave of the entire phonon system of the crystal, and in the second it is a wave of local temperature of phonons with energies on the order of $\hbar\Omega_0$, interacting strongly with one another and weakly with all other phonons. We present in this paper a quantitative description for $T \approx \hbar\Omega_0$. In this case the phonon-hydrodynamics equations can be obtained from the kinetic equation for acoustic phonons and soft-mode phonons by the standard Chapman-Enskog procedure (see, e.g., Ref. 2). We seek the phonon distribution function in the form of a Planck distribution with drift

$$N = n\left(\frac{\Omega_{j\mathbf{k}} - \mathbf{V}\mathbf{k}}{T}\right) + \text{nonequilibrium additions}, \tag{7}$$

where $\mathbf{V}$ is the drift velocity, $\mathbf{k}$ the wave vector of the phonons, and $\Omega_{j\mathbf{k}}$ the frequency of the phonons of the branch j.

The equations are of standard form[1,2]

$$D_{il}\dot{V}_l + S\frac{\partial T}{\partial r_i} + \lambda_{il}V_l - \nu_{ilmn}\frac{\partial^2 V_m}{\partial r_l \partial r_n} = 0, \tag{8}$$

$$C\dot{T}+TS\operatorname{div}V-\varkappa_{il}\frac{\partial^2 T}{\partial r_i \partial r_l}=0, \qquad (9)$$

where S and C are respectively the entropy density and heat capacity of the phonon gas, while the tensors D_{il}, λ_{il}, ν_{ilmn} and $\varkappa_{il}$ are expressed by equations analogous to those of ordinary dielectrics, but with the soft-mode branches included in the phonon spectrum:

$$D_{il}=\frac{\hbar^2}{T}\sum_j\int\frac{d^3k}{(2\pi)^3}k_ik_lN_0(N_0+1),$$

$$S=\frac{\hbar^2}{3T^2}\sum_j\int\frac{d^3k}{(2\pi)^3}k_i\frac{\partial\Omega}{\partial k_i}\Omega N_0(N_0+1),$$

$$\lambda_{il}=\frac{\hbar^2}{T}\sum_j\int\frac{d^3k}{(2\pi)^3}k_i\hat{I}_Uk_lN_0(N_0+1),$$

$$\nu_{ilmn}=\frac{\hbar^2}{T}\sum_j\int\frac{d^3k}{(2\pi)^3}\left\langle\!\left\langle k_i\frac{\partial\Omega}{\partial k_l}\right\rangle\!\right\rangle \times\hat{I}_N^{-1}N_0(N_0+1)\left\langle\!\left\langle k_m\frac{\partial\Omega}{\partial k_r}\right\rangle\!\right\rangle, \qquad (10)$$

$$\varkappa_{il}=\frac{\hbar^2}{T}\sum_j\int\frac{d^3k}{(2\pi)^3}\left\langle\!\left\langle\Omega\frac{\partial\Omega}{\partial k_i}\right\rangle\!\right\rangle\hat{I}_N^{-1}N_0(N_0+1)\left\langle\!\left\langle\Omega\frac{\partial\Omega}{\partial k_l}\right\rangle\!\right\rangle, \qquad (11)$$

where $\hat{I}_{U,N}$ are the linearized collision operators of the umklapp and normal processes, respectively. The operators are assumed to act on the functions of the wave vector and on the numbers of the phonon branch to the right of the operators. The symbol $\langle\langle\ldots\rangle\rangle$ denotes the following subtraction operator:

$$\langle\langle f\rangle\rangle=f-\Omega\left\{\sum_j\int\frac{d^3k}{(2\pi)^3}\Omega^2N_0(N_0+1)\right\}^{-1}\times\sum_j\int\frac{d^3k}{(2\pi)^3}f\Omega N_0(N_0+1)-\sum_l k_l\left\{\left\{\sum_j\int\frac{d^3k}{(2\pi)^3}k_l^2N_0(N_0+1)\right\}^{-1}\times\sum_j\int\frac{d^3k}{(2\pi)^3}fk_lN_0(N_0+1)\right\}, \qquad (12)$$

which ensures that expressions (10) and (11) are finite and single-valued. We use here and below the abbreviated notation

$$\Omega=\Omega_{\mathbf{k}j},\quad N_0=n(\Omega_{\mathbf{k}j}/T).$$

Just as in the case of an ordinary dielectric, the main damping of the second sound is described by the kinetic coefficients $\hat{\nu}$, $\hat{\varkappa}$, and $\hat{\lambda}$ (Refs. 1 and 2): the contribution of the terms with $\hat{\nu}$ or $\hat{\varkappa}$ to the damping is proportional to $\omega^2\tau_N$, and that of the terms with $\hat{\lambda}$ is independent of frequency and is proportional to τ_x^{-1}. In the cubic crystal of interest to us, the second-sound velocity w_{II} is connected with the coefficients of Eqs. (8) and (9) by the known relation[1,2]

$$w_{II}^2=TS^2/DC,\quad D\equiv D_{\parallel}=D_{22}=D_{33}.$$

Although the phonon spectrum used to calculate D, S, and C contains a soft mode and therefore differs substantially from the long-wave acoustic spectrum of an ordinary dielectric, estimates show that the second-sound velocity in a ferroelectric should be of the usual order of magnitude, i.e., of the order of the velocity of ordinary sound.

In the derivation of Eqs. (8) and (9) we have neglected the temperature dependence of the soft mode. We shall show that in the framework of our analysis this dependence can indeed be neglected. Inclusion in (7) of the temperature dependence of the spectrum frequencies does not interfere with the Chapman-Enskog procedure, but leads to additional terms

$$-\mu_{il}\frac{\partial\dot{T}}{\partial r_l}$$

in Eq. (8) and

$$\Delta C\dot{T}-\eta\ddot{T}-T\mu_{il}\frac{\partial\dot{V}_i}{\partial r_l}$$

in Eq. (9), where

$$\mu_{il}=\frac{\hbar^2}{T}\sum_j\int\frac{d^3k}{(2\pi)^3}\left\langle\!\left\langle\frac{\partial\Omega}{\partial T}\right\rangle\!\right\rangle\hat{I}_N^{-1}N_0(N_0+1)\left\langle\!\left\langle k_i\frac{\partial\Omega}{\partial k_l}\right\rangle\!\right\rangle, \qquad (13)$$

$$\eta=\hbar^2\sum_j\int\frac{d^3k}{(2\pi)^3}\left\langle\!\left\langle\frac{\partial\Omega}{\partial T}\right\rangle\!\right\rangle\hat{I}_N^{-1}N_0(N_0+1)\left\langle\!\left\langle\frac{\partial\Omega}{\partial T}\right\rangle\!\right\rangle, \qquad (14)$$

$$\Delta C=\frac{\hbar^2}{T}\sum_j\int\frac{d^3k}{(2\pi)^3}N_0(N_0+1)\Omega\frac{\partial\Omega}{\partial T}. \qquad (15)$$

The new kinetic coefficients $\hat{\mu}$ and η describe the additional contributions, proportional to ω^2, to the second sound damping. Standard order-of-magnitude estimates (see, e.g., Refs. 1 and 2) yield for (13) and (14) at $T\approx\hbar\Omega_0$

$$\mu\approx\frac{1}{\Gamma a^3}\left(\frac{T}{\Theta}\right)^3\xi, \qquad (16)$$

$$\eta\approx\frac{1}{\Gamma a^3}\left(\frac{T}{\Theta}\right)^3\xi^2, \qquad (17)$$

$$\xi\approx\frac{\Theta^2}{T}\frac{\partial\chi^{-1}}{\partial T}\approx\frac{\Gamma}{\Omega_0}, \qquad (18)$$

where a is the characteristic interatomic distance.

We wrote down (18) using (2) and the estimate

$$\partial\chi^{-1}/\partial T\approx T/\Theta M w^2,$$

obtained for $T\approx\hbar\Omega_0$ from the results of Ref. 11. As seen from (18), ξ is of the same order as the correlation parameter, so that $\xi\ll 1$ in the region where the Landau theory is valid. The damping contribution due to the coefficient η has a ready physical interpretation it is the analog, for second sound, of the Akhiezer mechanism, or of the fluctuational mechanism of ordinary-sound absorption.[1,12] This can be verified by comparing (14) with the known expression for the contribution from the Akhiezer mechanism to the viscosity tensor.[1] They differ by the interchange $\partial\Omega/\partial T\to\partial\Omega/\partial u_{il}$ (u_{il} is the strain tensor).

Comparison of the contributions made by η and $\hat{\mu}$ to the second sound with the contributions from $\hat{\varkappa}$ and $\hat{\nu}$ shows that the former are small relative to the parameter ξ. Since the kinetic equation used by us in the derivation of (8) and (9) is itself valid in terms of this parameter for the considered group of phonons, retention of the terms with $\hat{\mu}$ and η in the phonon-hydrodynamics equations is in exaggeration of the accuracy.

It is easy to verify that the correction ΔC (15) to the heat capacity is also small compared with C in terms of the parameter $1/\xi$. It should therefore also be neglected for the same reason.

4. LIGHT SCATTERING BY SECOND SOUND

Observation of light scattering by second-sound oscillations induced in an ordinary dielectric is a rather complicated experimental task.[4] In the case of thermal second-sound oscillations, the problem becomes in fact experimentally insoluble in view of the low scattered-light intensity and of the need to use very small scattering angles ($\lesssim 10^{-2}$).[13] We shall show that a much more favorable situation obtains in a ferroelectric, both with respect to intensity and with respect to the scattering geometry.

The need for using very small scattering angles was dictated by the "low-frequency" position of the existence window in an ordinary dielectric. As shown in Sec. 2, the "existence window" of the second sound in a ferroelectric is located in a higher-frequency region. The upper limit of the "frequency window" is a quantity of the order of the soft-mode damping Γ and easily reach several times ten gigahertz. This means that it is possible to have in a ferroelectric a weakly damped second sound with a wavelength of the order of that of light, and consequently light can be scattered by this excitation at angles that are not small.

We consider now the question of the intensity of the scattered light. A rough estimate for the intensity of light scattering by second-sound fluctuations is provided by the Landau-Placzek relation, by the Fabelinskiĭ formula,[14] or by the modified Landau-Placzek relation[13,15] for the central-peak intensity. Strictly speaking, however, the intensity of the central peak when it is split into a second-sound doublet does not remain constant. The point is that the central peak can be regarded, with good accuracy, as stemming from isobaric fluctuations of the entropy, while the fluctuations corresponding to second-sound excitations are no longer of isobaric origin. Allowance for this circumstance not only changes the result for the scattered-light intensity (although the intensity should remain of the same order, except in special "compensation" conditions, as in Ref. 13), but leads also to an additional anisotropy of this intensity (i.e., to a dependence on the direction of the transferred wave vector).

Confining ourselves, as above, to the case of a cubic crystal, we start from a coupled system of second-sound and elasticity-theory equations[1,2]:

$$\ddot{T}-w_{II}^2\frac{\partial^2 T}{\partial r_i^2}+\frac{\alpha KT}{C_u}\frac{\partial \ddot{u}_i}{\partial r_i}=0, \tag{19}$$

$$\ddot{u}-\rho^{-1}c_{ilmn}\frac{\partial^2 u_m}{\partial r_n \partial r_l}+\frac{\alpha K}{\rho}\frac{\partial T}{\partial r_i}=0,$$

where $\mathbf{u}$ is the acoustic displacement vector, c the elastic-moduli tensor, ρ the density, α and $K=c_{11}+2c_{12}$ the thermal-expansion coefficient and the hydrostatic compression modulus, respectively, and C_u the heat capacity at constant strain. Let us consider for this system the normal modes that propagate with a wave vector $\mathbf{q}$. Equations (19) are coupled as an anharmonically small effect of thermal expansion. Neglecting this effect, these modes are a temperature wave of amplitude $T_\mathbf{q}$ and three acoustic waves with amplitudes $A^{(s)}$ ($s=1,2,3$),

$$u_i=e_i^{(s)}(\mathbf{q})A^{(s)},$$

where $e_i^{(s)}(\mathbf{q})$ are the displacement polarization vectors in these waves. We determine the true normal modes of the system (19), $U^{(s)}$ and $\tau_\mathbf{q}$, in first order in the coupling constant of the equations; this constant is of the order of αT, and the modes are connected with A and $T_\mathbf{q}$ by the relations

$$iqA^{(s)}=U^{(s)}+\Lambda^{(s)}\tau_\mathbf{q}/T,\quad T_\mathbf{q}=\tau_\mathbf{q}-\Lambda^{(s)}c^{(s)}U^{(s)}/C_u, \tag{20}$$

where

$$\Lambda^{(s)}=\frac{n_i e_i^{(s)} w_0^2}{w_{(s)}^2-w_{II}^2}\alpha T,\quad w_0^2=\frac{K}{\rho},\quad w_{(s)}^2=\frac{c_{(s)}}{\rho},$$

$$c^{(s)}=c_{ijkl}e_i^{(s)}e_k^{*(s)}n_j n_l,\quad n_i=q_i/q, \tag{21}$$

the $\tau_\mathbf{q}$ mode corresponds to excitations of second sound with a wave vector $\mathbf{q}$. The differential (with respect to the directions) extinction coefficient for scattering of light by this mode, can be written, in analogy with Ref. 14, in the form

$$dh=\frac{\pi^2}{\lambda^4}\left(\frac{\partial \varepsilon_{il}}{\partial \tau}d_i'^{*}d_l\right)^2\langle\tau_\mathbf{q}\tau_{-\mathbf{q}}\rangle dO\, v, \tag{22}$$

where λ is the wavelength of the light in the crystal, $\mathbf{d}$ and $\mathbf{d}'$ are the polarization vectors of the incident and scattered light, $\langle\dots\rangle$ denotes thermodynamic averaging over a crystal of volume v, dO is the solid-angle element, ε_{il} is the dielectric tensor at the optical frequency,

$$\frac{\partial \varepsilon_{il}}{\partial \tau}=\frac{\partial \varepsilon_{il}}{\partial T}+\frac{\partial \varepsilon_{il}}{\partial u_{mp}}n_m e_p^{(s)}\Lambda^{(s)}/T. \tag{23}$$

Using the known relations for the probability of the fluctuations of the thermodynamic quantities and relations (20), we easily obtain

$$\langle\tau_q\tau_{-q}\rangle=T/vC_u[1+\Lambda_{(s)}^2c^{(s)}/C_uT]\approx T/vC_u. \tag{24}$$

The term in the square brackets can be neglected compared with unity: order-of-magnitude estimates show that it is anharmonically small. For the considered case of a displacive ferroelectric at $T\approx\hbar\Omega_0$, this term is of the order of the correlation parameter ξ. Ultimately, using relations (16)–(19), we can write

$$\frac{dh}{dO}=\frac{\pi^2}{\lambda^4}\frac{T^2}{C_u}\left[d_n'^{*}d_m\left(\frac{\partial\varepsilon_{nm}}{\partial T}+\frac{\partial\varepsilon_{nm}}{\partial u_{il}}N_{il}\alpha\right)\right]^2, \tag{25}$$

where

$$N_{il}=w_0^2\frac{e_i^{(s)}e_p^{(s)}n_p n_l}{w_{(s)}^2-w_{II}^2}. \tag{26}$$

In the ease of an elastically isotropic medium $N_{il}\propto n_i n_l$, the dependence of N_{il} on $\mathbf{n}$ for a cubic crystal is more complicated and reflects the cubic anisotropy of the elastic spectrum.

The anisotropy of the elastic spectrum is thus manifested as an additional anisotropy of the intensity of light scattering by second sound fluctuations. Such an anisotropy appears in pure form in scattering in which the polarization is not changed and the vectors **d** and **d**′ are parallel to a fourfold axis. Then

$$\frac{dh}{dO}=\frac{\pi^2T^2}{\lambda^4C_u}\left[\frac{\partial\varepsilon_{11}}{\partial T}+\frac{\partial\varepsilon_{11}}{\partial u_{22}}\left(\frac{\cos^2\gamma}{w_L{}^2-w_{II}{}^2}+\frac{\sin^2\gamma}{w_T{}^2-w_{II}{}^2}\right)\alpha w_0{}^2\right]^2, \quad (27)$$

where γ is the angle between the oscillation polarization vector in the "longitudinal" sound wave, while **n**, w_L, and w_T are the velocities of the "longitudinal" and "transverse" acoustic waves propagating along **n**.

It can be seen that the angular dependence of the differential extinction coefficient, described by (27), drops out in the case of an elastically isotropic spectrum, for in that case w_L, w_T, and γ are independent of the vector **n** (which lies in a plane $\perp$**d**) and $\gamma=0$.

The physical cause of the manifestation of the elastic-spectrum anisotropy in the anisotropy of the intensity of light scattering by second-sound fluctuations can be easily explained: since $\partial\varepsilon/\partial T$ and $\alpha\partial\varepsilon/\partial u_{il}$ are more likely to be of the same order, the amplitudes of the strains accompanying the thermal waves are important for the scattered-light intensity. Clearly, the amplitude of a strain corresponding to any acoustic wave connected with a thermal one by Eqs. (19) is larger the smaller the velocity difference between the second sound and the corresponding ordinary sound. The size of the "hydrostatic compression component" in the wave is also important. The structure of the tensor N_{il} (26) is just a manifestation of these two features. Only the first factor, naturally, plays a role for the directions along which a purely longitudinal acoustic wave can propagate. Assuming in the foregoing example that the second sound propagates along twofold and fourfold axes, it is easy to obtain for the ratios of the corresponding scattered-light intensities

$$\frac{I_{[100]}}{I_{[110]}}=\left(\frac{\partial\varepsilon_{\parallel}/\partial T+\alpha(\partial\varepsilon_{\parallel}/\partial u_{22})(c_{\parallel}+2c_{12})(c_{\parallel}-\rho w_{II}{}^2)^{-1}}{\partial\varepsilon_{\parallel}/\partial T+\alpha(\partial\varepsilon_{\parallel}/\partial u_{22})(c_{\parallel}+2c_{12})[(c_{11}+c_{12})/2+c_{44}-\rho w_{II}{}^2]^{-1}}\right)^2.$$

We present also an equation for the intensity ratio $I_{[100]}$ of light scattering by second sound for **d**‖**d**′‖[100] and **n**‖[010], and for the ratio $I_{\rm MB}$ for longitudinal sound in the same geometry

$$\frac{I_{[100]}}{I_{\rm MB}}=\frac{Tc_{11}}{C_u}\left(\frac{\partial\varepsilon_{11}/\partial T}{\partial\varepsilon_{11}/\partial u_{22}}+\alpha\frac{c_{11}+2c_{12}}{c_{11}-\rho w_{II}{}^2}\right)^2. \quad (28)$$

Note that to stay consistently in the framework of the weakly harmonic model of displacive ferroelectrics (see, e.g., Ref. 8), then $I_{[100]}/I_{\rm MB}\approx Tc_{11}\alpha^2/C_u$ turns out to be of the order of the correlation parameter ξ for $T\approx\hbar\Omega_0$.

Recall that the analysis in the present section pertains to low temperatures, $T\ll\Theta$. The relations obtained are equally valid for ordinary dielectrics and for low-temperature ferroelectrics. In the former case they are apparently only of academic interest, in view of the extreme difficulty of experimentally observing light scattering by second-sound fluctuations in an ordinary dielectric. For a low-temperature ferroelectric, these relations hold for a situation that is realistic from the experimental point or view, but in the relatively narrow temperature interval $T\approx\hbar\Omega_0$. As indicated in Sec. 2, high-frequency second sound can exist in a ferroelectric in a considerably larger temperature interval, but for $T\gg\hbar\Omega_0$ it will already be a local-temperature wave of soft-mode phonons and of acoustic ones with energies $\approx\hbar\Omega_0$. Clearly, in this case the intensity of the light scattered by the second sound can no longer be directly connected with macroscopic characteristics of the crystal, as was the case at $T\approx\hbar\Omega_0$. We do not analyze here the case $T\gg\hbar\Omega_0$ quantitatively. We can suggest, however, a rough estimate for the ratio of the intensity I_{II} of light scattered by "high-temperature" second sound to that of Mandel'shtam-Brillouin scattering by acoustic phonons, $I_{\rm MB}$, at the same temperature:

$$\frac{I_{II}}{I_{\rm MB}}\approx\frac{TK\alpha^2(T=\hbar\Omega_0)}{C_u(T=\hbar\Omega_0)}, \quad (29)$$

where $\alpha(T=\hbar\Omega_0)$ and $C_u(T=\hbar\Omega_0)$ are the values of α and C_u at a temperature equal to the energy of the soft-mode phonon limit. Let us show how this estimate was obtained. First, we took the scattering source to be opto-elastic interactions. Second, it is clear from the reasoning behind the derivation of (25) that Eq. (29) should contain a "thermal expansion coefficient" and a "thermal expansion coefficient" and a "heat capacity" for a local temperature of phonons with energy on the order of $\hbar\Omega_0$. To estimate such a "thermal expansion coefficient" we used the known phonon-thermodynamics relation[1,2]

$$T^2K\alpha=-\hbar^2\sum_j\int\frac{d^3k}{(2\pi)^3}\Omega\frac{\partial\Omega}{\partial u_{ii}}N_0(N_0+1), \quad (30)$$

and also the order-of-magnitude relation

$$\partial\Omega_0/\partial u_{ii}\approx\Omega_0(\Omega_D/\Omega_0)^2,$$

which is valid for soft-made phonons. The summation and the integration in (30), however, were only over phonons with energy $\lesssim\hbar\Omega_0$. The "heat capacity" for the local temperature was estimated similarly.

5. INFLUENCE OF DEFECTS ON SECOND SOUND IN A FERROELECTRIC

Just as in the case of an ordinary dielectric, scattering by crystal imperfections of a ferroelectric should cause narrowing of the second-sound "frequency window" on account of the decrease of $\tau_\varkappa$. The interaction of the (soft-mode) phonons with defects in ferroelectrics is more noticeably pronounced than that of acoustic phonons having the same frequency in ordinary dielectrics.[16] The increase of the characteristic frequency of the second-sound window on going from an ordinary dielectric to a displacive ferroelectric should therefore be accompanied by a decrease of the phonon-impurity collision times τ_d. One can visualize a situation in which a wide high-frequency window exists for second sound in an ideally pure ferroelectric but not in a real ferroelectric with defects. Let us estimate the defect density for which weakly damped second sound can exist in a ferroelectric. In view of the great variety of crystal imperfections (charged, uncharged, or polarized point defects; dislocations, crystallite boundaries, and others) it is desirable to obtain such an estimate for a realistic physical situation. To assess the scale of the discussed effects, however, we obtain an estimate for the simplest case of point defects that introduce a strong but local perturbation both into the kinetic energy and in the matrix of the lattice force constant (the mass of the subtituent atom differs substantially from that of the substituted one, and the force constants are noticeably altered near the defect).

Estimating τ_d for soft-mode long-wave phononons, we use a scheme described in Ref. 2. It is easy to verify by using Eqs. (9.16)–(9.19) and (12.4) of Ref. 2 that the contribution to τ_d^{-1} from the perturbation of the kinetic energy by the considered type of defect is of the same order as for acoustic phonons in an ordinary dielectric:

$$\tau_d^{-1}\approx N_d\Omega_D(\Omega_0/\Omega_D)^4, \tag{31}$$

where N_d is the atomic concentration of the defects.

The contribution to τ_d^{-1} from the perturbation of the force constants by the matrix defect can be estimated from relations (9.25), (9.28), (12.2) and (12.4) of Ref. 2, but with allowance for the fact that the soft mode is an optical phonon [it suffices to retain in (9.25) the zeroth expansion term of the exponential]. This contribution turns out to be substantially larger than (31) and than the corresponding contribution for acoustic phonons of the same energy in an ordinary dielectric, and is given by

$$\tau_d^{-1}\approx N_d\Omega_D. \tag{32}$$

The defects permit second sound to exist in a ferroelectric if the proper (anharmonic) damping Γ of the soft mode is much larger than their contribution τ_d^{-1} to the damping. For the considered type of uncharged point defects this condition, with allowance for (2) and (32), is equivalent to

$$N_d \ll T/Mw^2. \tag{33}$$

6. POSSIBILITIES OF OBSERVING SECOND SOUND IN FERROELECTRICS

Promising substances for observation of second sound excitations in light scattering are the virtual ferroelectrics $SrTiO_3$ and $KTaO_3$ at temperatures $T\approx 20$–40 K. The soft mode in these crystals has at these temperatures a frequency $\Omega_0\approx T/\hbar$ and a damping on the order of several cm^{-1}, so that second sound with frequencies $\omega \ll 100$ GHz is possible. In especially clean crystals, the lower limit of the second-sound existence window is determined by umklapp processes [Eq. (6)], and the window itself can reach 3–4 decades. The actual lower limit of the window should probably be determined by phonon scattering from the crystal imperfections. At sufficient density of the latter, there may be no window at all. We present an estimate of the concentration N_d of point defects of the type considered in Sec. 5, a concentration for which second sound can exist. Using (33) and putting $Mw^2\approx 10^5$ K, we get $N_d \ll 10^{-4}$. It must be noted that this estimate is patently approximate. To estimate the real second-sound existence window for a specific crystal it is desirable to have detailed information on the phonon-impurity scattering. This information can be extracted, for example, by theoretical reduction of the thermal-conductivity data.[4]

The intensity of light scattered by second-sound fluctuations can be calculated from the equations of Sec. 4. The ratio of the integral intensity of a second-sound line to one from thermal acoustic phonons is given, in order of magnitude, by the correlation parameter $\xi=\Gamma/\Omega_0\approx\Theta/Mw^2\approx 10^{-2}$. Note that for second sound in an ordinary dielectric this ratio would be determined by the substantially smaller parameter $(T/\Theta)^3T/Mw^2$. The relatively high location of the existence window in frequency makes it possible to use a geometry with relatively large scattering angles in experiments on light scattering from second sound.

The crystals mentioned are not the only possible objects for second-sound observations in ferroelectrics. The following must be taken into account when searching for other objects. First, second sound can exist also in dielectrics with sufficiently high transition temperatures (see Sec. 1). Second, although we have discussed in the present paper only cubic ferroelectrics, a similar situation with second sound can possibly be realized in uniaxial displacive ferroelectrics with a weakly polar soft mode (see, e.g., Ref. 17).

We point out one more circumstance that may be useful, in our opinion, for experimental observation of second sound in a ferroelectric. Application of an electric field or a change of temperature can cause a strong change of the soft-mode frequency. Such a restructuring of the phonon spectrum can strongly influence second-sound excitations. For example, these excitations should vanish if the soft-mode frequency is greatly increased.

7. COMPARISON WITH THE RESULTS OF REFS. 5 AND 6

Let us compare our results with those of Sakhnenko and Timonin[4] on additional sound-like excitations that can appear in the light-scattering spectrum. In Ref. 6 was considered an initial phonon spectrum containing only a single branch—a soft mode with an isotropic dispersion law $\Omega^2(k)=\Omega_0^2+ck^2$; it was established there that an additional excitation appears here, with a dispersion law $\omega^2=ck^2$ which is weakly damped at $\omega\gg\max(\Gamma,\Omega_0^2/\Gamma)$. It is known that such kinetic properties of phonon spectra are exceedingly sensitive to details of their spectra,[2] and it is therefore immaterial, from our point of view, whether the indicated excitations will exist in a crystal with a real anisotropic soft-mode spectrum. If such excitations do exist, they are weakly damped for the considered ferroelectric with phonon soft mode only at rather high frequencies $\omega\gg\Omega_0/\xi$. Actually,[2)] however, they can be only short-wave with frequencies $\gtrsim 100\ cm^{-1}$. It is clear that objects such as weakly damped excitations should not be manifested in the scattered-light spectrum. Thus, different soundlike excitations are investigated in the present paper and in Ref. 6. Only the temperature waves considered in the present paper can occur as weakly damped in experiments on light scattering.

The soundlike mode recorded in the computer experiments of Schneider and Stoll,[5] judging from all the foregoing, is likewise not second sound in its classical meaning (see, e.g., Ref. 2). This is indicated by the following circumstances: 1) the high-frequency $\omega \gtrsim \Gamma$ of the recorded mode; 2) the mode is recorded also when the phonon spectrum constitutes only one optical branch with 15% variation of the frequency over the Brillouin zone; clearly, in such a phonon spectrum the times to establish the temperature and the quasimomentum should be of the same order, i.e., a window for the existence of second sound should be present.

Just as in the case of Ref. 6, the high-frequency character of the soundlike excitation recorded in Ref. 5 makes it difficult to observe it by the light-scattering method.

[1)]The importance of taking this circumstance into account in the analysis of the thermal conductivity of ferroelectrics was first pointed out by Levanyuk *et al.*[10]

[2)]Phonon soft modes usually have frequencies not lower than 5–10 cm^{-1}

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