EFFECT OF THERMAL CONDUCTIVITY ON THE DIELECTRIC PERMITTIVITY AND PROPAGATION OF SOUND IN SOLIDS

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The thermal conductivity and low frequency kinetic effects in solids are analyzed by the temperature diagram technique in combination with the application of the inhomogeneous kinetic equation. The corresponding dispersion of the dielectric permittivity in ferroelectric substances and the elastic moduli in solid dielectrics are investigated. A microscopic expression for the Green's function is given for the hydrodynamic region of small **k** and ω for all temperatures. In particular, the expression describes the effects associated with second sound. Some features of the inelastic scattering cross sections for a crystal in the case of small energy and quasi-momentum transfer are discussed.

INTRODUCTION

In the imposition of a variable external field—electromagnetic, elastic and so on—on a medium, two basic relations are possible between the frequency of the field and the thermal relaxation time. If the field changes more rapidly than the establishment of thermal equilibrium is carried out, the oscillations are adiabatic; in the opposite case, the temperature remains constant and the oscillations are isothermal. The order of the values of the frequency ω and the wave vector **k** at which the character of the oscillations change can be found with the help of the heat conduction equation

$$\partial T / \partial t = \chi \Delta T. \tag{1}$$

It is clear from (1) that the oscillations are adiabatic for $\omega \gg \chi k^2$ and isothermal for $\omega \ll \chi k^2$. Here, whereas the characteristics of the adiabatic and isothermal oscillations—the dielectric permittivity, the speed of sound—are determined from thermodynamics, the description of the oscillations in the transitional region $\omega \sim \chi k^2$ requires a kinetic consideration. The present work is devoted to the microscopic consideration of the corresponding dispersion for the case of the study of the dielectric permittivity $\epsilon_{\alpha\beta}(\mathbf{k}, \omega)$ in ferroelectrics, the elastic moduli $c_{\alpha\beta\gamma\delta}(\mathbf{k}, \omega)$ in solid dielectrics, and also the features of low temperature kinetics associated with second sound.

The dispersion indicated leads to characteristic features in the correlation (Green's) functions of phonons $G(k, \omega)$ for small k and ω. Therefore, although in experiments on the high frequency measurements of $\epsilon_{\alpha\beta}$ and $c_{\alpha\beta\gamma\delta}$ are usually made in the adiabatic regime $\omega \gg \chi k^2$ only, in experiments on inelastic scattering, for example, of neutrons by a crystal, the study of $G(\mathbf{k}, \omega)$ in principle for arbitrary **k** and ω and, in particular, of the features of the transitional region $\omega \sim \chi k^2$.

For the "hydrodynamic" region considered of small ω and **k**, for not too low T, the expression for the susceptibilities can be found simply by the methods of macroscopic kinetics. This is done in Sec. 2. However, a microscopic analysis based on lattice dynamics is also of interest. Such an analysis enables us to connect $\epsilon(\mathbf{k}, \omega)$ and $c(\mathbf{k}, \omega)$ with the Green's function. It makes it possible to trace the microscopic picture and the region of applicability of the results, and also the transition to high frequency, "non-hydrodynamic" kinetics. In ferroelectrics, the micro-consideration allows a better understanding of the difference in the effect of relaxation processes on $\epsilon(\mathbf{k}, \omega)$ above and below the transition point (which was discussed previously on the basis of a rough model[1]), and in elastic moduli $c(\mathbf{k}, \omega)$—to follow quantitatively the appearance of second sound and the related dispersion of the kinetic coefficients as the temperature is lowered.[2,3] Finally, the expressions derived below for the kinetic coefficients may be useful for the study of temperature dependences, for example, in the vicinity of phase transitions.

The consideration is carried out by the methods of the temperature diagram technique on the basis of the inhomogeneous kinetic equation used earlier by Sham[2] for the consideration of second sound in solids. A general expression is found in Sec. 3 with the help of this equation for the singular part of the Green's function due to the heat conduction. Use of the results of the theory of ferroelectricity[4] permits us to connect these singularities with the singularities of $\epsilon(\mathbf{k}, \omega)$ and trace the agreement with the phenomenological expression for ϵ. An analytic study of the elastic moduli $c(\mathbf{k}, \omega)$ is given in Sec. 4. A general microscopic expression is given for $c(\mathbf{k}, \omega)$ in the hydrodynamic region of small ω and **k** for arbitrary **T**. The results obtained give, in particular, the quantitative expression obtained earlier by Shklovskiĭ[5] for the relation between "thermoelastic" and viscous sound damping. The given expression also describes the transition of the relaxation of the "thermoconduction mode" in second sound with decrease in T and the dispersion effects of heat conduction, viscosity and sound absorption, associated with it. These latter have been discussed before phenomenologically.[3] The features in the elastic scattering of neutrons at small **k** and ω, associated with the dispersion considered, are discussed briefly in Sec. 5.

2. PHENOMENOLOGICAL DESCRIPTION

Let us consider the effect of heat conduction on the dielectric permittivity $\epsilon_{\alpha\beta}(\mathbf{k}, \omega)$. The expression for the inverse susceptibility tensor $\zeta = 4\pi(\epsilon - 1)^{-1}$ in a variable field and with account of temperature oscillations can be written in the form

$$\zeta_{\alpha\beta} = \frac{dE_\alpha}{dP_\beta} = \zeta_{\alpha\beta}{}^T - \frac{1}{4\pi}\left(\frac{\partial S}{\partial P_\alpha}\right)_T \frac{dT'}{dP_\beta'}. \quad (2)$$

Here $\mathbf{P}$ is the polarization, $\zeta^T_{\alpha\beta} = (4\pi)^{-1}\partial^2 F/\partial P_\alpha \partial P_\beta$ the static, isothermal value of $\zeta_{\alpha\beta}$; $F(P, T)$ and $S = -(\partial F/\partial T)_P$ are the free energy and entropy per unit volume; T' and P' are the variable components of the temperature and the polarization. For simplicity, we consider only the case of a "clamped" crystal,[4,6] i.e., the oscillations are of sufficiently high frequency that the acoustic deformations $u'_{\alpha\beta}$ do not succeed in following the electric field and the piezoelectric coupling of the elastic and electric oscillations vanishes: $u'_{\alpha\beta} = 0$.

The quantities T' and P' can be connected by means of the heat conduction equation[7]. Furthermore, setting T' and P' proportional to $\exp(i\mathbf{k}\cdot\mathbf{r} - i\omega t)$, we have, from (2),

$$\zeta_{\alpha\beta}(\mathbf{k}, \omega) = \zeta_{\alpha\beta}{}^T = \frac{T}{4\pi C_P}\left(\frac{\partial E_\alpha}{\partial T}\right)_P \left(\frac{\partial E_\beta}{\partial T}\right)_P \frac{\omega}{\omega + i\chi_{\nu\delta}k_\nu k_\delta}. \quad (3)$$

Here C_P is the heat capacity per unit volume for constant polarization, $\chi_{\gamma\delta}$ is the temperature conductivity tensor. Here and below, repeated Greek indices indicate summation from 1 to 3. In correspondence with what was said in the Introduction, for $\omega \ll \chi k^2$ the inverse susceptibility ζ takes on the isothermal value ζ^T, and for $\omega \gg \chi k^2$ the adiabatic value ζ^S. In ferroelectrics, the quantity $(\partial E_\alpha/\partial T)_P$ is proportional to the polarization P_α, so that the difference between ζ^S and ζ^T is important only in the ferroelectric phase and then only for those components of $\zeta_{\alpha\beta}$ for which $P_\alpha \neq 0$ and $P_\beta \neq 0$.

In the consideration of the dispersion of the elastic moduli, we limit ourselves for simplicity to the consideration of centrosymmetric crystals in which there is no piezoeffect.[8] Then, solving the equation of elastic oscillations under the action of an external force density $\mathbf{f}(\mathbf{r}, t)$ with account of viscosity and temperature changes by the same method as above, we obtain

$$[-\rho\omega^2\delta_{\alpha\beta} + c_{\alpha\gamma\delta\beta}(\mathbf{k}, \omega)k_\gamma k_\delta]\,u_\beta' = f_\alpha(\mathbf{k}, \omega), \quad (4a)$$

$$c_{\alpha\beta\gamma\delta}(\mathbf{k}, \omega) = c^T_{\alpha\beta\gamma\delta} + \frac{\omega}{\omega + i\chi_{\mu\nu}k_\mu k_\nu}\frac{T}{C_u}\left(\frac{\partial S}{\partial u_{\alpha\beta}}\right)_T \left(\frac{\partial S}{\partial u_{\gamma\delta}}\right)_T - i\omega\eta_{\alpha\beta\gamma\delta}. \quad (4b)$$

Here ρ is the density, u' the displacement, C_u the heat capacity at constant deformation, and c^T and η the tensors of isothermal elastic moduli and viscosity. As above, it is easy to see that for $\omega \gg \chi k^2$ the tensor $c_{\alpha\beta\gamma\delta}$ undergoes a transition to the adiabatic value $c^S_{\alpha\beta\gamma\delta}$. In an isotropic body, the difference between the adiabatic sound velocity and the isothermal one exists only for longitudinal oscillations, since the transverse shear oscillations do not give rise to changes in volume and temperature.

3. MICROSCOPIC CONSIDERATION OF THE DISPERSION OF $\epsilon(\mathbf{k}, \omega)$

The microscopic method of calculation of the dielectric permittivity $\epsilon(\mathbf{k}, \omega)$ for small $\mathbf{k}$ was discussed in[9,10,4]. According to[9], to find ϵ by diagram methods, it is necessary to find the polarization operator for the interaction of the long-wave electromagnetic field with the medium, i.e., the set of diagrams that are not resolved into uncoupled parts over the single photon line. In our case of a dielectric crystal, the operator H_{em} of interaction with the long-wave electromagnetic field can be written in the form[10,4]

$$H_{em} = -\sum_{\mathbf{r}i} e_i \mathbf{u}_\mathbf{r}{}^i \mathbf{E}_\mathbf{r} + v_c \sum_\mathbf{r} (\varepsilon_\infty - 1)_{\alpha\beta}\frac{E_\mathbf{r}{}^\alpha E_\mathbf{r}{}^\beta}{8\pi} + \frac{v_c}{8\pi}\sum_\mathbf{r} (\mathbf{E}_\mathbf{r}{}^2 + \mathbf{H}_\mathbf{r}{}^2). \quad (5)$$

Here $\mathbf{u}^i_\mathbf{r}$ and e_i are the displacement and the effective charge of the i-th ion in the cell $\mathbf{r}$; v_c is the cell volume, $\mathbf{E}_\mathbf{r}$ and $\mathbf{H}_\mathbf{r}$ the mean macroscopic fields, and the component with $(\epsilon_\infty - 1)$ describes the effect of the polarizability of the ion.

By the methods of[9,11] we can obtain the following expression for $\epsilon_{\alpha\beta}(\mathbf{k}, \omega)$ from (5):

$$\varepsilon_{\alpha\beta}(\mathbf{k}, \omega) = \varepsilon_\infty^{\alpha\beta} + \frac{4\pi}{v_c}\sum_{i,l} e_i e_l G_{Rs}^{i\alpha, l\beta}(\mathbf{k}, \omega). \quad (6)$$

Here G_{Rs} is the Fourier component of the retarded correlator of the displacements $\mathbf{u}^i$ and $\mathbf{u}^l$:

$$G_{Rs}^{i\alpha, l\gamma}(\mathbf{k}, \omega) = i\sum_\mathbf{r}\int_0^\infty dt\, e^{i\omega t - i\mathbf{k}\mathbf{r}}\,\mathrm{Sp}(\rho_0[u_\mathbf{r}{}^{i\alpha}(t), u_0{}^{l\gamma}(0)])_s,$$
$$\rho_0 = e^{-\beta H}/\mathrm{Sp}\, e^{-\beta H}, \quad (7)$$

$\beta = 1/T$, $\mathbf{u}_\mathbf{r}(t) = e^{iHt}\mathbf{u}_\mathbf{r}e^{-iHt}$ and the index s on G indicates that in the Hamiltonian H of the crystal only the short-range part of the interactions is taken into account, but the interaction with the macroscopic field $\mathbf{E}(\mathbf{k}, \omega)$ is not considered. In diagram language, the latter means the absence noted above in graphs for G of single photon lines with frequency ω and momentum $\mathbf{k}$. The complete correlation function $G_R(\mathbf{k}, \omega)$ (which can be studied experimentally, for example, in the scattering of neutrons) connected with G_{Rs} by equation similar to the Maxwell equations in the medium,[4] and are simply expressed in terms of G_{Rs}. If we neglect the anharmonic terms in H in (7) and transform from the displacements $\mathbf{u}^i_\mathbf{r}$ to the normal phonon coordinates $\xi_{\mathbf{k}j}$, by diagonalizing the harmonic part H_0 of the Hamiltonian:[12]

$$\mathbf{u}_\mathbf{r}{}^i = \frac{1}{\sqrt{N}}\sum_{\mathbf{k}j} \mathbf{v}_{\mathbf{k}j}{}^i \xi_{\mathbf{k}j} e^{i\mathbf{k}\mathbf{r}}, \quad H_0 = \frac{1}{2}\sum_{\mathbf{k}j}(\dot\xi_{\mathbf{k}j}\dot\xi_{-\mathbf{k}j} + \omega_{\mathbf{k}j}{}^2\xi_{\mathbf{k}j}\xi_{-\mathbf{k}j}),$$
$$(G_{Rs}^{i\alpha, l\beta}(\mathbf{k}, \omega))_0 = \sum_j \frac{v_{\mathbf{k}j}{}^{i\alpha} v_{-\mathbf{k}j}^{l\beta}}{\omega_{\mathbf{k}j}{}^2 - \omega^2} \quad (8)$$

(where N is the number of cells, $\mathbf{v}^i_{\mathbf{k}j}$ the polarization vectors), then (6) goes over into the well-known dispersion formula for ϵ[10].

In the following, in the discussion of $\epsilon(\mathbf{k}, \omega)$; we limit ourselves for simplicity to ferroelectrics considered in[4] of the displacement type, having a cubic symmetry above the transition (for example, perovskites). In these materials, at low $\mathbf{k}$ and ω, we have $\epsilon(\mathbf{k}, \omega) \gg \epsilon_\infty \sim 1$, and, as was discussed in[4], of all the phonon branches j in G_{Rs}, only the critical, anomalously soft optical branches, which are described by the coordinates $\xi_{kj} = \xi_{k,c\alpha}$ (where $\alpha = 1, 2, 3$ denotes the polarization of the branch at $\mathbf{k} = 0$), are important. For these branches, the anharmonic corrections are important, so that (8) is not directly applicable. Formula (6) takes the form

$$\varepsilon_{\alpha\beta}(\mathbf{k}, \omega) = \lambda G_{Rs}^{\alpha\beta}(\mathbf{k}, \omega),$$

$$\lambda = \frac{4\pi}{3v_c}\Big(\sum_{i,\alpha} e_i v_{c\alpha}{}^i\Big)^2 \equiv \frac{4\pi z_c{}^2}{v_c}. \tag{9}$$

Here $G_{Rs}^{\alpha\beta}$ is determined by Eq. (7) with the substitutions $u^{i\alpha} \to \xi_c^\alpha$ and $u^{l\beta} \to \xi_c^\beta$. A relation of the form (9) was obtained previously[4] by comparison of the equations for G with the Maxwell equations in the medium; however, the relaxation effects and the damping were not taken into account.

The usual method of calculating the retarded functions $G_R(\mathbf{k}, \omega)$ is by finding the temperature Green's function $G(\mathbf{k}, i\omega_n)$ and its analytic continuation from the discrete points $i\omega_n = 2in\pi T$ of the imaginary axis into the complex plane ω.[9,13,2,4] If we write the Dyson equation for G in the usual form:

$$G^{-1}(\mathbf{k}, i\omega_n) = G_0{}^{-1}(\mathbf{k}, i\omega_n) + \Pi(\mathbf{k}, i\omega_n), \tag{10}$$

where Π is the irreducible self-energy part,[9] then the investigation of the singularities of $G_R(\mathbf{k}, \omega)$ at small k and ω of interest to us reduces to the study of the singularities of the analytic continuation of $\Pi(\mathbf{k}, \omega)$. As was discussed, these singularities reflect the effect of thermal relaxation, i.e., processes which, in the system of phonons considered, are described by the kinetic equation. Therefore, the problem reduces to the establishment of the connection of the "single-particle" oscillations, described by $G(\mathbf{k}, \omega)$, with the oscillations of the phonon distribution functions, which are described by the kinetic equation.

Detailed investigations were carried out previously by Éliashberg for the case of a Fermi liquid[13] and by Sham[2] for the case considered, that of a system of phonons. For the study of the singularities of Π, it is sufficient to investigate the difference $\delta\Pi(\mathbf{k}, \omega) = \Pi(\mathbf{k}, \omega) - \Pi(\mathbf{k}, 0)$ and limit ourselves in it to singularities at small ω and k.[13,2] The "isothermal" limit $\Pi(\mathbf{k}, 0) = \Pi(\mathbf{k}, i\omega_n)|_{n=0}$ is calculated according to ordinary perturbation theory and gives the renormalized phonon spectrum; summed with $G_0^{-1}(\mathbf{k}, 0)$, the quantity $\Pi(\mathbf{k}, 0)$ determines the isothermal value $\zeta^T(\mathbf{k}, 0)$ of the inverse susceptibility, according to (9).

Graphically, the equations for the singular part of Π are shown in Fig. 1.[13,2] Here the internal lines of the graphs correspond to the complete Green's functions with account, in particular, of the electromagnetic interaction (5) and the damping. In the lowest approximation of perturbation theory, it suffices, to limit ourselves to graph 1b in the kernel of the integral (kinetic) equation for the vertex, i.e., to an account of three-phonon interactions only, $H_{int}^{(3)}$:

$$H_{int}^{(3)} = \frac{1}{3!}\sum_{\substack{i_1 i_2 i_3\\ \mathbf{p}_1\mathbf{p}_2\mathbf{p}_3}} V^{i_1 i_2 i_3}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3)\,\xi_{\mathbf{p}_1 i_1}\xi_{\mathbf{p}_2 i_2}\xi_{\mathbf{p}_3 i_3}\Delta(\mathbf{p}_1 + \mathbf{p}_2 + \mathbf{p}_3). \tag{11}$$

Here $\Delta(\mathbf{p})$ is equal to unity when p is equal to the reciprocal lattice vector, and equal to zero otherwise.[12]

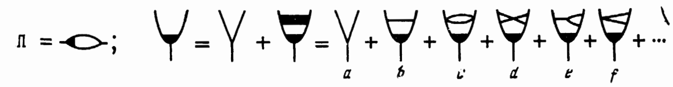

FIG. 1

After analytic continuation with respect to the frequency,[13,2] the equation for $\delta\Pi$ takes the form

$$\delta\Pi_{\alpha\beta} = \frac{1}{T}\sum_{\mathbf{p}i}\frac{1}{2\omega_{\mathbf{p}i}} V^{\alpha ii}\Big(-\mathbf{k}, \frac{\mathbf{k}}{2} - \mathbf{p}, \frac{\mathbf{k}}{2} + \mathbf{p}\Big) N_{\mathbf{p}i}(N_{\mathbf{p}i} + 1) f_{\mathbf{p}i}{}^\beta(\mathbf{k}, \omega), \tag{12}$$

where f_{pi}^β satisfies the equation

$$i(\omega - \mathbf{k}\mathbf{v}_{\mathbf{p}i}) N_{\mathbf{p}i}(N_{\mathbf{p}i} + 1) f_{\mathbf{p}i}{}^\beta = \frac{i\omega}{2\omega_{\mathbf{p}i}} V^{\beta ii}\Big(\mathbf{k}, \mathbf{p} - \frac{\mathbf{k}}{2}, -\mathbf{p} - \frac{\mathbf{k}}{2}\Big) N_{\mathbf{p}i}(N_{\mathbf{p}i} + 1) + \hat{I}(f_{\mathbf{p}i}{}^\beta), \tag{13}$$

and the collision integral $\hat{I}(f)$ has the usual form:[12]

$$\begin{aligned} I(\hat{f}_{\mathbf{p}i}{}^\beta) = \sum_{\mathbf{q}j} I_{\mathbf{p}i,\mathbf{q}j} f_{\mathbf{q}j}^\beta = \frac{\pi}{4}\sum_{\mathbf{q}j,\mathbf{f}l}\Big\{ &\frac{|V^{ijl}(\mathbf{p}, \mathbf{q}, -\mathbf{f})|^2}{\omega_{\mathbf{p}i}\omega_{\mathbf{q}j}\omega_{\mathbf{f}l}}(N_{\mathbf{p}i} + 1)(N_{\mathbf{q}j} + 1) \\ &\times N_{\mathbf{f}l}(f_{\mathbf{p}i}{}^\beta + f_{\mathbf{q}j}{}^\beta - f_{\mathbf{f}l}{}^\beta)\Delta(\mathbf{p} + \mathbf{q} - \mathbf{f})\,\delta(\omega_{\mathbf{p}i} + \omega_{\mathbf{q}j} - \omega_{\mathbf{f}l}) \\ &+ \frac{1}{2}\frac{|V^{ijl}(\mathbf{p}, -\mathbf{q}, -\mathbf{f})|^2}{\omega_{\mathbf{p}i}\omega_{\mathbf{q}j}\omega_{\mathbf{f}l}}(N_{\mathbf{p}i} + 1) N_{\mathbf{q}j} N_{\mathbf{f}l}(f_{\mathbf{p}i}{}^\beta - f_{\mathbf{q}j}{}^\beta - f_{\mathbf{f}l}{}^\beta) \\ &\times \Delta(\mathbf{p} - \mathbf{q} - \mathbf{f})\,\delta(\omega_{\mathbf{p}i} - \omega_{\mathbf{q}j} - \omega_{\mathbf{f}l})\Big\}. \end{aligned} \tag{14}$$

In (12)–(14), $N_{pi} = [\exp(\beta\omega_{pi}) - 1]^{-1}$ is the Bose function, $\mathbf{v}_{pi} = \partial\omega_{pi}/\partial\mathbf{p}$ is the group velocity. In[2], Eq. (3.10), which is similar to (13), has the form of a set of equations for the two functions $X(\mathbf{p}, \omega_p)$, which is analogous to our f_{pi}^β, and $X(\mathbf{p}, -\omega_p)$. However, since the potential $V^{\beta ii}(\mathbf{k}, \mathbf{p} - \mathbf{k}/2, -\mathbf{p} - \mathbf{k}/2)$ is even in p, we can establish the fact that $X(\mathbf{p}, -\omega_p) = X(-\mathbf{p}, \omega_p)$, so that the system reduces to Eq. (13) for f_{pi}^β.[1)] The ratio of the collision term $\hat{I}(f)$ to the left side of (13) is equal to $\overline{\Gamma}/\omega$ or $\overline{\Gamma}/\mathbf{k}\cdot\mathbf{v}$ in order of magnitude, where $\overline{\Gamma}$ is some mean damping of the thermal phonons (for the more precise evaluation, see below). Therefore, in the region of large ω and k ($\omega, \mathbf{k}\cdot\mathbf{v} \gg \Gamma$), we can use perturbation theory and in first approximation, simply discard $\hat{I}(f)$ in (13).

In the case of interest to us, namely the "hydrodynamic" region $\omega, \mathbf{k}\cdot\mathbf{v} \ll \overline{\Gamma}$, the solution of the inhomogeneous kinetic equation will be sought with the help of an expansion in the eigenfunctions φ_{pi}^n of the collision operator

$$f_{\mathbf{p}i}{}^\beta(\mathbf{k}, \omega) = \sum_{n=0}^{\infty} a_n{}^\beta(\mathbf{k}, \omega)\varphi_{\mathbf{p}i}{}^n, \quad \hat{I}(\varphi_{\mathbf{p}i}{}^n) = \gamma_n\varphi_{\mathbf{p}i}{}^n. \tag{15}$$

Here $\gamma_n \geq 0$ is the eigenvalue of the operator $\hat{I}$. Inasmuch as the kernel of the integral equation $I_{pi,qj}$ is symmetric and even relative to change in sign of p and q, the function φ_{pi}^n can be regarded as orthnormalized and possessing a definite parity in p. The function $\varphi_{pi}^0 = \text{const}\cdot\omega_{pi}$ corresponds to the smallest eigenvalue $\gamma_0 = 0$, and describes the change in the equilibrium Bose distribution function for small temperature

1)In passing, we note an error in the text of the Sham paper: [2] in Eq. (2.1), there should be a plus sign instead of minus in front of the first component in the brackets. In correspondence with this, the definitions of the functions Y^+ and Y^- in (3.13) must be interchanged.

changes[12]; in the expansion (15), φ^0 will play a fundamental role. For low temperatures $T \ll \Theta_D$, when the quasi-momentum p is an integral of the motion, with exponential accuracy, the first three odd solutions $\varphi^\nu_{pi} \approx c^\nu_\alpha p_\alpha$ are also important; $\alpha = 1, 2, 3$, the eigenvalues of which γ_ν are exponentially small.[12] For $\omega > \gamma_\nu$ these functions describe phenomena connected with second sound, but in this section, we limit ourselves to the region of small ω and not too small T, for which $\omega \ll \gamma_\nu$.

Substituting the expansion (15) in (13), multiplying (13) by φ^n_{pi}, and summing over p and i, we get

$$i\omega \sum_m S_{nm} a_m{}^\beta - i \sum_m (\mathbf{kv})_{nm} a_m{}^\beta = i\omega V_{n\mathbf{k}}{}^\beta + \gamma_n a_n{}^\beta. \quad (16)$$

Here

$$S_{nm} = \sum_{\mathbf{p}i} \varphi_{\mathbf{p}i}{}^n N_{\mathbf{p}i}(N_{\mathbf{p}i}+1)\varphi_{\mathbf{p}i}{}^m,$$

$$\mathbf{v}_{nm} = \sum_{\mathbf{p}i} \varphi_{\mathbf{p}i}{}^n N_{\mathbf{p}i}(N_{\mathbf{p}i}+1)\mathbf{v}_{\mathbf{p}i}\varphi_{\mathbf{p}i}{}^m,$$

$$V_{n\mathbf{k}}{}^\beta = \sum_{\mathbf{p}i} \varphi_{\mathbf{p}i}{}^n \frac{1}{2\omega_{\mathbf{p}i}} V^{\beta ii}\left(\mathbf{k}, \mathbf{p}-\frac{\mathbf{k}}{2}, -\mathbf{p}-\frac{\mathbf{k}}{2}\right) N_{\mathbf{p}i}(N_{\mathbf{p}i}+1). \quad (17)$$

The condition $\omega, \mathbf{k}\cdot\mathbf{v} \ll \bar{\Gamma}$ is equivalent to the condition $a^\beta_n \ll a^\beta_0 (n > 0)$. In this case, with accuracy to first order in ω and k, we get from (16)

$$a_n{}^\beta = i\frac{(\omega_{0n} - \mathbf{kv}_{0n})a_0{}^\beta - \omega V_{n\mathbf{k}}{}^\beta}{\gamma_n},$$

$$a_0{}^\beta = \omega\left(V_{0\mathbf{k}}{}^\beta + i\sum_{n>0}\frac{\omega_{0n}V_{n\mathbf{k}}{}^\beta}{\gamma_n}\right)\left(\omega_{00} + i\sum_{n>0}\frac{\omega_{0n}{}^2 + (\mathbf{kv})_{0n}{}^2}{\gamma_n}\right)^{-1}. \quad (18)$$

Here and below, $\omega_{mn} = \omega S_{mn}$. The denominator a^β_0 in (18) must generally be expanded in powers of ω, but it more convenient to do this later. Formulas (18) allow us to make more precise the conditions of applicability of the hydrodynamic approximation:

$$\omega_{mn} \ll \gamma_n, \quad (\mathbf{kv})_{mn} \ll \gamma_n, \quad \omega_{00}V_{n\mathbf{k}}{}^\beta \ll \gamma_n V_{0\mathbf{k}}{}^\beta \quad (n>0). \quad (19)$$

Limiting ourselves to the lowest terms in ω and k in the expansion (15), we get from (18)

$$\delta\Pi_{\alpha\beta}(\mathbf{k},\omega) = \frac{\omega}{T}\frac{V^\alpha_{0,-\mathbf{k}}V_{0\mathbf{k}}{}^\beta}{\omega S_{00} + i\sum_{n>0}(\mathbf{kv})_{0n}{}^2/\gamma_n}$$

$$= \frac{\omega}{T^3}\frac{V^\alpha_{0,-\mathbf{k}}V_{0\mathbf{k}}{}^\beta}{v_c C(\omega + i\chi_{\gamma\delta}k_\gamma k_\delta)} \quad (20)$$

In Eq. (20), the normalization constant of the function $\varphi^0_{pi} = \text{const}\cdot\omega_{pi}$ is contracted and for simplification of notation, we shall set $\varphi^0_{pi} = \omega_{pi}$ everywhere below. Here the quantity

$$\frac{1}{T^2 v_c}S_{00} = \frac{1}{T^2 v_c}\sum_{\mathbf{p}i}\omega_{\mathbf{p}i}{}^2 N_{\mathbf{p}i}(N_{\mathbf{p}i}+1)$$

$$= \frac{1}{v_c}\frac{\partial}{\partial T}\sum_{\mathbf{p}i}\omega_{\mathbf{p}i}N_{\mathbf{p}i}$$

is the limiting heat capacity C of the Bose gas of phonons. The quantities $\chi_{\gamma\delta}$, as is seen from the comparison given below with the results of Sec. 2, represent temperature conductivity tensor;

$$\chi_{\gamma\delta} = \frac{1}{S_{00}}\sum_{n>0}\frac{v_{0n}{}^\gamma v_{0n}{}^\delta}{\gamma_n}. \quad (21)$$

The terms in $\delta\Pi$ following in ω, omitted in (20), describe the effect of the viscosity of the phonon gas and are discussed in Sec. 4.

In the derivation of Eqs. (12)–(21), the specific features of the ferroelectrics were not employed, so that by α and β in them we meant arbitrary phonon ranches. It is seen from (20) and (17) that the effect of the thermal relaxation on the dispersion of the optical branches depends on the symmetry of the crystal In "central" crystals, in which each atom is the center of symmetry, the three-phonon potential $V^{\alpha ii}(\mathbf{k}, -\mathbf{p}-\mathbf{k}/2, -\mathbf{p}-\mathbf{k}/2)$ is proportional to k when $\mathbf{k} \to 0$,[14,15] so that $\delta\Pi$ in (20) is proportional to k^2 and is small. For a noncentral crystal, $\delta\Pi(0, \omega) \neq 0$ and thermal dispersion exists.

The ferroelectrics considered are central above the transition point T_0; here the polarization $\mathbf{P} = 0$ and, in accord with (3), $\xi^S = \xi^T$. We shall show that the relations (20), (10), and (9) also transform into (3) below T_0. In the given case, the total heat capacity, except for the phonon part C, which is taken into account in (20), contains contributions connected with the dependence of the polarization P on T, so that C in (20) represents the heat capacity C_p. Moreover, a certain contribution to C gives the dependence of the critical frequencies ω_{pc} on T, which was not taken into account in (20); however, in the heat capacity, this gives a small correction of higher order in the anharmonicity. Therefore, to prove the identity of (20), (10) and (9) with (3) as $\mathbf{k} \to 0$, it suffices to prove the relation

$$\frac{1}{T^2 z_c}V_{0\mathbf{k}}{}^\alpha\Big|_{\mathbf{k}=0} = \frac{1}{z_c}\sum_{\mathbf{p}i}\frac{\partial N_{\mathbf{p}i}}{\partial T}V^{\alpha ii}(0,\mathbf{p},-\mathbf{p})$$

$$= \frac{\partial E_\alpha}{\partial T} = \frac{\partial^2 F}{\partial P_\alpha \partial T}, \quad (22)$$

where Z_c is the same as in (9). The free energy $F(\mathbf{P}, T)$ in the materials considered was studied in[4]. Figure 2 shows several graphs for F contributing to $\partial E/\partial T$, taken from Eqs. (15) of[4].

The circles on this drawing correspond to the mean equilibrium value $\langle x_c \rangle = \mathbf{P}v_c z_c^{-1}$. The analogous graphical expansion of the vertex $V_3^\alpha = V^{\alpha ii}(0, \mathbf{p}, -\mathbf{p})$ is also given. We consider, for example, the first components a and a′ in these expansions. Comparing the graphs and the analytic expressions, we get

$$F^{(a)} = \frac{v_c}{4}\sum_{\mathbf{p}i}V^{\alpha\beta ii}(0,0,\mathbf{p},-\mathbf{p})\frac{N_{\mathbf{p}i}}{\omega_{\mathbf{p}i}}\frac{P_\alpha P_\beta}{z_c{}^2}$$

$$V_3^{(a')\alpha} = \frac{v_c P_\beta}{2z_c\omega_{\mathbf{p}i}}V^{\alpha\beta ii}(0,0,\mathbf{p},-\mathbf{p}). \quad (23)$$

Here $V^{\alpha\beta ii}$ is the 4-phonon vertex of interaction of the critical-polarization phonons α and β with the phonons of the i-th branch. It is seen that the quantities (23) are connected by the relation (22). Termwise equality of the remaining components of Fig. 2 is proved similarly.

Thus, the considered dispersion of ϵ exists only below the transition point and is described by Eqs. (3) and (20). A similar problem was considered earlier[1] qualitatively using a model example, by the method of Mandel'shtam-Leontovich, with introduction of the phenomenological relaxation time τ^{-1}. Comparison of (3) and (20) with the results of[1] shows that one can

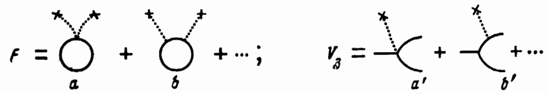

FIG. 2

describe the thermal relaxation by formulas of the Mandel'shtam-Leontovich type if one takes the quantity $\chi_{\alpha\beta}k_\alpha k_\beta$ to mean the relaxation frequency τ^{-1}.

As was already noted, in high-frequency measurements of the permittivity $\epsilon(\mathbf{k}, \omega)$, usually only the adiabatic regime $\omega \gg \chi k^2$ is realized, so that one can observe the considered "optical" thermal dispersion only in experiments on inelastic scattering, which are discussed in Sec. 5. The corresponding contributions to the cross section can be described by using (34), (10), (20), and (9). The answers are similar to those given in (35) below; the fundamental difference is that the given dispersion appears only below the transition point and is significant only in transverse branches (and not in the longitudinal, as in (35)), inasmuch as only these branches are low-lying in the given ferroelectrics.[4]

4. DISPERSION AND DAMPING OF FIRST AND SECOND SOUND

The expression for the complex elastic modulus tensor $c_{\alpha\beta\gamma\delta}(\mathbf{k}, \omega)$ in terms of the Green's function can be found by the usual methods.[9,11] We define the acoustical coordinate $\mathbf{u}_\mathbf{r}$, for example, as the coordinate of the center of gravity of the cell:[14]

$$\mathbf{u}_\mathbf{r} = \Big(\sum m_i\Big)^{-1} \sum m_i \mathbf{u}_\mathbf{r}^i.$$

Then the Hamiltonian of the interaction with the external force density $\mathbf{f}$ is written in the form

$$V(t) = -\sum_\mathbf{r} \mathbf{u}_\mathbf{r}\mathbf{f}(\mathbf{r}, t)v_c.$$

Integrating in the usual way[11] the equation for the matrix density $\rho = \rho_0 + \rho'$ in the field of the small external force $\mathbf{f}(\mathbf{r}, t) = \mathbf{f}' e^{i\mathbf{k}\cdot\mathbf{r} - i\omega t}$, we have

$$u_\mathbf{r}'^\alpha = \mathrm{Sp}(u_\mathbf{r}^\alpha \rho') = iv_c \sum_{\mathbf{r}'} \int_{-\infty}^{t} dt' f_\beta'(\mathbf{r}', t')$$
$$\times \mathrm{Sp}(\rho_0[u_\mathbf{r}^\alpha(t), u_{\mathbf{r}'}^\beta(t')]) = D_R^{\alpha\beta}(\mathbf{k}, \omega) f_\beta' v_c e^{i\mathbf{kr} - i\omega t}. \tag{24}$$

Here ρ_0 is the same as in (7) and $D_R^{\alpha\beta}$ is the retarded Green's function of the components u^α and u^β. Comparing (24) with the macroscopic equations (4a), we find

$$-\rho\omega^2\delta_{\alpha\beta} + c_{\alpha\gamma\delta\beta}(\mathbf{k}, \omega) k_\gamma k_\delta = v_c^{-1}(D_R^{-1}(\mathbf{k}, \omega))_{\alpha\beta}. \tag{25}$$

The Dyson equation for D has the form (10), with the substitution $G \to D$. By defining the isothermal tensor $c_{\alpha\beta\gamma\delta}k_\gamma k_\delta$ as

$$v_c^{-1}D_{\alpha\beta}^{-1}(\mathbf{k}, 0) = v_c^{-1}D_{\alpha\beta}^{-1}(\mathbf{k}, i\omega_n)\big|_{n=0},$$

we have, from (10) and (25),

$$[c_{\alpha\gamma\delta\beta}(\mathbf{k}, \omega) - \overset{T}{c}_{\alpha\gamma\delta\beta}] k_\gamma k_\delta = v_c^{-1}(\Pi_{\alpha\beta}(\mathbf{k}, \omega)$$
$$- \Pi_{\alpha\beta}(\mathbf{k}, 0)) \equiv v_c^{-1}\delta\Pi_{\alpha\beta}(\mathbf{k}, \omega). \tag{26}$$

Here $\Pi_{\alpha\beta}(\mathbf{k}, \omega)$ is the analytic continuation of the irreducible self-energy part of the Green's temperature function $D_{\alpha\beta}(\mathbf{k}, i\omega_n)$.

Using (26) and the results of Sec. 3, we consider the effect of thermal relaxation on the elastic moduli and the sound propagation. We shall show first that Eqs. (26) and (20) are identical with (4b) when $\mathbf{k}, \omega \to 0$. To this end, we note that the quantity $V_{0\mathbf{k}}^\alpha$ in (20) has the form

$$V_{0\mathbf{k}}^\alpha = -iT^2 k_\beta \frac{\partial}{\partial T} \sum_{\mathbf{p}j} N_{\mathbf{p}j}\omega_{\mathbf{p}j}^2 \lambda_{\mathbf{p}j}^{\alpha\beta},$$

$$\lambda_{\mathbf{p}j}^{\alpha\beta} = \frac{i}{2\omega_{\mathbf{p}j}^2} \frac{\partial}{\partial k_\beta} V^{\alpha jj}\Big(\mathbf{k}, \mathbf{p} - \frac{\mathbf{k}}{2}, -\mathbf{p} - \frac{\mathbf{k}}{2}\Big)\Big|_{\mathbf{k}=0} \tag{27}$$

for small $\mathbf{k}$. Here it is taken into account that the potential of interaction with the acoustic branch α vanishes linearly with $\mathbf{k}$ as $\mathbf{k} \to 0$[10,12] and $\lambda_{\mathbf{p}j}^{\alpha\beta}$ is the Grüneisen coefficient, which determines the change in the spectrum for deformation of the crystal:[5,16]

$$\omega_{\mathbf{p}j}(u_{\alpha\beta}) = \omega_{\mathbf{p}j}(0)(1 - u_{\alpha\beta}\lambda_{\mathbf{p}j}^{\alpha\beta}).$$

The term δF in the free energy that is linear in the deformation and determines the thermal expansion,[4] and the corresponding derivative $\partial S/\partial u_{\alpha\beta}$ in (4b) are equal to

$$\delta F = -\frac{u_{\alpha\beta}}{v_c} \sum_{\mathbf{p}i} \omega_{\mathbf{p}i} \lambda_{\mathbf{p}i}^{\alpha\beta} \Big(N_{\mathbf{p}i} + \frac{1}{2}\Big),$$

$$\frac{\partial S}{\partial u_{\alpha\beta}} = \frac{1}{v_c}\frac{\partial}{\partial T} \sum_{\mathbf{p}i} N_{\mathbf{p}i}\omega_{\mathbf{p}i}\lambda_{\mathbf{p}i}^{\alpha\beta}. \tag{28}$$

The heat capacity of the phonon gas C in (20) does not take into account the temperature dependence of $u_{\alpha\beta}$, i.e., it is the heat capacity at constant deformation C_u. Thus, with account of (27) and (28), Eqs. (20) and (26) are identical with (4b).

In the propagation of ordinary "first" sound in a solid, the quantity k is equal to ω/u_s, where u_s is the sound velocity. In this case, at small ω and not too low T, when the inequalities (19) are satisfied, the sound wave is adiabatic, $\omega \gg \chi k^2$, as can be established by estimating χ by means of (21). We find the expression (26) for $\delta\Pi$ in this case, with accuracy to within linear terms in ω. Using (12), (15), (18), and (20), we get

$$\frac{1}{v_c}\delta\Pi_{\alpha\beta} = k_\gamma k_\delta \Big\{ \frac{\lambda_0^{\alpha\gamma}\lambda_0^{\beta\delta}}{Tv_c S_{00}} - i\omega \Big[\frac{\lambda_0^{\alpha\gamma}\lambda_0^{\beta\delta}}{Tv_c S_{00}}$$
$$\times \sum_{n>0} \frac{(\mathbf{kv})_{0n}^2}{\gamma_n \omega^2} + \frac{1}{Tv_c S_{00}^2} \sum_{n>3} \frac{1}{\gamma_n} (S_{0n}\lambda_0^{\alpha\gamma}$$
$$- S_{00}\lambda_n^{\alpha\gamma})(S_{0n}\lambda_0^{\beta\delta} - S_{00}\lambda_n^{\beta\delta})\Big]\Big\}. \tag{29}$$

Here it has been taken into account that $V_{n\mathbf{k}}^\alpha = ik_\gamma\lambda_n^{\alpha\gamma}$, where

$$\lambda_n^{\alpha\gamma} = \sum_{\mathbf{p}i} \varphi_{\mathbf{p}i}^n N_{\mathbf{p}i}(N_{\mathbf{p}i} + 1)\omega_{\mathbf{p}i}\lambda_{\mathbf{p}i}^{\alpha\gamma}.$$

Since $\omega_{\mathbf{p}i}$ is even in $\mathbf{p}$, the first and second components in the square brackets contain only odd and even functions $\varphi_{\mathbf{p}i}^n$, respectively.

The expression in the square brackets in (29) describes the sound damping. The first term in these brackets gives the damping which results from the thermal conductivity, the so-called thermoelastic damping, which can be expressed in terms of thermodynamic quantities and $\chi_{\alpha\beta}$. The second component, as is evident from comparison with (4b), is the viscosity tensor $\eta_{\alpha\beta\gamma\delta}$. The damping expression, which is analogous to (29), was obtained by Shklovskiĭ[5] in the τ-approximation. It was noted by him that the ratio of the thermo-elastic damping to the viscous damping is proportional to the square of the ratio of group velocity, averaged in some way, to the sound velocity u_s, which can explain the smallness of this ratio at high temperatures in crystals with small anisotropy[5,16]. Formula (29) gives the quantitative expression for this assertion.

Up to now, we have considered the case for not too low temperatures and small ω. For low $T \ll \Theta_D$, as was recalled above, three first values of γ_ν, $\nu = 1, 2, 3$, corresponding to the functions $\varphi^\nu_{pi} = c^\nu_\alpha p_\alpha$, tend exponentially to zero, inasmuch as only Umklapp processes make a contribution to the quantity γ_ν, in accord with (15).[12] Therefore, even at low frequencies, the conditions (19) for $n = 1, 2, 3$ will be violated, while the hydrodynamic condition of (19) will be satisfied for the remaining "normal" γ_n with $n > 3$, which fall off with T in a power-law fashion. By assuming that (19) is valid only for $n > 3$, and the relation among ω, $\mathbf{k}$ and γ_ν is arbitrary, we can obtain, by the same method as above, the general expression for $D_R^{-1}(\mathbf{k}, \omega)$, which describes the dispersion of the sound velocity, of the viscosity and the thermal conductivity over the whole range of temperatures T and "hydrodynamic" k and ω:

$$(D_R^{-1}(\mathbf{k}, \omega))_{\alpha\beta} = -M\omega^2\delta_{\alpha\beta} + v_c \overset{T}{c}_{\alpha\gamma\delta\beta} k_\gamma k_\delta$$
$$+ \frac{\omega}{T} k_\gamma k_\delta \left[\frac{\lambda_0^{\alpha\gamma}\lambda_0^{\beta\delta} + i\Lambda^{\alpha\gamma\delta\beta}(\mathbf{k}, \omega)}{\omega S_{00} - (\mathbf{kv})_{0\nu}\Sigma_{\nu\mu}(\mathbf{kv})_{\mu 0} + 2iS_{00}\Gamma(\mathbf{k}, \omega)} - i\sum_{n>3} \frac{\lambda_n^{\alpha\gamma}\lambda_n^{\beta\delta}}{\gamma_n} \right]. \tag{30}$$

Here M is the mass of the cell,

$$\Lambda^{\alpha\gamma\delta\beta}(\mathbf{k}, \omega) = \sum_{n>3} [\omega_{0n} - (\mathbf{kv})_{0\mu}\Sigma_{\mu\nu}(\mathbf{kv})_{\nu n}](\lambda_0^{\alpha\gamma}\lambda_n^{\beta\delta} + \lambda_n^{\alpha\gamma}\lambda_0^{\beta\delta}), \tag{31}$$

$$2S_{00}\Gamma(\mathbf{k}, \omega) = \sum_{n>3} \gamma_n^{-1} \Big\{ \omega_{0n}^2 + (\mathbf{kv})_{0n}^2 - 2(\mathbf{kv})_{0\mu}\Sigma_{\mu\nu}[\omega_{\nu n}(\mathbf{kv})_{n0} + (\mathbf{kv})_{\nu n}\omega_{n0}]$$
$$+ (\mathbf{kv})_{0\lambda}\Sigma_{\lambda\mu}[\omega_{\mu n}\omega_{n\nu} + (\mathbf{kv})_{\mu n}(\mathbf{kv})_{n\nu}]\Sigma_{\nu\rho}(\mathbf{kv})_{\rho 0} \Big\}. \tag{32}$$

The matrix $\hat{\Sigma} = (\hat{\omega} + i\hat{\gamma})^{-1}$, where $\hat{\omega} + i\hat{\gamma}$ is a 3×3 matrix of the quantities $\omega_{\mu\nu} + i\gamma_{\mu\nu} = \omega S_{\mu\nu} + i\gamma_\nu\delta_{\mu\nu}$. In place of the basis functions φ^ν, in which the γ matrix is diagonal, one can use in (30)—(32) any other orthonormalized linear combination of components of the quasi-momentum p_α, for example, such in which not $\gamma_{\mu\nu}$ but $\omega_{\mu\nu} = \omega S_{\mu\nu}$ is diagonal; this can be convenient in the description of second sound. The quantities Λ and Γ are successive terms in ω and $\mathbf{k}$ of the expansion of the numerator and denominator and in sound propagation they describe the damping processes. For compactness in the formula (30), as above, it is more convenient not to expand the denominator in powers of k and ω, i.e., to keep the component with Γ in the denominator.

For $\hat{\omega} \ll \hat{\gamma}$, we obtain from (30)—(32), as before, the results (21) for the thermal conductivity and (29) for the sound propagation. For $\hat{\omega} \gg \hat{\gamma}$ (which, with account of (19), is possible only for $T \ll \Theta_D$) formulas (30)—(32) describe phenomena connected with second sound. Here we use the results obtained previously by other methods,[2,3,17] and also the microscopic expression for the phenomenological parameters introduced earlier.[3] In the given region, the values of λ_n are small, like $T^5\Theta_D^{-5}$, so that, in accord with (30), D_R has a pole close to the pole point of $\delta\Pi$, i.e., close to the point $\omega^2 = \rho_{\alpha\beta}k_\alpha k_\beta$, where $\rho_{\alpha\beta} = v_{0\mu}S^{-1}_{\mu\nu}v^\beta_{\nu 0} \cdot S^{-1}_{00}$. This equation determines the velocity of second sound and is identical with the result of[17] (if in the expressions for $B_{\alpha\beta}$ in (17) and (34) of that reference we introduce under the summation sign the factor ω_{ks}, which was omitted in the transition from (8) to (17)). In particular, for a cubic crystal, in which $\rho_{\alpha\beta} = \delta_{\alpha\beta}c^2_{II}$, a formula is obtained which is similar to the case of an isotropic body:[17]

$$c_{II}^2 = \frac{\overline{u_s^{-3}}}{3\overline{u_s^{-5}}}, \quad \overline{u_s^{-m}} = \frac{1}{3}\sum_{i=1}^{3} \int \frac{d\mathbf{n}}{4\pi} u_{si}^{-m}(\mathbf{n}), \tag{33}$$

where $\mathbf{n} = \mathbf{p}/p$ and summation is carried out over the three acoustic branches.

The damping Γ_{II} of second sound is composed of Γ^u_{II}, which is connected with scattering processes, and the "normal" scattering Γ^n_{II}, which is proportional to ω^2. The damping Γ^u_{II} is obtained from expansion of the quantity

$$(\hat{\omega} + i\hat{\gamma})^{-1} = \hat{\omega}^{-1} - i\hat{\omega}^{-1}\hat{\gamma}\hat{\omega}^{-1}$$

in the second component of the denominator of (30). Inasmuch as the thermal conductivity (21) is determined from the components with γ_ν at these temperatures,

$$S_{00}\chi_{\alpha\beta} = v_{0\nu}^\alpha (\hat{\gamma})_{\nu\mu}^{-1} v_{\mu 0}^\beta,$$

then the damping Γ^u_{II} can be expressed in terms of the coefficient of thermal conductivity and the matrix $\rho_{\alpha\beta}$ introduced above, which determines the velocity of second sound:[3]

$$2\Gamma_{II}^u = \omega^{-2}k_\alpha k_\beta \rho_{\alpha\gamma}\chi_{\gamma\delta}^{-1}\rho_{\delta\beta}.$$

The value of Γ^n_{II} is given by the expression $\Gamma(\mathbf{k}, \omega)$ in (32), if we neglect $\hat{\gamma}$ in series with $\hat{\omega}$ in $\hat{\omega} + i\hat{\jmath}$ in it, and set $\omega^2 = k_\alpha k_\alpha \rho_{\alpha\beta}$. Components connected with the numerator of (30) are proportional to the quantities λ_n and λ_0, and in the velocity and in the damping give small corrections of relative order $T^4\Theta_D^{-4}$, which can be established by estimating the corresponding integrals.

On going (with change of ω or T) through the region $\hat{\omega} \sim \hat{\gamma}$ the collective oscillation, second sound, transforms into a purely relaxational "heat conduction mode": the pole denominator in (30) transforms into the expression $\omega + i\chi_{\alpha\beta}k_\alpha k_\beta$ of Eq. (20). Here, a significant dispersion of the kinetic coefficients—thermal conductivity, viscosity, sound absorption—also takes place. The phenomena appearing here were discussed phenomenologically by Gurevich and Éfros.[3] Equations (30)—(32) make possible a unified study of the corresponding dispersions and description of the transition region $\hat{\omega} \sim \hat{\gamma}$.

5. SINGULARITIES IN THE INELASTIC SCATTERING CROSS SECTIONS AT SMALL ENERGY AND QUASI-MOMENTUM TRANSFER

We shall consider how the hydrodynamic dispersion described above arises in inelastic scattering of neutrons or light from a crystal. The Van Hove formula (see, for example,[18]) for the cross section of coherent scattering of a neutron of energy ϵ, with the energy ω and momentum $\mathbf{q} = \mathbf{G} + \mathbf{k}$ transferred to the crystal (here $\mathbf{G}$ is the reciprocal lattice vector and $\mathbf{k}$ lies in the first Brillouin zone), can be transformed by the methods of[9,19] to the form

$$\frac{d^2\sigma^{\mathrm{coh}}}{d\Omega dE} = \sqrt{\frac{\varepsilon-\omega}{\varepsilon}} \sum_{ij\mathbf{R}} a_i a_j^* \int_{-\infty}^{\infty} \frac{dt}{2\pi} \exp(i\omega t)$$

$$\times \mathrm{Sp}[\rho_0 \exp\{-i\mathbf{q}\mathbf{R}_i(0)\} \exp\{i\mathbf{q}\mathbf{R}_j(t)\}] = \sqrt{\frac{\varepsilon-\omega}{\varepsilon}} \sum_{ij} a_i a_j^*$$

$$\times \exp\{i\mathbf{q}(\rho_j - \rho_i)\} \frac{q_\alpha q_\gamma \operatorname{Im} G_R^{i\alpha, j\gamma}(\mathbf{k}, \omega)}{2\pi(1-\exp(-\beta\omega))}. \qquad (34)$$

Here a_i is the amplitude (length) of the coherent scattering by the i-th atom of the cell, ρ_i is the equilibrium coordinate of the i-th atom in the cell, the quantities $\mathbf{q}\cdot\mathbf{u}$ are considered small, so that the exponents $\exp(i\mathbf{q}\cdot\mathbf{u})$ are expanded in a series, and G_R is the Fourier component of the retarded Green's function of the displacements $u_{\mathbf{r}}^{i\alpha}(0)$ and $u_{\mathbf{r}}^{j\gamma}(t)$, which is determined by Eq. (7) (without the index s). In light scattering, it is necessary to omit the factor $(\epsilon-\omega)^{1/2}\epsilon^{-1/2}$ in (34), and to replace the scattering length a_i by the electromagnetic amplitude $(e^2/mc^2)F_i$, where F_i is the form factor of the i-th ion.[12]

For small $\mathbf{k}$, ω, the largest contribution to (34) is made by the acoustic branches. In the case of a neutral symmetric crystal, the atoms of the cell in these branches at small $\mathbf{k}$ move as a unit, and one can use as acoustic coordinates, as above, the coordinates of the center of mass of the cell (and for the optical, any set of relative coordinates[14]). In this case, $G_R^{i\alpha,j\gamma}$ in (34) is replaced by the quantity $D_R^{\alpha\gamma}$ (25), (30).

We shall illustrate the character of the resultant singularities in the dependence of the cross section on $\mathbf{k}$ and ω by an example of an isotropic solid. We consider initially not very low T, when for $c_{\alpha\beta\gamma\delta}(\mathbf{k}, \omega)$ in (25) we can use Eq. (4b). Setting

$$\chi_{\alpha\beta} = \chi\delta_{\alpha\beta}, \quad \eta_{\alpha\gamma\delta\beta}k_\gamma k_\delta = k_\alpha k_\beta \eta_l + (k^2\delta_{\alpha\beta} - k_\alpha k_\beta)\eta_t,$$

we have in this case

$$\frac{d^2\sigma^{\mathrm{coh}}}{d\Omega dE} = \sqrt{\frac{\varepsilon-\omega}{\varepsilon}} \left| \sum_i a_i \exp(i\mathbf{q}\rho_i) \right|^2 \frac{T}{2\pi M} \left\{ (\mathbf{qk})^2 \left(\frac{\eta_l}{\rho} + \frac{\alpha u_{lS}\chi k^2}{\omega^2+\chi^2k^4} \right) \right.$$

$$\times \left[\left(k^2u^2_{lT} + \alpha u^2_{lS} \frac{k^2\omega^2}{\omega^2+\chi^2k^4} - \omega^2 \right)^2 + \omega^2k^4 \left(\frac{\eta_l}{\rho} + \frac{\alpha u_{lS}^2\chi k^2}{\omega^2+\chi^2k^4} \right)^2 \right]^{-1}$$

$$\left. + \frac{[k^2G^2 - (\mathbf{kG})^2]\eta_t/\rho}{(k^2u_t^2-\omega^2)^2 + \omega^2k^4\eta_t^2/\rho^2} \right\}. \qquad (35)$$

Here u_{lT} and u_{lS} are the isothermal and adiabatic velocities of longitudinal sound; u_t is the velocity of transverse sound; the quantities $\alpha = 1 - u_{lT}^2 u_{lS}^{-2}$ is proportional to the difference $C_pC_V^{-1} - 1$ and for a solid it is small.[7,2] In the transition from (34) to (35), it is taken into account that for the hydrodynamic $\omega \ll \overline{\Gamma}$ considered, the quantity $\beta\omega = \omega/T \ll 1$.

Substitution in (34) of the general formula (30) allows us also to investigate the effect on the scattering of the dispersion associated with the appearance of second sound. Thus, in the region of low T and not too large ω, in which the viscous and the normal damping $\Gamma_{\mathrm{II}}^{\mathrm{n}}$ are small, of comparatively the same order as the thermoelastic damping and $\Gamma_{\mathrm{II}}^{\mathrm{u}}$, so that one can neglect the terms $\Gamma(\mathbf{k}, \omega)$, $\Lambda(\mathbf{k}, \omega)$ and the last component in (30), Eq. (34) in the elastic, isotropic case takes the form

$$\frac{d^2\sigma^{\mathrm{coh}}}{d\Omega dE} = \sqrt{\frac{\varepsilon-\omega}{\varepsilon}} \left| \sum_i a_i e^{i\mathbf{q}\rho_i} \right|^2 \frac{T}{2\pi M} \left\{ (\mathbf{qk})^2 \chi\alpha u_{lS}^2 c_{\mathrm{II}}^4 k^2 \right.$$

$$\times [\omega^2 c_{\mathrm{II}}^4 (k^2u_{lS}^2 - \omega^2)^2 + \chi^2 [(k^2u_{lS}^2 - \omega^2)(k^2c_{\mathrm{II}}^2 - \omega^2)$$

$$\left. - \alpha u_{lS}^2 c_{\mathrm{II}}^2 k^4]^2]^{-1} + R_t \right\} \qquad (36)$$

Here R_t denotes the second, transverse component in the curly brackets in (35), and the speed of second sound c_{II} is given by Eq. (33) with u_{Si} independent of n. It is seen that for $\chi\omega > c_{\mathrm{II}}^2$ there is a maximum in the cross section (36) corresponding to second sound, although its intensity is approximately $1/\alpha$ that of first sound. For $\chi\omega \ll c_{\mathrm{II}}^2$, (36) goes over into (35) with $\eta_l = 0$.

The relations (35) and (36) describe the shape of the lines of the quasielastic peaks in the hydrodynamic region ω, $\mathbf{k}\cdot\mathbf{v} \ll \overline{\Gamma}$, and are similar to the corresponding expressions for the liquid, which are discussed, for example, in[20]. The quantitative difference consists fundamentally in the fact that in a solid the given region is always narrow (compared, for example, with the Debye temperature Θ_D) which makes its experimental study difficult. However, in principle, all the dispersion discussed here can be studied in scattering experiments. The dispersion connected with the thermal conductivity is important in the region of comparatively large $\mathbf{k}$ and small $\omega \sim \chi k^2$, in which the viscosity, account of which is necessary in sound damping, is unimportant.

In conclusion, we note that our results illustrate the considerations of Kadanoff and Martin[20] on the singularities of the correlation functions $G(\mathbf{k}, \omega)$ in the region of hydrodynamic ω and $\mathbf{k}$. From considerations of correspondence with hydrodynamics, these authors proved the presence of singularities of G for small $\mathbf{k}$ and ω and noted that the nontrivial form of these singularities can indicate the difficulty of the microscopic calculation of G in the given region. Our results show how one can complete these calculations in the case of a solid. This example can be useful in the consideration of more complicated phenomena, for example, in the study of relaxation in spin systems.

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