THEORY OF LIGHT SCATTERING NEAR SECOND ORDER PHASE TRANSITIONS

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It is shown that in the study of molecular scattering of light near points of second order phase transitions, in addition to the fluctuations of the characteristic parameter, it is also necessary to take into account certain other quantities, for example, the density of the material. The total and spectral intensities of the scattered light are calculated. In the formula for the total intensity, account of the fluctuations of quantities that are different from the characteristic parameter leads to a change in the constant coefficient, and this change is shown to be very important as a rule. The effect of the phase transition on the spectral composition of the light scattered by fluctuations of quantities which differ from the characteristic parameter is studied. The behavior of the Mandel'shtam-Brillouin component is studied in great detail. It is shown under what conditions the position of the components can change significantly near the phase transition point.

FLUCTUATIONS of the characteristic parameter of a second-order phase transition increase in the neighborhood of the transition point, as is well known.[1] This is made manifest, in particular, in the increase in the intensity of scattered light.[2 3] In the theoretical consideration of this phenomenon,[2, 4-7] attention was paid only to fluctuations of the characteristic parameter. Together with this, in a number of cases, the fluctuations of the characteristic parameter and of other quantities cannot be regarded as independent. The present work consists essentially in the generalization of the results of researches[4-7] to include account of this circumstance.

It will be shown below that near the phase transition point, fluctuations increase not only in the characteristic parameter, but also in certain quantities which differ from it. Although the contribution of the fluctuations of quantities which differ from the characteristic parameter can be as important or even more important than the contribution of the fluctuations of the characteristic parameter, in the final formula for the total intensity, this appears only in the more or less considerable renormalization of one of the coefficients. Accurate to the given renormalization, the conclusions obtained in [6] relative to the temperature evolution of the spectral composition of the light scattered by the fluctuations of the transition parameter remain in force. So far as the spectral composition of the light scattered by the fluctuations of the other quantities is concerned in other words, the spectral components which are not connected with the fluctuations of the characteristic parameter, then it is seen that the presence of the phase transition has a significant effect on their location only when the frequencies corresponding to these components are sufficiently low.

1. Let us consider the fluctuations, close to the transition point, of quantities which differ from the characteristic parameter (η). Let ξ be one of these quantities. We represent it in the form

$$\xi = \xi_0(\eta) + \xi', \tag{1}$$

where $\xi_0(\eta)$ represents the "equilibrium" value of ξ, corresponding to a given fixed value of η (see below, Eq. (4)). From (1) we have for ξ

$$\Delta\xi = \Delta\xi_0(\eta) + \Delta\xi' = (d\xi_0/d\eta)_{\eta=\eta_0}\Delta\eta + \Delta\xi', \tag{2}$$

where η_0 is the value of η in the state of complete thermodynamic equilibrium.

Expanding the density of the thermodynamic potential $\Phi(P, T, \xi, \eta)$ in a series about the point $\xi = \xi_0(\eta_0)$, $\eta = \eta_0$, and substituting Eq. (2) in place of $\Delta\xi$, we obtain

$$\begin{aligned}\Delta\Phi &= \frac{1}{2}\left[\left(\frac{\partial^2\Phi}{\partial\eta^2}\right)_0(\Delta\eta)^2 + 2\left(\frac{\partial^2\Phi}{\partial\xi\,\partial\eta}\right)_0\Delta\eta\,\Delta\xi + \left(\frac{\partial^2\Phi}{\partial\xi^2}\right)_0(\Delta\xi)^2\right] \\ &= \frac{1}{2}[a(\Delta\eta)^2 + 2b\,\Delta\eta\,\Delta\xi + c(\Delta\xi)^2] = \frac{1}{2}\left\{\left[a + 2b\left(\frac{d\xi_0}{d\eta}\right)_{\eta=\eta_0}\right.\right. \\ &\left.\left. + c\left(\frac{d\xi_0}{d\eta}\right)^2_{\eta=\eta_0}\right](\Delta\eta)^2 + 2\left[b + \left(\frac{d\xi_0}{d\eta}\right)_{\eta=\eta_0}\right]\Delta\xi'\Delta\eta + c(\Delta\xi')^2\right\} \\ &= \frac{1}{2}[\tilde{a}(\Delta\eta)^2 + 2\tilde{b}\,\Delta\eta\,\Delta\xi' + c(\Delta\xi')^2].\end{aligned} \tag{3}$$

The coefficient $\tilde{b}$ in (3) is equal to zero, as can easily be seen, and consequently, the quantities ξ' and η are statistically independent ($\langle\Delta\xi'\Delta\eta\rangle = 0$, the symbol $\langle\ \rangle$ denotes averaging). In fact, $\xi_0(\eta)$ is determined from the equation

$$(\partial\Phi/\partial\xi)_{\xi=\xi_0(\eta)} = 0. \tag{4}$$

Differentiating both parts of (4) with respect to η and setting $\eta = \eta_0$ in the result, we obtain

$$\left(\frac{\partial^2\Phi}{\partial\xi\,\partial\eta}\right)_0 + \left(\frac{\partial^2\Phi}{\partial\xi^2}\right)_0\left(\frac{\partial\xi_0}{d\eta}\right)_{\eta=\eta_0} = \tilde{b} = 0. \tag{5}$$

For the fluctuation ξ we now have

$$\langle(\Delta\xi)^2\rangle = (d\xi_0/d\eta)^2_{\eta=\eta_0}\langle(\Delta\eta)^2\rangle + \langle(\Delta\xi')^2\rangle. \tag{6}$$

Close to the phase transition point, as follows from (3) and (5),

$$\langle(\Delta\eta)^2\rangle = \frac{k_BT}{V}\frac{1}{\tilde{a}} \sim \frac{1}{|T-\Theta|}, \tag{7}$$

where Θ is the transition temperature. Therefore, if the value of $(d\xi_0/d\eta)_{\eta=\eta_0}$ falls off close to the phase transition point more slowly than $\sim|T-\Theta|$, the fluctuations of ξ increase as $T \rightarrow \Theta$. On the other hand,

$$\langle(\Delta\xi')^2\rangle = \frac{k_B T}{V}\frac{1}{c},$$

and the coefficient c does not depend in any material way on the temperature (in the framework of the Landau theory,[8] which we shall use everywhere below, we can set $c(T) = c(\Theta)$).

Thus, in the calculation of the intensity of the anomalous scattering near the phase transition point, it suffices to take into account only the first term in (2).

The problem of the character of the change of the fluctuations of any particular quantity near the transition point reduces, as we shall see, to the clarification of the temperature dependence of $(d\xi_0/d\eta)_{\eta=\eta_0}$. For example, if we are interested in the fluctuations of the material density ρ, then we have from $\rho_0(\eta) = \rho_0(0) + \chi\eta^2$,

$$\left(\frac{d\rho_0}{d\eta}\right)_{\eta=\eta_0} = \begin{cases} 0 & \text{for } T > \Theta \text{ (symmetric phase)} \\ 2\chi\eta_0 & \text{for } T < \Theta \text{ (nonsymmetric phase)} \end{cases}$$

Hence for $T > \Theta$, the nearness of the transition point does not manifest itself in the value of the density fluctuations. So far as the case $T < \Theta$ is concerned, for a phase transition which coincides with the so-called critical point of second order phase transitions (CP), $\eta_0 \sim (\Theta - T)^{1/4}$. Therefore, $\langle(\Delta\rho)^2\rangle \sim (\Theta - T)^{1/2}$. At the same time, for phase transitions which are far from the CP, $\eta_0 \sim (\Theta - T)^{1/2}$, whence it follows that $\langle(\Delta\rho)^2\rangle$ does not increase as $T \to \Theta$.

2. The scattering of light which is accompanied by a small change in frequency, can be described as the result of spatially inhomogeneous fluctuations of the dielectric tensor of the medium ε_{ik}. The fluctuations of ε_{ik} are produced here by fluctuations of quantities which characterize the position of the atomic nuclei of the system. We have

$$\Delta\varepsilon = \varepsilon_\eta \Delta\eta + \sum_i \varepsilon_i \Delta\xi_i, \tag{8}$$

where

$$\varepsilon_\eta = (\partial\varepsilon/\partial\eta)_{\xi_i=\xi_{i0}(\eta_0),\ \eta=\eta_0},$$
$$\varepsilon_i = (\partial\varepsilon/\partial\xi_i)_{\xi_i=\xi_{i0}(\eta_0),\ \eta=\eta_0},$$

In the calculation of the intensity of anomalous scattering, one can set

$$\Delta\xi_i \approx \Delta\xi_{i0}(\eta) = (d\xi_{i0}/d\eta)_{\eta=\eta_0}\Delta\eta. \tag{9}$$

as was pointed out above. Substituting (9) in (8), we get

$$\Delta\varepsilon = \left[\varepsilon_\eta + \sum_i \varepsilon_i \left(\frac{d\xi_{i0}}{d\eta}\right)_{\eta=\eta_0}\right]\Delta\eta \equiv \tilde{\varepsilon}_\eta \Delta\eta. \tag{10}$$

In [2, 4-7], the terms under the summation sign in Eq. Eq. (10) were discarded in the calculation of $\langle(\Delta\varepsilon)^2\rangle$, as a result of which the coefficient ε_η appears in the final formula for the total intensity, and not $\tilde{\varepsilon}_\eta$. The error which arises in the use of the coefficient ε_η in place of $\tilde{\varepsilon}_\eta$ can be very substantial. In the case of the $\alpha \rightleftharpoons \beta$ transition in quartz (see, for example, [9]), the ratio $\varepsilon_\eta/\tilde{\varepsilon}_\eta \lesssim 0.2$, which upon conversion to intensity, means that of the total scattered light, only ~ 4% is scattered directly by fluctuations of the characteristic parameter; the remaining part is scattered by fluctuations of other quantities, principally by fluctuations of the density. The renormalized coefficient $\tilde{\varepsilon}_\eta$ is easily connected with experimentally observed quantities. For this, we note that for $T < \Theta$, in the state of complete equilibrium,[1)]

$$\varepsilon_{\text{eq}}(P, T) = \varepsilon(P, T, \xi_{i0}(\eta_0), \eta_0(T)),$$

$$\left(\frac{\partial\varepsilon_{\text{eq}}}{\partial T}\right)_P = \left(\frac{\partial\varepsilon}{\partial T}\right)_{P,\eta} + \left[\left(\frac{\partial\varepsilon}{\partial\eta}\right)_{\eta=\eta_0} + \sum_i \left(\frac{\partial\varepsilon}{\partial\xi_i}\right)_\eta \left(\frac{d\xi_{i0}}{d\eta}\right)_{\eta=\eta_0}\right]\left(\frac{d\eta_0}{dT}\right)_P$$
$$\equiv \left(\frac{\partial\varepsilon}{\partial T}\right)_{P,\eta} + \tilde{\varepsilon}_\eta \left(\frac{d\eta_0}{dT}\right)_P \approx \tilde{\varepsilon}_\eta \left(\frac{d\eta_0}{\partial T}\right)_P.$$

Similarly

$$\left(\frac{\partial^2\Phi_{\text{eq}}}{\partial T^2}\right)_P = \left(\frac{\partial^2\Phi}{\partial T^2}\right)_{P,\eta} - \left(\frac{\partial^2\Phi}{\partial\eta^2}\right)_{P,\eta=\eta_0}\left(\frac{\partial\eta_0}{\partial T}\right)_P^2 \equiv \left(\frac{\partial^2\Phi}{\partial T^2}\right)_{P,\eta} - \tilde{a}\left(\frac{\partial\eta_0}{\partial T}\right)_P^2$$

or

$$\tilde{a}\left(\frac{\partial\eta_0}{\partial T}\right)^2 = \frac{1}{T}\Delta C_P,$$

where ΔC_P is the difference in the heat capacity at constant pressure below and above the transition point. Now, using (7) and (10), we can easily obtain the result

$$\langle(\Delta\varepsilon)^2\rangle = \frac{k_B T^2}{V}\left(\frac{\partial\varepsilon_{\text{eq}}}{\partial T}\right)_P^2 \Big/ \Delta C_P. \tag{11}$$

We turn our attention to the fact that, inasmuch as ΔC_P is either a constant or (in the case of transitions close to the CP) increases as $(\Theta - T)^{-1/2}$; the anomaly in the light scattering is connected exclusively with the increase of the quantity $(\partial\varepsilon_{\text{eq}}/\partial T)_P$ close to the transition point, while two cases can be distinguished, depending on the character of the symmetry of the material and the transformation properties of the characteristic parameter:

1) $\varepsilon_{\text{eq}}(P, T) - \varepsilon_{\text{eq}}(P, T > \Theta) \sim \eta_0^2(P, T)$ for $T < \Theta$;
2) $\varepsilon_{\text{eq}}(P, T) - \varepsilon_{\text{eq}}(P, T > \Theta) \sim \eta_0(P, T)$ for $T < \Theta$.

In the first case, the derivative $(\partial\varepsilon_{\text{eq}}/\partial T)_P$ increases as $T \to \Theta$ only for transitions close to the CP, in which connection the anomaly in the light scattering is observed only for such transitions (the temperature dependence of η_0 is treated at the end of the previous section).

In the second case, $(\partial\varepsilon_{\text{eq}}/\partial T)_P$ increases for transitions both close to and far away from the CP. Furthermore, in this case, in contrast with the previous one, the scattering anomaly also takes place in the symmetric phase (see [4]).

3. Let us consider the question of the spectral composition of the scattered light. Changes in the spectral picture of the scattered light near the phase transition point are obviously connected with the change in the time characteristic of the fluctuations of the various quantities. The evolution of the spectrum of fluctuations of the characteristic parameter of the transition was considered in [6] without account of the coupling of η with other quantities. It was shown in this case that, because of the fact that the system loses rigidity at the

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[1)] The better agreement with experiment, [3] obtained in [5] in the estimate of the total intensity of scattered light near the $\alpha \rightleftharpoons \beta$ transition point in quartz, is explained by the fact that in the estimate the quantity $(\partial\varepsilon_{\text{eg}}/\partial T)_P/(\partial\eta_0/\partial T)_P$ is used as the coefficient ε_η in the formula for the total intensity. This quantity represents none other than the coefficient $\tilde{\varepsilon}_\eta$ whose value for quartz exceeds the value of the coefficient ε_η by order of magnitude.

phase transition point relative to changes in η, the fluctuations of η, even if they are far from the transition point and have an oscillatory character, are strongly retarded near the transition point, so that, beginning with some temperature (generally speaking, not very close to Θ), the oscillations give way to a relaxation, while the relaxation time increases as $|T - \Theta|^{-1}$ as $T \rightarrow \Theta$. Thus the fluctuations near the transition point are fundamentally very slow fluctuations. Correspondingly, in the spectrum of the light scattered by the fluctuations of η, there is an unshifted component near the transition point whose width decreases as $T \rightarrow \Theta$.

Proceeding to the study of the fluctuations of quantities other than the characteristic parameter, we divide the quantities whose fluctuations are connected with the fluctuations of η into two groups. To the first group, we assign those quantities ξ which cannot be linearly connected with η; that is, those quantities (for example, the density), for which only a quadratic connection is possible: $\xi \sim \eta^2$. In the second group, we place those quantities which are linearly related to η.

We begin with the quantities of the first group. The thermodynamic potential has the following form in this case:

$$\Phi(\xi, \eta) = \Phi_0 + \alpha\eta^2 + \frac{\beta}{2}\eta^4 + \frac{\gamma}{6}\eta^6 + \frac{\chi}{2}\xi^2 + d\eta^2\xi. \qquad (12)$$

The coefficient $b = 2d\eta_0$, which is equal to zero for $T > \Theta$, corresponds to the potential in the form (12) in Eq. (3). Thus, the fluctuations of ξ and η are independent for $T > \Theta$.

For $T < \Theta$, the fluctuations of ξ, as is seen from Eqs. (2)–(5), can be split into two parts: the fluctuations of ξ' about the state of partial equilibrium, corresponding to a given instantaneous value of η, and the fluctuations $\xi_0(\eta)$, for which ξ changes along with η. Since η changes very slowly in the vicinity of the transition point, the fluctuations of ξ' in this case proceed at almost constant η, and the presence of the phase transition has little effect on them. Account of the same slow fluctuations $\xi_0(\eta)$, which take place in synchronism with the fluctuations of η, leads only to a renormalization of the coefficient of proportionality between $\Delta\varepsilon$ and $\Delta\eta$, just as in the calculation of the total scattering intensity. Thus, if the fluctuations of ξ bear an oscillatory character, and are accompanied by a change in ε_{ik}, then a combination line is observed in the spectrum of scattered light near the transition point. The frequency of this line is determined at $T = \Theta$ by the value of c in Eq. (3), which has the meaning of an elastic modulus, corresponding to a change in ξ at constant η.

As one moves away from the transition point, the relaxation time of the characteristic parameter

$$\tau \sim a^{-1} = {}^1\!/_4\eta_0^2(\sqrt{\beta^2 - 2\alpha\gamma} + d^2/\chi)$$

will decrease; sufficiently far from the transition point, it can be seen to be less than the period of oscillation of ξ. Then the frequency of the fluctuations of ξ will be determined not by the quantity c, but by $c_{st} = c - b^2/a$, which has the meaning of a "static" elastic modulus, which takes into account the change in ξ for change in η. Consequently, for $T < \Theta$, the location of sufficiently low-frequency lines can depend significantly on the temperature. The most low-lying combination lines are, as is well known, the Mandel'shtam-Brillouin (MB) components. The indicated temperature dependence for them has been observed experimentally in triglycin sulfate.[10]

However, it can be shown that even far away from the transition point, the relaxation time of the characteristic parameter is greater than the period of the hypersonic oscillations corresponding to the MB components. In this case, the presence of a phase transition has no effect on the temperature dependence of the position of these components and the higher-lying combination lines.

We go on to the consideration of fluctuations of quantities of the second group, i.e., those which in the symmetric phase possess a linear coupling between ξ and η. The thermodynamic potential in this case has the form

$$\Phi = \Phi_0 + \alpha\eta^2 + \frac{\beta}{2}\eta^4 + \frac{\gamma}{6}\eta^6 + d\xi\eta + \frac{\chi}{2}\xi^2. \qquad (13)$$

We shall consider the transition for zero force connected with ξ. Here, as it is not difficult to establish, the temperature of the phase transition is determined not from the condition $\xi = 0$, as was the case when the transition took place at $\alpha = 0$, but from the condition of vanishing of the determinant $ac - b^2$, which consists of the second derivatives entering into (3). These derivatives are expressed in terms of the expansion coefficients in the following form:

$$a = \begin{cases} 4\eta_0^2(\sqrt{\beta^2 - 2\alpha\gamma} + d^2/\chi), & T < \Theta \\ 2\alpha, & T > \Theta \end{cases}$$
$$b = d, \qquad c = \chi.$$

For $T = \Theta$, consequently, the quantities $(\partial^2\Phi/\partial\eta^2)_{F_\xi} = \tilde{a} = a - d^2/\chi$ (F_ξ is the force conjugate to ξ) and $(\partial^2\Phi/\partial\xi^2)_{F_\eta} = \chi - d^2/a$ (F_η is the force conjugate to η) vanish; the quantity $(\partial^2\Phi/\partial\eta^2)_\xi$ remains finite at the transition point. Inasmuch as the relaxation time of the characteristic parameter $\tau \sim (\partial^2\Phi/\partial\eta^2)^{-1}$, then for $T \rightarrow \Theta$, τ—the relaxation time of η for $F_\xi = 0$—increases without bound, the relaxation time of η for fixed ξ (in what follows, this is denoted simply by τ) remains finite at the transition point.

In studying the fluctuations of ξ, it is generally necessary to compare the period of the fluctuations with the two relaxation times. It is obvious that if this period is shown to be much less than τ everywhere in space (and thereby greater than $\tilde{\tau}$), then, just as was done above, the fluctuations of ξ can be divided into slow (ξ_0) and fast (ξ'), and the presence of a phase transition has no effect on the fast fluctuations, which take place at constant η. We shall consider, by means of a specific example, what possibilities exist in the case of quantities the frequency of whose fluctuations is comparatively small.

We shall investigate the low-frequency portion of the spectrum of light scattered in a uniaxial ferroelectric, which lacks a center of inversion in the para phase (Rochelle salt, KDP). Near the transition point, the slowest fluctuations for such a material will be the fluctuations of the vector of spontaneous polarization **P** and the components of the elastic deformation tensor u_{ik}. We choose the direction of spontaneous polarization along the z axis. In considering the fluctuations of P, we can assume that $P_x = P_y = 0$. This is an excellent approximation, inasmuch as, for T close to Θ, $\varepsilon_{zz} \gg \varepsilon_{xx}, \varepsilon_{yy}$. Among the quantities u_{ik} for the materials considered, only the deformation u_{xy} changes the value of P_z; therefore, it suffices to consider only its fluctuations. The thermodynamic potential as a function of P_z and u_{xy} has the form

$$\Phi = \Phi_0 + \alpha P_z^2 + \frac{\beta}{2} P_z^4 + \frac{\gamma}{6} P_z^6 + dP_z u_{xy} + \frac{\mu}{2} u_{xy}^2 - \frac{E_z P_z}{2}. \tag{14}$$

Here μ is the shear modulus, d the piezoelectric modulus, and E_z the electric field produced by the inhomogeneously polarized crystal.[11] For waves propagating perpendicular to the z axis (in what follows, we shall consider only such waves), the electric field can be assumed to be zero.

We proceed to the writing of the equations of motion. In materials about which we have been speaking, the appearance of P_z is associated with a certain ordering process (evidently of a relaxational character). It is natural therefore to assume that the term in the equation of motion for P_z with the second time derivative can be omitted. We have simply

$$\gamma \dot{P}_z = \partial\Phi / \partial P_z, \tag{15}$$

where γ is some constant which can be considered to be independent of the temperature in the vicinity of the transition point. We have the standard equation of motion for u_{xy} [2)]

$$\rho \ddot{u}_{xy} + \mu\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right) u_{xy} + d\left(\frac{\partial}{\partial x^2} + \frac{\partial}{\partial y^2}\right) P_z = 0. \tag{16}$$

From (15) and (16) it is easy to obtain expressions for

$$\langle |P_z(\omega, \mathbf{k})|^2\rangle, \quad \langle P_z^*(\omega, \mathbf{k}) u_{xy}(\omega, \mathbf{k})\rangle, \quad \langle |u_{xy}(\omega, \mathbf{k})|^2\rangle,$$

by carrying out calculations in the usual manner.[8] Here $P_z(\omega, \mathbf{k})$ and $u_{xy}(\omega, \mathbf{k})$ are the space-time Fourier components of the functions $P_z(\mathbf{r}, t)$ and $u_{xy}(\mathbf{r}, t)$. Knowing these quantities and the character of the dependence of the tensor ε_{ik} on P_z and u_{xy} we obtain a formula for the spectral intensity of the light $I(\omega)$ scattered at a given angle.

In the case of KDP and Rochelle salt, only a single component of the tensor ε_{ik}, namely ε_{xy}, depends on u_{xy} and P_z, and the dependence of ε_{xy} on u_{xy} is much more important than on P_z (we recall that the intensity of the MB components exceeds the intensity of the other combination components by a factor 10^2). Keeping this in view, we set

$$\varepsilon_{xy} \cong g u_{xy}. \tag{17}$$

Correspondingly, we have for $I(\omega)$

$$I(\omega = \Omega - \Omega_0) \sim \langle |u_{xy}(\omega, \mathbf{k})|^2\rangle = \frac{k_B T}{\pi\omega V} \frac{\mu''}{(\mu' - \rho\omega^2/k^2)^2 + \mu''^2} \tag{18}$$

where μ' and μ'' are the real and imaginary parts of the complex shear modulus

$$\tilde{\mu}(\omega) = \mu - d^2 / (a - i\omega\gamma), \tag{19}$$

and the value of k is given by the angle of the light scattering and is equal to the difference of the wave vectors of the scattered and incident light. According to (18), the location of the secondary maxima of $I(\omega)$ (corresponding to the MB components) is determined by the relation

$$\omega_{max}^2 = {}^1/_3\, \omega_0^2 \{2 - z^2 + [(2 - z^2)^2 + 3(2z^2 - 2zz_0 - 1)]^{1/2}\}, \tag{20}$$

where we have introduced the notation

$$\omega_0 = \sqrt{\rho / \mu k^2}, \quad z = a(T) / \omega_0\gamma, \quad z_0 = z(\Theta).$$

Analysis of Eq. (20) leads to the following results:

1. Far from the transition point, the maximum of $I(\omega)$ for $\omega = 0$, which corresponds to the unshifted component Ω_0, is lacking. It appears for the temperature T_1, defined from the relation

$$z(T_1) = {}^1/_2 (z_0 + \sqrt{z_0^2 + 2}).$$

2. If $z_0 > 3/2\sqrt{2}$, then the appearance of the central maximum is accompanied by the disappearance of the secondary maxima. Thus the MB components for $T \rightarrow \Theta$ approach one another and at a temperature T_1 merge into the unshifted line.

3. If $(2 - \sqrt{3})^{1/2} < z_0 < 3/2\sqrt{2}$, then the central maximum appears before the MB components are merged. The temperature T_2 of merging of the MB components is determined by the condition of the vanishing of the root of the expression in (20).

4. If $z_0 < (2 - \sqrt{3})^{1/2}$, then the MB components are present even for $T = \Theta$. Their location can be determined by setting $z = z_0$ in (20). For $1/2 < z_0 < (2 - \sqrt{3})^{1/2}$, the distance between the MB components decreases as we approach the transition point, becoming minimal for $T = \Theta$. For $z_0 = 1/2$, the MB components come together only up to $T_3 \neq \Theta$, after which the distance between the MB components begins to increase for further approach of T to Θ.

The shifts of the MB components, which are not accompanied by their merging in the vicinity of the transition point, take place in crystals of Rochelle salt. Here[3, 12] $\alpha(\Theta) \approx 1.6 \times 10^{-2}$, $\gamma \sim 10^{-11} - 10^{-10}$ sec^{-1}, the sound velocity far from the transition point $c_0 = \omega_0/k \approx 2.5 \times 10^5$ cm/sec, while k is about 10^5 cm^{-1} for irradiation of the crystal by visible light. Then for the quantity $z_0 = a(\Theta)/\gamma\omega$, we obtain the numerical value $\sim 10^{-1} - 10^{-2}$, corresponding to the case $z_0 < 1/2$.

For KDP crystals, $\tau^{-1}(\Theta) > 10^{10}$,[12] so that the case $z_0 > 3/2\sqrt{3}$ is clearly realizable, i.e., the MB components merge into the unshifted Rayleigh line for $T \rightarrow \Theta$.

We emphasize that in any comparison of the results obtained above with experiment, one must not forget their approximate character. The presence of finite friction in the acoustic branch, as well as the account in Eq. (17) of the team proportional to the fluctuations of P_x, can bring some change into the picture of the distribution of scattered light over the frequencies.

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2) In Eq. (16), the term containing friction is omitted. Account of it has only a slight effect on the results.

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