Inelastic scattering of light by fluctuations of the order parameter in nematic liquid crystals

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The line shape of the scattering due to singular longitudinal and biaxial fluctuations of the order parameter in nematic liquid crystals is calculated. Qualitative effects by which the character of the fluctuations can be determined from the line shape are indicated.

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I. INTRODUCTION

Scattering of light in liquid crystals, particularly near the phase-transition point, has been recently investigated intensively both theoretically¹ and experimental $ly.^2$ As a rule, when it comes to the nematic phase, only light scattering due to fluctuations of the director is considered (with the exception of Ref. 3, which will be discussed later). This is generally speaking justified, since the main cause of the anomalously strong scattering in liquid crystals is the fluctuation of the orientation of the director. The reason is that nematic liquid crystals are degenerate systems (or systems with broken continuous symmetry) and therefore uniform transverse fluctuations (i. e., uniform rotations of the director in all of space) need not overcome a barrier to become excited (i. e., they are Goldstone fluctuations).

It is known, however,¹⁻³ that the transverse fluctuations do not lead to light scattering if the polarization satisfies certain conditions. For example, there is no scattering if the vectors of the initial and final polarizations are in the equatorial plane, i. e., in the plane perpendicular to the director. More accurately speaking, the transverse scattering is weak in a small angle interval such that the polarization is close to the equatorial plane. The second simplest case in which the transverse fluctuations do not lead to light scattering occurs when the wave vectors of the incident and scattered light are in the equatorial plane, and the polarizations are parallel to the director. In this region the light scattering is determined by fluctuations of another type, namely longitudinal and biaxial fluctuations.

We define as longitudinal the fluctuations of the modulus of the order parameter. Biaxial fluctuations describe the deviations of the order parameter from the uniaxial tensor defined by the director. Pokrovskii and one of us (Kats)⁴ calculated the integrated (with respect to frequency) intensity of the light scattering connected with the longitudinal and biaxial fluctuations. It is experimentally more convenient, however, to measure not the integrated but the spectral intensity, i. e., the line shape. This is due both to the higher accuracy of such a procedure and to the fact that in the spectral intensity it is easier to separate effects that stem from impurities and other defects. To calculate the spectral intensity it is necessary to investigate the dynamics of the fluctuations of the order parameter. The present paper is devoted to this investigation.

Stratonovich³ has obtained certain results pertaining to the same question. We call attention out to the difference between our study and his. As to the longitudinal fluctuations, Stratonovich³ considered only the socalled classical fluctuations of the modulus s of the order parameter Q_{ij} , i.e., he determined the fluctuations $\langle \delta s^2 \rangle$ directly from the expansion of the Landau free energy. At $T < T_c$ (T_c is the temperature of the transition into an isotropic liquid) these fluctuations are not singular when the fluctuation wave vector $q \rightarrow 0$. These fluctuations are present, generally speaking, in any system that undergoes a phase transition, and one can say that they manifest no liquid-crystal properties whatever. We, on the other hand, consider singular longitudinal scattering, which is typical only of such a degenerate system as a nematic liquid crystal. Such singular longitudinal fluctuations are connected with the presence of Goldstone transverse fluctuations (in this case, the fluctuations of the director). As shown in Ref. 4, the singular longitudinal fluctuations predominate at wavelengths $\lambda \ge 5000$ Å. In Ref. 3 there was considered also the dynamics of biaxial fluctuations. The intensity of light scattering is determined by the fluctuations of the anisotropic part of the dielectric constant ε_{ii} , which is connected with the fluctuations of the order-parameter tensor Q_{ii} . Stratonovich,³ however, does not take into account the interaction of the fluctuations Q_{ij} and of the velocity \mathbf{v} of the liquid crystal. It is known, however,^{1,2} that the time dependence of the fluctuations of Q_{ij} (and consequently also the spectral intensity of the light scattering) can be strongly influenced by the hydrodynamic motion. This is the cause, for example, of the birefringence in the stream. In the present paper the cross sections for the singular longitudinal and biaxial scattering are calculated with account taken of the hydrodynamic interaction.

II. LONGITUDINAL SCATTERING OF LIGHT

The order parameter of the nematic liquid crystal Q_{ii} can be written in the following most general form:

$$Q_{ij} = s(n_i n_j - \frac{1}{3} \delta_{ij}) + \delta Q_{ij}^{\perp}, \tag{1}$$

where the transverse fluctuation δQ_{ij}^{\perp} satisfies the orthogonality conditions

$$n_i n_j \delta Q_{ij} = 0, \quad \delta Q_{ii} = 0.$$

The conditions (2) can be satisfied in general form by introducing the new variables⁴

$$\delta Q_{i_{j}}^{\perp} = \xi_{1}(n_{i}e_{1j} + n_{j}e_{1i}) + \xi_{2}(n_{i}e_{2j} + n_{j}e_{2i}) + \xi_{3}(e_{1i}e_{2j} + e_{1j}e_{2i}) + \xi_{4}(e_{1i}e_{1j} - e_{2i}e_{2j}),$$
(3)

where the director **n** and the unit vectors \mathbf{e}_1 and \mathbf{e}_2 make up a right-hand triad of unit vectors. The parameters ξ_1 and ξ_2 describe the fluctuation of the director, while ξ_3 and ξ_4 describe the biaxial fluctuation.

As already indicated in the Introduction, strong transverse fluctuations lead in degenerate systems to weaker but likewise singular longitudinal fluctuations. The corresponding connection is expressed by the so-called modulus-conservation principle:

$$2s\delta s = -(\delta Q_{ij}^{\perp})^2. \tag{4}$$

This local equality takes place in coordinate space, while in momentum space it is necessary to calculate the integral of the transverse correlators; this can be symbolically represented in the form of a loop diagram (see the figure). The lines of the diagram correspond to the correlators of the fluctuations of the director¹⁾ $\xi_{1,2}$

$$I_{\alpha}(\mathbf{q},\omega) = \int d\mathbf{r} \int dt \exp(i\mathbf{q}\mathbf{r} - i\omega t) \langle \xi_{\alpha}(\mathbf{r},t) \xi_{\alpha}(0,0) \rangle, \qquad (5)$$

where $\alpha = 1, 2$. The transverse correlator is calculated from the linearized equations of hydrodynamics and is well known¹:

$$I_{\alpha}(\mathbf{q},\omega) = \frac{2I_{\alpha}(\mathbf{q})\Gamma_{\alpha}(\mathbf{q})}{\omega^{2} + \Gamma_{\alpha}^{2}(\mathbf{q})},$$
(6)

where

$$V_{\alpha}(\mathbf{q}) = \frac{T}{K_{\alpha\alpha}q_{x}^{2} + K_{33}q_{z}^{2}}, \quad \Gamma_{\alpha} = \frac{A_{\alpha}P_{\alpha}}{\gamma_{1}P_{\alpha} - C_{\alpha}B_{\alpha}},$$
(7)

and A_{α} , C_{α} , P_{α} , and B_{α} are equal to:

$$A_{1} = K_{11}q_{x}^{2} + K_{33}q_{z}^{2}, \quad C_{1} = -\frac{1}{2q_{x}} [\gamma_{1}q^{2} + \gamma_{2}(q_{x}^{2} - q_{z}^{2})],$$

$$P_{1} = \frac{1}{2q^{2}} (2\eta_{1}q_{x}^{4} + 2\eta_{2}q_{z}^{4} + \alpha_{m}q_{x}^{2}q_{z}^{2}), \quad B_{1} = \frac{q_{z}}{q^{2}} (\alpha_{3}q_{x}^{2} - \alpha_{2}q_{z}^{2}),$$

$$A_{2} = K_{22}q_{x}^{2} + K_{33}q_{z}^{2}, \quad C_{2} = \frac{1}{2} (\gamma_{2} - \gamma_{1})q_{z}, \quad (8)$$

$$P_{2} = \frac{1}{2} (\alpha_{4}q_{x}^{2} + 2\eta_{2}q_{z}^{2}), \quad B_{2} = \alpha_{2}q_{z}.$$

Here α_i are the standard symbols for the Leslie coefficients, while η_1 , η_2 , and α_m denote certain combinations of α_i :

 $\eta_1 = \frac{1}{2} (\alpha_3 + \alpha_4 + \alpha_6), \qquad \eta_2 = \frac{1}{2} (\alpha_4 + \alpha_5 - \alpha_2),$ $\alpha_m = 2 (\alpha_1 + \alpha_4) + \alpha_5 + \alpha_5 + \alpha_5 - \alpha_2.$

The coefficients γ_1 and γ_2 describe the rotation friction:

 $\gamma_1 = \alpha_3 - \alpha_2 = \alpha_3 - \alpha_6, \quad \gamma_2 = \alpha_3 + \alpha_2.$

Unfortunately, the longitudinal fluctuations can be determined from (6)-(8) only numerically. At a sufficient degree of accuracy for the establishment of qualitative relations, however, one can use the following approximation. In practically all nematic liquid crystals the parameters α_1 and α_3 are small, while the remaining $\alpha_2-\alpha_6$ are larger by at least one order of

magnitude. Thus, for example, in the MBBA crystal at $T_c - T = 10$ K we have $\alpha_1 = 6.5$, $\alpha_3 = -1.2$, $\alpha_2 = -77.5$, $\alpha_4 = 83.2$, $\alpha_5 = 46.3$, $\alpha_6 = -34.4$. The signs of the "large" Leslie viscosity coefficients are also fixed. In this approximation (whose accuracy is ~1%) $C_{\alpha}B_{\alpha} \ll \gamma_1P_{\alpha}$. We therefore have from (7)

$$\Gamma_{\alpha} = A_{\alpha} / \gamma_{1}. \tag{9}$$

With the same accuracy (~1%), however, we can neglect the anisotropic elastic moduli. Thus, ultimately,

$$\Gamma = \frac{K}{\gamma_1} q^2, \quad I(\mathbf{q}) = \frac{T}{Kq^2}, \quad I(\mathbf{q}, \omega) = \frac{2T}{\gamma_1} \frac{1}{\omega^2 + (K^2/\gamma_1^2) q^4}.$$
 (10)

It is more convenient to carry out the calculations in coordinate space:

$$I(\mathbf{r},t) = \frac{2T}{\gamma_1} \frac{1}{(2\pi)^4} \int d\mathbf{q} \, d\omega \exp(i\omega t - i\mathbf{q}\mathbf{r}) \frac{1}{\omega^2 + (K^2/\gamma_1^2)q^4}.$$

The integral with respect to frequency can be easily obtained:

$$I(\mathbf{r},t) = \frac{2T}{\gamma_1} \frac{1}{8\pi^3} \int \frac{d\mathbf{q}}{q^2} \exp\left(-i\mathbf{q}\mathbf{r} - \frac{K}{\gamma_1} q^2 t\right)$$

We next calculate the integral over the angles:

$$I(\mathbf{r},t) = \frac{T}{\gamma_i} \frac{1}{2\pi^2} \frac{1}{r} \int_0^{\infty} \frac{dq}{q} \sin(qr) \exp\left(-\frac{K}{\gamma_i} q^2 t\right).$$
(11)

The integral in (11) is well known (see, e.g., Ref. 5):

$$I(\mathbf{r},t) = \frac{T}{4\pi K} \frac{1}{r} \Phi\left(r/2\left(\frac{K}{\gamma_{i}}t\right)^{\gamma_{i}}\right), \qquad (12)$$

where

$$\Phi(x) = \frac{2}{\sqrt{\pi}} \int \exp(-u^2) du.$$

The longitudinal fluctuation is

$$\delta Q_{ij}^{\parallel} = \delta s(n_i n_j - \frac{1}{3} \delta_{ij}).$$
(13)

From the modulus conservation principle (4) we have

$$G(\mathbf{r}, t) = \langle \delta s(r, t) \delta s(0, 0) \rangle = I^2(\mathbf{r}, t)/4s^2,$$
(14)

from which we get

$$G(\mathbf{r},t) = \frac{T^2}{64\pi^2 K^2 s^2} \frac{1}{r^2} \Phi^2 \left(r/2 \left(\frac{K}{\gamma_1} t \right)^{1/2} \right).$$
(15)

Equation (15) solves in principle the problem of the intensity of light scattering by longitudinal fluctuations. However, a direct Fourier transformation of (15) is quite difficult to obtain.

It is more convenient to use first a partial Fourier transform only with respect to the coordinate. We again have from (4)

$$G(\mathbf{q},t) = \frac{1}{4s^2} \int I\left(\mathbf{p} + \frac{\mathbf{q}}{2}, t\right) I\left(\mathbf{p} - \frac{\mathbf{q}}{2}, t\right) d^3p.$$
(16)

In turn we obtain from (10) an expression for I(p, t):

$$I(\mathbf{p},t) = \frac{T}{K} \frac{1}{p^2} \exp\left(-\frac{K}{\gamma_1} p^2 t\right).$$
 (17)

Substituting (17) in (16) and calculating the integral with respect to the angles, we obtain

$$G(\mathbf{q},t) = \frac{\pi}{8s^2} \frac{T^2}{K^2} \exp\left(-\frac{K}{\gamma_1} t \frac{q^2}{2}\right) \frac{1}{q} \cdot \int_{0}^{\infty} \frac{p \, dp}{\Delta} \exp\left(-2\frac{K}{\gamma_1} p^2 t\right) \ln\left|\frac{\Delta + pq}{\Delta - pq}\right|,$$
(18)

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where $\Delta = p^2 + q^2/4$.

It is seen from (18) that the main contribution to the integral comes from the region $q \ll p$. Expanding the integrand in this region, we obtain accurate to $(q/p)^3$

$$G(\mathbf{q},t) = \frac{\pi^2}{8s^2} \frac{T^2}{K^2} \frac{1}{q} \left[1 - \Phi\left(\frac{K}{\gamma_1} \frac{qt^{\gamma_1}}{2^{\gamma_1}}\right) \right].$$
(19)

Formula (19) can be easily Fourier-transformed with respect to time

$$G(\mathbf{q},\omega) = \frac{\pi^2 T^2}{8s^2 K^2} \left[\omega^2 + \frac{K^2}{4\gamma_1^2} q^4 \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K^2}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1} q^2 \right]^{-1/2} \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K^2}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1} q^2 \right]^{-1/2} \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K^2}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1} q^2 \right]^{-1/2} \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K^2}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1} q^2 \right]^{-1/2} \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K^2}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1} q^2 \right]^{-1/2} \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K^2}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1} q^2 \right]^{-1/2} \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K^2}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^2 \right]^{-1/2} \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K^2}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^2 \right]^{-1/2} \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^2 \right]^{-1/2} \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^2 \right]^{-1/2} \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^2 \right]^{-1/2} \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^2 \right]^{-1/2} \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^2 \right]^{-1/2} \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^2 \right]^{-1/2} \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^4 \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^4 \right]^{-1/2} \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^4 \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^4 \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^4 \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^4 \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^4 \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^4 \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^4 \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^4 \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{4\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^4 \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{2\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^4 \right]^{-1/2} \left[\left\{ \omega^2 + \frac{K}{2\gamma_1^2} q^4 \right\}^{1/2} + \frac{K}{2\gamma_1^2} q^4 \right]^{-1/2} \left[\left\{ \omega^2$$

The principal consequence of (20) is that the line shape is not Lorentzian. At $\omega \gg q$ we have $G(\mathbf{q}, \omega) \sim 1/\omega^{3/2}$ and at $q \gg \omega$ we have $G(\mathbf{q}, \omega) \sim 1/q^3$.

The light-scattering cross section can be easily connected with the quantity $G(\mathbf{q}, \omega)$. The differential cross section in the frequency interval $d\omega$ and in the solid angle interval $d\Omega$ is

$$\frac{d\sigma}{d\omega d\Omega} = \frac{\omega^*}{32\pi^3} \langle \delta \varepsilon_{ij} \delta \varepsilon_{mn} \rangle p_i p_m p_j' p_n', \qquad (21)$$

where $\delta \omega_{ij}$ is the fluctuation of the dielectric tensor, **p** and **p'** are the polarization vectors of the incident and scattered light (we use a system of units in which the speed of light c=1). By virtue of the symmetry, $\delta \varepsilon_{ij}$ is connected with the fluctuation of the order parameter $\delta Q_{ij}^{"}$ by the linear relation

$$\delta \varepsilon_{ij} = M \delta Q_{ij}^{\parallel}. \tag{22}$$

From (22), (21), and (13) we have

$$\frac{d\sigma}{d\omega \, d\Omega} = \frac{\omega^4 M^2}{32\pi^3} \left[(\mathbf{pn}) (\mathbf{p'n}) - \frac{1}{3} (\mathbf{pp'}) \right]^2 G(\mathbf{q}, \omega).$$
(23)

III. BIAXIAL SCATTERING OF LIGHT

To calculate the spectral intensity connected with the biaxial fluctuations it is necessary to know the dynamics of the fluctuations. The system of the basic equations of motion is conveniently written in the form of the Langrange variational equations obtained from the least-action principle.⁶ To this end it suffices to specify the density of the Lagrangian. In the case of interest to us, of a medium in thermodynamic equilibrium, the role of the Lagrangian is assumed by the difference between the kinetic energy E and the free energy F:

$$\mathcal{L}=E-F.$$

By using the free energy F, we aim by the same token to confine ourselves beforehand to processes at constant volume and temperature. To take into account the relaxation processes it is necessary also to specify the density of the dissipation function R. As the independent variables that characterize the state of the liquid crystal at the point \mathbf{r} and at the instant of time twe choose the displacement vector $\mathbf{u}(\mathbf{r}, t)$ [the velocity of the liquid crystal is $\mathbf{v}(\mathbf{r}, t) = \dot{\mathbf{u}}(\mathbf{r}, t)$] and the order parameter $Q_{ij}(\mathbf{r}, t)$. As usual, we assume that the liquid crystal is not compressible.

(25)

The kinetic-energy density takes the form $E = \rho u^2/2$,

where $\rho = \text{const}$ is the density of the medium.

The expression for the free energy in terms of the variables ξ_i was obtained in Ref. 4:

$$F = K_1 q^2 \sum_{i=1}^{\infty} |\xi_i|^2 + \frac{1}{2} K_2 q^2 [(\xi_1 \cos \theta + \xi_3 \sin \theta)^2 + \xi_2^2 + \xi_4^2 \sin^2 \theta - \xi_2 \xi_1 \sin 2\theta] + \frac{1}{2} \Delta (\xi_3^2 + \xi_4^2),$$
(26)

where K_1 and K_2 are certain combinations of the Frank moduli. $(k_1 = k \text{ and } k_2 = 0 \text{ in the case } K_{11} = K_{22} = K_{33} = K);$ θ is the angle between the vectors n and q, and Δ characterizes the energy needed to excite the biaxial fluctuation. In the Landau theory

$$\Delta = \frac{20}{9}B^2/C,$$

where B and C are the coefficients of Q_{ij}^3 and Q_{ij}^4 , respectively.

For our purposes, the linearized hydrodynamics equations are sufficient. It is therefore enough to take into account in the density of the dissipation function the quadratic dependence on the velocity gradients $\partial v_i / \partial x_j$ and on the rate of "rotation" of the order parameter relative to the liquid crystal:

 $\dot{Q}_{ij} - Q_{ij} \times \frac{1}{2}$ rot v.

It is convenient to introduce the corresponding symmetrical tensors

$$A_{ij} = \frac{1}{2} (\partial v_i / \partial x_j + \partial v_j / \partial x_i),$$

$$N_{ij} = \dot{Q}_{ij} - (e_{inm}Q_{jm} + e_{jnm}Q_{im}) \omega_n,$$
(27)

where $\omega = (1/2)$ curl **v** and e_{inm} is a fully antisymmetrical tensor.

The function R is constructed from Q_{ij} and from expressions quadratic in A_{ij} and N_{ij} . At the required accuracy, with allowance for symmetry considerations and orthogonality (2), we can write

$$R = \mu_{1}(A_{ij})^{2} + \mu_{2}(N_{ij})^{2} + \mu_{3}N_{ij}A_{ij} + \mu_{4}Q_{ij}A_{ji}A_{ii} + \mu_{3}Q_{ij}N_{ji}N_{ii} + \mu_{3}Q_{ij}A_{ii}N_{ji} + \mu_{7}(Q_{ij}A_{ij})^{2} + \mu_{8}(Q_{ij}N_{ij})^{2} + \mu_{9}(Q_{ij}N_{ij})(Q_{ij}A_{ij}).$$
(28)

Some of the viscosity coefficients μ_i can be expressed in terms of the Leslie coefficients, while three coefficients represent new "biaxial" viscosities.

The general form of the variational equations of motion with a certain set of variables $m_i(\mathbf{r}, t)$ is given by the known expression

$$\frac{\partial}{\partial t} \left(\frac{\delta \mathscr{L}}{\delta \dot{m}_i} \right) - \frac{\delta \mathscr{L}}{\delta m_i} + \frac{\partial}{\partial x_{\mathfrak{s}}} \frac{\partial \mathscr{L}}{\delta (\partial m_i / \partial x_{\mathfrak{s}})} = - \frac{\delta R}{\delta m_i} + \frac{\partial}{\partial x_{\mathfrak{s}}} \frac{\delta R}{\delta (\partial m_i / \partial x_{\mathfrak{s}})}.$$
 (29)

The employed independent variables m_i are the three components of the displacement vector **u** and the four parameters ξ_i that characterize the fluctuation of $Q_{\alpha\beta}$.

The spectrum of the natural oscillations (and consequently the scattering line shape) is next determined from formulas (25)-(29). The problem reduces to a system of algebraic equations or to the calculation of a 7×7 determinant of rather general form. For example,

$$N_{ij}^{2} = Q_{ij}^{2} + i\xi_{1} [(\mathbf{e},\mathbf{q})(\mathbf{n}\mathbf{v}) - (\mathbf{e},\mathbf{v})(\mathbf{n}\mathbf{q})] + i\xi_{2} [(\mathbf{e}_{2}\mathbf{q})(\mathbf{n}\mathbf{v}) - (\mathbf{e}_{2}\mathbf{v})(\mathbf{n}\mathbf{q})] + 2[(\mathbf{n}\mathbf{v})^{2}\mathbf{q}^{2} + (\mathbf{n}\mathbf{q})^{2}\mathbf{v}^{2} - 2(\mathbf{n}\mathbf{v})(\mathbf{q}\mathbf{v})(\mathbf{n}\mathbf{q})], N_{ij}A_{ij} = iQ_{ij}(q_{i}v_{j} + q_{j}v_{i}) - [(\mathbf{n}\mathbf{q})^{2}\mathbf{v}^{2} - (\mathbf{n}\mathbf{v})^{2}\mathbf{q}^{2}]$$

etc., i.e., most elements of the determinant are different from zero. Of course, such a calculation cannot be carried out in analytic form.

However, in our case of light scattering in the optical band there is one simplifying circumstance. The point is that at typical values of the parameters the quantity Δ in (26) is very large compared with Kq^2 :

$$\Delta \sim 4 \times 10^7 \text{ erg/cm}^3$$
, $Kq^2 \sim 10^4 \text{ erg/cm}^3$ ($q \sim 10^6 \text{ cm}^{-1}$).

Therefore we can put q=0 in all the equations of motion for the parameters ξ_3 and ξ_4 . This cannot be done in the equations for ξ_1 and ξ_2 , which do not contain the large parameter Δ . We thus obtain the following system of equations.

$$(-i\omega\gamma_{1}+A_{1})\xi_{1}+iC_{1}c_{2}+K_{2}q^{2}\cos\theta\sin\theta\xi_{3}=0,$$

$$(-i\omega\rho+P_{1})c_{3}-\omega B_{1}\xi_{1}=0,$$

$$-i\omega\nu\xi_{3}+A\xi_{3}=0$$
(30)

(ν is a certain combination of the coefficients μ_i).

We obtain similarly the system of equations for the parameters v_y , ξ_2 , and ξ_4^* .

We note here that the mode corresponding to biaxial fluctuations is not hydrodynamic in the literal sense, i. e., its frequency ω does not vanish as $q \rightarrow 0$. However, although the wavelengths corresponding to the parameter Δ are small, they are still much larger than the intermolecular distances (~300 Å). The corresponding fluctuations can therefore be treated macroscopically.

To determine the spectrum of the scattered light, it is convenient to take in (30) the Laplace transform with respect to time

$$\xi_1(l) = \int dt \exp\left(-lt\right) \xi_1(t)$$

etc. We then obtain

$$\begin{aligned} (\gamma_{1}l+A_{1})\xi_{1}(l) + iC_{1}v_{x}(l) + \frac{l}{2}K_{2}q^{2}\sin 2\theta\xi_{3}(l) = \gamma_{1}\xi_{1}(0), \\ (\rho l+P_{1})v_{x}(l) - ilB_{1}\xi_{1}(l) = \rho v_{x}(0) - iB_{1}\xi_{1}(0), \\ v / \xi_{3} + \Delta\xi_{3} = v\xi_{3}(0), \end{aligned}$$
(31)

where $\xi_i(0)$ and $v_x(0)$ are the initial values of the corresponding parameters. From (31) we easily obtain

$$\begin{aligned} \xi_{2}^{*}(l) &= v \xi_{2}^{*}(0) / (vl + \Delta), \ \xi_{1}(l) &= \{ [\gamma_{1} l \rho + \gamma_{1} P_{1} - C_{1} B_{1}] \xi_{1}(0) - i \rho C_{1} v_{z}(0) \\ &+ i /_{z} K_{2} q^{2} \sin 2\theta \xi_{3}^{*}(l) \} \{ l^{2} \rho \gamma_{1} + l (\rho A_{1} + \gamma_{1} P_{1} - C_{1} B_{1}) + A_{1} P_{1} \}^{-1}. \end{aligned}$$
(32)

Similar formulas hold also for $\xi_4^*(l)$ and $\xi_2(l)$. To calculate the scattering cross section, we write down from symmetry considerations

$$\delta \varepsilon_{\alpha\beta} = M \delta Q_{\alpha\beta}^{\perp} + N (n_{\alpha} n_{1} \delta Q_{1\beta}^{\perp} + n_{\beta} n_{1} \delta Q_{\alpha1}^{\perp}).$$
(33)

If we confine ourselves to the case of scattering in the equatorial plane, we obtain from (32), (33), and (21)

$$\frac{d\sigma}{d\omega \, d\Omega} = \frac{M^2}{32\pi^3} \, \omega^4 \frac{T}{v \omega^2 + \Delta^2} \,. \tag{34}$$

In a more general geometry, the cross section for biaxial scattering is determined also by the correlator $\langle \xi_3^2 \rangle$ and by the mixed correlators $\langle \xi_1 \xi_3^* \rangle$ and $\langle \xi_2 \xi_4^* \rangle$ [the corresponding integral formulas are given in Ref. 4, and their generalization to inelastic scattering can be obtained from (32) in analogy with (34)].

IV. CONCLUSION

The principal results of the paper are the following. There exist geometrical conditions under which transverse fluctuations do not lead to scattering of light. Under these conditions the spectral intensity is determined by the dynamics of the longitudinal and biaxial fluctuations. If the wave vectors of the incident and scattered light lie in an equatorial plane, i. e., in a plane perpendicular to the director), while the polarization vectors of the incident and scattered rays are parallel to the director, the entire effect is determined only by the longitudinal scattering.

On the other hand, if the polarization vectors of the incident and scattered light lie in the equatorial plane, longitudinal and biaxial scattering takes place. However, if in addition the polarization vectors of the incident and scattered light are mutually orthogonal, then the intensity of the longitudinal scattering vanishes. To observe effects connected with longitudinal and biaxial scattering, it is necessary to satisfy these geometrical conditions with accuracy $5-10^{\circ}$ and to use samples with transverse dimensions not larger than 1 cm and not thicker than 0.1 mm. Otherwise the longitudinal and biaxial scattering will be masked by the effects of transverse single and multiple scattering.

We have calculated in this paper the line shapes of the longitudinal and biaxial scattering [formulas (23) and (34)]. The principal qualitative effects by which the character of the fluctuations can be determined from the line shape are the following:

1) In the longitudinal scattering, the line does not have a Lorentz shape.

2) In biaxial scattering, the line width does not depend on the wave vector transferred in the scattering.

These two conclusions differ substantially from the results obtained for scattering connected with the fluctuations of the director. This scattering has a Lorentz line shape, and a line width $\sim q^2$.

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¹⁾In this notation we neglect the contribution made to the longitudinal modes by the pure relaxation of s, which is not hydrodynamic at all in this region.

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