

Light scattering in gyrotropic media

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The scattering of light is considered with allowance for spatial dispersion of the medium. The Green's function for an electromagnetic field in a homogeneous medium with spatial dispersion is written down and the respective scattering intensity is calculated. The results are used to investigate the scattering of light in isotropic gyrotropic media. Fluctuations of the dielectric tensor (symmetric scattering) and of the gyrotropy (antisymmetric scattering) are taken into account. Scattering associated with fluctuations of the order parameter is considered in detail for physically diverse cases: the mixture stratification point, the phase transition in an intrinsic ferroelectric, the transition from the isotropic to the ordered phase of a cholesteric liquid crystal. The description is carried out in terms of circularly polarized waves. The angular and polarization dependences of the scattering intensity are calculated.

Light-scattering methods have been extensively applied in the investigation of the most diverse physical systems. The theory is based on a number of simplifying assumptions such as the possibility of restricting the treatment of the problem to the single-scattering approximation, optical isotropy of the medium, absence of spatial dispersion, neglect of gyrotropy, etc.^{1,2} At present the necessity often arises of treating quite complicated systems in which these assumptions break down. Thus, for example, in the investigation of scattering near the second-order phase transition points, higher orders of scattering prove to be important,^{3,4} and in the study of fluctuations in the ordered phase of nematic liquid crystals it is necessary to take account of the optical anisotropy peculiar to these objects^{5,7} as well as higher orders of scattering.^{5,6}

Recently great interest is attached to phase transitions of the type isotropic liquid—cholesteric liquid crystal (CLC), especially in connection with the existence of intermediate “blue” phases.⁸ The application of light-scattering methods to these systems requires an account of such specific properties as gyrotropy, which leads in particular to a rotation of the plane of polarization and circular dichroism.

The ensuing problem of constructing a theory of scattering in gyrotropic media is of independent interest. First, as is well known, the natural waves of such a medium are circularly polarized. Therefore it is most suitable to conduct the experiment in precisely these polarizations. This requires that the light scattering be described in terms of circular or elliptical polarizations. Second, in such media the dielectric tensor contains an antisymmetric part, whose fluctuations give rise to so-called antisymmetric scattering.¹ This contribution to the scattering can arise in the presence in the medium of a pseudovector corresponding to the intrinsic optical activity (a concrete example of such scattering, due to fluctuations of the magnetic moment close to the Curie point in a dielectric ferromagnet, was considered in Ref. 9) as well as in the case in which it is necessary to take account of spatial dispersion (natural optical activity). From this point of view the given problem leads to the more general problem of constructing a light-scattering theory which takes account of the spatial dispersion of the medium.

The present article is dedicated to a study of the effects of spatial dispersion, and in particular of intrinsic gyrotropy,

on the light scattering properties of a medium. The Green's function of the electromagnetic field in a homogeneous medium with spatial dispersion is constructed (taking account of anisotropy, gyrotropy, and absorption), and the corresponding light intensity is calculated. Both symmetric and antisymmetric scattering are investigated. The description is in terms of the natural waves of the medium. The scattering associated with fluctuations of the order parameter in optically isotropic, gyrotropic media near second-order phase transition points is considered in detail for physically diverse situations: the stratification point of a mixture (scalar order parameter), the phase transition in an intrinsic ferroelectric (vector order parameter), and the transition to the ordered phase of a CLC (here the order parameter is a traceless symmetric tensor of second rank).

1. LIGHT SCATTERING IN A MEDIUM WITH SPATIAL DISPERSION

The optical properties of a medium are characterized by the dielectric tensor $\epsilon_{\alpha\beta}$, which relates the electric field intensity \mathbf{E} with the electric induction \mathbf{D} . In particular, light scattering in a random-inhomogeneous medium can be described as taking place on fluctuational inhomogeneities of $\epsilon_{\alpha\beta}$.¹

It is well known that in general the relation between \mathbf{D} and \mathbf{E} is nonlocal. At the same time, the local form of the coupling equations is commonly used in the study of light scattering.^{1,2} This is explained by the fact that in most cases in optics the spatial dispersion is small—of the order of a/λ , where a is the characteristic structural or molecular dimension, and λ is the wavelength of the light.^{1,10} Along with this, there are effects in which the role of spatial dispersion is decisive, e.g., the phenomenon of optical activity (gyrotropy).

Various formulations exist of the electrodynamics of gyrotropic media.^{1,10,11} This has to do with the ambiguity of the definition of the electrical induction \mathbf{D} and the magnetic field intensity \mathbf{H} . We will use the form of the material equations commonly used in the electrodynamics of media with spatial dispersion^{1,10}:

$$D_{\alpha}(\mathbf{r}, t) = \int d\mathbf{r}_1 dt_1 f_{\alpha\beta}(\mathbf{r}, \mathbf{r}_1; t, t_1) E_{\beta}(\mathbf{r}_1, t_1), \quad (1)$$

$$\mathbf{H}(\mathbf{r}, t) = \mathbf{B}(\mathbf{r}, t),$$

where \mathbf{B} is the magnetic induction. The integration over t_1 is carried out by virtue of the causality principle over the limits $-\infty < t_1 \leq t$. Below we will assume that the dependence of $f_{\alpha\beta}$ on the variables t and t_1 is significantly slower than on their difference $t - t_1$. In this case the kernel $f_{\alpha\beta}(\mathbf{r}, \mathbf{r}_1; t, t_1)$ satisfies the Onsager symmetry principle $f_{\alpha\beta}(\mathbf{r}, \mathbf{r}_1; t, t_1) = f_{\beta\alpha}(\mathbf{r}_1, \mathbf{r}; t, t_1)$.^{10,12}

Substituting Eq. (1) into Maxwell's equations, and assuming the medium to be homogeneous on average, we obtain a closed equation for the field \mathbf{E} , which we write in the form

$$(\text{rot rot } \mathbf{E})_{\alpha} + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \int \bar{\epsilon}_{\alpha\beta}(\mathbf{r} - \mathbf{r}_1, t - t_1) E_{\beta}(\mathbf{r}_1, t_1) d\mathbf{r}_1 dt_1 = -\frac{1}{c^2} \frac{\partial^2}{\partial t^2} \int U_{\alpha\beta}(\mathbf{r}, \mathbf{r} - \mathbf{r}_1; t, t - t_1) E_{\beta}(\mathbf{r}_1, t_1) d\mathbf{r}_1 dt_1. \quad (2)$$

Here $\bar{\epsilon}_{\alpha\beta}(\mathbf{r} - \mathbf{r}_1, t - t_1) = \langle f_{\alpha\beta}(\mathbf{r}, \mathbf{r}_1; t, t_1) \rangle$ has the sense of a mean nonlocal dielectric tensor, and

$$U_{\alpha\beta}(\mathbf{r}, \mathbf{r} - \mathbf{r}_1; t, t - t_1) = f_{\alpha\beta}(\mathbf{r}, \mathbf{r}_1; t, t_1) - \bar{\epsilon}_{\alpha\beta}(\mathbf{r} - \mathbf{r}_1, t - t_1)$$

is its fluctuating part. The angular brackets denote the statistical average.

In what follows we will be interested in the intensity of the scattered light, which is defined by the quantity $\langle E'_{\alpha} E_{\beta}^{*\prime} \rangle$, where $\mathbf{E}' = \mathbf{E} - \langle \mathbf{E} \rangle$ is the field of the scattered wave. As is well known, the influence of fluctuations in a random-inhomogeneous medium in addition to directly causing light scattering; leads to the replacement of $\bar{\epsilon}_{\alpha\beta}$ by the effective dielectric tensor $\epsilon_{\alpha\beta}$.¹³ In the propagation of the light through the medium this gives such effects as attenuation of the mean field $\langle \mathbf{E} \rangle$, and in a gyrotropic medium also fluctuational rotation of its plane of polarization.^{14,15} To take account of these scattering effects, we will carry out the description in terms of the effective Green's function $T_{\alpha\beta}$.¹⁶ In the single-scattering approximation

$$\langle E'_{\alpha}(\mathbf{r}, t) E_{\beta}^{*\prime}(\mathbf{r}, t) \rangle = \int T_{\alpha\nu}(\mathbf{r} - \mathbf{r}_1, t - t_1) T_{\beta\mu}(\mathbf{r} - \mathbf{r}_2, t - t_2) \times \langle U_{\nu\rho}(\mathbf{r}_1, \mathbf{r}_1 - \mathbf{r}_3; t_1, t_1 - t_3) U_{\mu\sigma}(\mathbf{r}_2, \mathbf{r}_2 - \mathbf{r}_4; t_2, t_2 - t_4) \rangle \langle E_{\rho}(\mathbf{r}_3, t_3) \rangle \times \langle E_{\sigma}(\mathbf{r}_4, t_4) \rangle d\mathbf{r}_3 d\mathbf{r}_4 dt_3 dt_4. \quad (3)$$

The usual experimental setup in light-scattering experiments corresponds to the far-field approximation $r \gg \lambda$. The mean field $\langle E(\mathbf{r}, t) \rangle$ is here a sum of plane normal waves of the form

$$E_{\rho} \mathbf{e}^{(j)} \exp[i(\mathbf{k}_{\rho}^{(j)} \mathbf{r} - \omega_{\rho} t)]. \quad (4)$$

Here $\mathbf{e}^{(j)}$ and $\mathbf{k}_{\rho}^{(j)}$ are the effective polarizations and the wave vectors. If we substitute Eq. (4) into Eq. (3) and take account of the fact that the frequency of the incident light ω_{ρ} is much greater than ω_U , the characteristic frequency of variation of $\bar{U}(\mathbf{r}, \rho; t, \tau)$ with respect to the variable t , then it can be easily shown that to find the scattering intensity it is sufficient to know the Green's function $T_{\alpha\beta}$ in the (\mathbf{r}, ω) representation in the far field.

Since media with spatial dispersion are in many cases anisotropic (e.g., gyrotropic crystals), we will construct the

Green's function $\hat{T}(\mathbf{r}, \omega)$ with account of both spatial dispersion and optical anisotropy. It is convenient to write it in terms of the polarization vectors $\mathbf{e}^{(j)}$ and the wave vectors $\mathbf{k}_{(j)}$ of the normal waves in the medium:

$$T_{\alpha\beta}(\mathbf{r}, \omega) = \frac{(\omega/c)^2}{4\pi r} \sum_{j=1}^3 \sum_{\mathbf{k}=\mathbf{k}_{st}^{(j)}} \frac{e^{i\mathbf{k}\mathbf{r}}}{t^{(j)}(\mathbf{k})} e_{\alpha}^{(j)}(\mathbf{k}) e_{\beta}^{(j)}(-\mathbf{k}). \quad (5)$$

Here

$$t^{(j)}(\mathbf{k}) = [1 - (\mathbf{k}\mathbf{e}^{(j)}(\mathbf{k}))^2/k^2] (\det \hat{N}^{(j)})^{1/2},$$

$$\hat{N}^{(j)}(\mathbf{k}) = \epsilon^{1/2} \nabla_{\mathbf{k}_{\perp}} \otimes \nabla_{\mathbf{k}_{\perp}} \mathbf{k}_{(j)}(\mathbf{k}, \omega), \quad j=1, 2,$$

$$t^{(3)}(\mathbf{k}) = -(\det \hat{N}^{(3)})^{1/2} (c/\omega)^2,$$

$$\hat{N}^{(3)}(\mathbf{k}) = \epsilon^{1/2} \nabla_{\mathbf{k}_{\perp}} \otimes \nabla_{\mathbf{k}_{\perp}} [e_{\alpha}^{(3)}(-\mathbf{k}) \epsilon_{\alpha\beta}(\mathbf{k}, \omega) e_{\beta}^{(3)}(\mathbf{k})],$$

$\hat{N}^{(j)}$ are 2×2 matrices, and \mathbf{k}_{\perp} is the component of \mathbf{k} orthogonal to \mathbf{r} . For $j=1$ and 2 , $\mathbf{e}^{(j)}(\mathbf{k}) = \epsilon^{-1}(\mathbf{k}, \omega) \mathbf{d}^{(j)}$, $\mathbf{d}^{(j)}$ are eigenvectors, and $(\omega/c)^2 \mathbf{k}_{(j)}^2(\mathbf{k}, \omega)$ are eigenvalues of the two-dimensional tensor $\hat{\epsilon}^{-1}(\mathbf{k}, \omega)$ acting in a plane orthogonal to \mathbf{k} [we assume that $\det \hat{\epsilon}(\mathbf{k}, \omega) \neq 0$]. On the dispersion surfaces $k^2 - k_{(j)}^2(\mathbf{k}, \omega) = 0$ the corresponding vectors \mathbf{k} are the wave vectors of the normal waves in the medium, and \mathbf{e}^j are their polarizations. The third term in Eq. (5) corresponds to the longitudinal waves $e^{(3)}(-\mathbf{k}) \| e^{(3)}(\mathbf{k}) \| \mathbf{k}$, and the equality $e_{\alpha}^{(3)}(-\mathbf{k}) \epsilon_{\alpha\beta}(\mathbf{k}) e_{\beta}^{(3)}(\mathbf{k}) = 0$ is the condition of their existence. The second sum in Eq. (5) is over the stationary points $\mathbf{k}_{st}^{(j)}$ lying on the corresponding dispersion surface ($k^2 - k_{(j)}^2(\mathbf{k}) = 0$ for $j=1, 2$ and $e_{\alpha}^{(3)} \epsilon_{\alpha\beta} e_{\beta}^{(3)} = 0$ for $j=3$), in which the outward normal is aligned with \mathbf{r} .

For the case of a medium without spatial dispersion a formula analogous to Eq. (5) was obtained in Ref. 7 (see also Ref. 5). Formula (5) differs from the results of Refs. 5 and 7 in that it takes into account additional waves that are possible in a medium with spatial dispersion.¹⁰ In addition, in contrast with Refs. 5 and 7, the tensor $\epsilon_{\alpha\beta}(\mathbf{k}, \omega)$ is neither symmetric nor Hermitian, but does obey the Onsager relation $\epsilon_{\alpha\beta}(\mathbf{k}, \omega) = \epsilon_{\beta\alpha}(-\mathbf{k}, \omega)$. As a result, it was necessary to use two systems of vectors $\{\mathbf{e}^{(j)}(\mathbf{k})\}$ and $\{\mathbf{e}^{(j)}(-\mathbf{k})\}$ in Eq. (5).

Substituting expressions (4) and (5) into Eq. (3), we obtain

$$\langle E'_{\alpha}(\mathbf{r}, t) E_{\beta}^{*\prime}(\mathbf{r}, t) \rangle = \frac{1}{16\pi^2 r^2} |E_{\rho}^{(i)}|^2 \sum_{j=1}^3 \sum_{\mathbf{k}=\mathbf{k}_{st}^{(j)}} \frac{V_{\text{eff}}^{(j)}}{|t^{(j)}(\mathbf{k})|^2} \times e^{-2\mathbf{k}''\mathbf{r}} \int \frac{d\omega}{2\pi} G_{\nu\rho\mu\lambda}(\mathbf{q}', \mathbf{k}_{\rho}^{(i)}, -\mathbf{k}_{\rho}^{(i)*}; \omega, \omega_{\rho}, -\omega_{\rho}) \times e_{\rho}^{(i)} e_{\lambda}^{(i)*} e_{\nu}^{(j)}(-\mathbf{k}) e_{\mu}^{(j)*}(-\mathbf{k}) e_{\alpha}^{(j)}(\mathbf{k}) e_{\beta}^{(j)*}(\mathbf{k}). \quad (6)$$

Here the index (i) refers to the incident light, and the index (j) , to the scattered light; $E_{\rho}^{(i)}$, $\mathbf{k}_{\rho}^{(i)}$, and $e^{(i)}$ are the amplitude, wave vector, and polarization of the incident light, $\mathbf{q} = \mathbf{q}_{(i,j)} = \mathbf{k}_{st}^{(j)} - \mathbf{k}_{\rho}^{(i)}$, $\mathbf{q}' = \text{Re } \mathbf{q}$, $\mathbf{k}_{st}'' = \text{Im } \mathbf{k}_{st}$, $G_{\nu\rho\mu\lambda}(\mathbf{q}', \mathbf{k}_{\rho} - \mathbf{k}_{\rho}^*, \omega, \omega_{\rho}, -\omega_{\rho})$ is the Fourier transform of the correlation function

$$G_{\nu\rho\mu\lambda}(\mathbf{r}_1 - \mathbf{r}_2, \rho_1, \rho_2; t_1 - t_2, \tau_1, \tau_2) = \langle U_{\nu\rho}(\mathbf{r}_1, \rho_1; t_1, \tau_1) U_{\mu\lambda}(\mathbf{r}_2, \rho_2; t_2, \tau_2) \rangle, \quad (7)$$

$$V_{\text{eff}}^{(j)} = \int \exp(2\mathbf{q}''\mathbf{R}) d\mathbf{R}, \quad \mathbf{q}'' = \text{Im } \mathbf{q}, \quad \mathbf{R} = (\mathbf{r}_1 + \mathbf{r}_2)/2,$$

where the points \mathbf{r}_1 and \mathbf{r}_2 belong to the scattering volume V .

In the derivation of formulation (6) it was assumed that $r \gg V^{1/3} \gg \lambda$.

Formula (6) makes it possible to calculate the intensity of the scattered light—the modulus of the Poynting vector \mathbf{S} , which in a medium with spatial dispersion is connected with the correlator $\langle E_\alpha E_\beta^* \rangle$ by the relation^{10,12}:

$$S_\alpha = \frac{c^2}{8\pi\omega} \operatorname{Re} \left[k_\alpha \delta_{\beta\nu} - k_\nu \delta_{\alpha\beta} - \frac{1}{2} \frac{\omega^2}{c^2} \frac{\partial \varepsilon_{\nu\beta}(\mathbf{k}, \omega)}{\partial k_\alpha} \right] \langle E_\beta E_\nu^* \rangle, \quad (8)$$

where \mathbf{E} is one of the normal modes of the scattered field, and \mathbf{k} is its wave vector.

2. THE MATERIAL EQUATION IN A MEDIUM WITH WEAK SPATIAL DISPERSION. GYROTROPY

Let us consider an inhomogeneous medium in the case of weak spatial dispersion (we neglect henceforth temporal dispersion). The kernel $f_{\alpha\beta}(\mathbf{r}, \mathbf{r}_1)$ in Eq. (1), which characterizes the nonlocality of the medium, is nonzero in the region $|\mathbf{r} - \mathbf{r}_1| \sim a$. The weakness of the spatial dispersion corresponds to the smallness of the parameters a/λ (in optics it is usually $\sim 10^{-2} - 10^{-3}$ [Ref. 10]). This makes it possible to expand $E(\mathbf{r}_1, t_1)$ in Eq. (1) in a series under the integral in the neighborhood of the point $\mathbf{r}_1 = \mathbf{r}$ and to obtain the material equation, with accuracy to terms of the order of $(a/\lambda)^3$, in the form

$$D_\alpha(\mathbf{r}) = W_{\alpha\beta}(\mathbf{r}) E_\beta(\mathbf{r}) + P_{\alpha\beta\gamma}(\mathbf{r}) \nabla_\gamma E_\beta(\mathbf{r}) + Q_{\alpha\beta\gamma\delta}(\mathbf{r}) \nabla_\gamma \nabla_\delta E_\beta(\mathbf{r}). \quad (9)$$

The Onsager symmetry $f_{\alpha\beta}(\mathbf{r}, \mathbf{r}_1) = f_{\beta\alpha}(\mathbf{r}_1, \mathbf{r})$ for the kernel \hat{f} leads to the following relations between the coefficients $W_{\alpha\beta}(\mathbf{r})$, $P_{\alpha\beta\gamma}(\mathbf{r})$, and $Q_{\alpha\beta\gamma\delta}(\mathbf{r})$:

$$\begin{aligned} W_{\alpha\beta}(\mathbf{r}) - W_{\beta\alpha}(\mathbf{r}) &= \nabla_\gamma P_{\alpha\beta\gamma}(\mathbf{r}) - \nabla_\gamma \nabla_\delta Q_{\alpha\beta\gamma\delta}(\mathbf{r}), \\ P_{\alpha\beta\gamma}(\mathbf{r}) + P_{\beta\alpha\gamma}(\mathbf{r}) &= 2\nabla_\mu Q_{\alpha\beta\gamma\mu}(\mathbf{r}), \\ Q_{\alpha\beta\gamma\delta}(\mathbf{r}) &= Q_{\beta\alpha\gamma\delta}(\mathbf{r}) = Q_{\alpha\beta\delta\gamma}(\mathbf{r}). \end{aligned} \quad (10)$$

Analogous relations can also be obtained by taking account of the subsequent terms in the expansion of \mathbf{D} in terms of the derivatives of $\mathbf{E}(\mathbf{r})$. Let us now turn our attention to the difference between the parameters of the expansions used in the derivation of Eq. (9) and Eqs. (10). Equation (9) is valid for $a/\lambda \ll 1$, and Eqs. (10) are valid for $a/r_0 \ll 1$, where r_0 is the characteristic dimension of the inhomogeneity of $f_{\alpha\beta}(\mathbf{r}, \mathbf{r}_1)$ with respect to the variable \mathbf{r} .

It is convenient to transform from the tensors \hat{W} , \hat{P} , and \hat{Q} , with account taken of relations (10), to $\hat{\varepsilon}^{(10)}$, the symmetric part of \hat{W} , and to $2\hat{\gamma}$, the antisymmetric part (with respect to $\alpha\beta$) of $P_{\alpha\beta\gamma}$. Then the material Eq. (9) takes the form

$$\begin{aligned} D_\alpha(\mathbf{r}) &= [\varepsilon_{\alpha\beta}^{(10)}(\mathbf{r}) + \nabla_\nu \gamma_{\alpha\beta\nu}(\mathbf{r})] E_\beta(\mathbf{r}) \\ &+ [2\gamma_{\alpha\beta\nu}(\mathbf{r}) + \nabla_\delta Q_{\alpha\beta\nu\delta}(\mathbf{r})] \nabla_\nu E_\beta(\mathbf{r}) \\ &+ Q_{\alpha\beta\nu\delta}(\mathbf{r}) \nabla_\nu \nabla_\delta E_\beta(\mathbf{r}), \end{aligned} \quad (11)$$

where

$$\begin{aligned} \varepsilon_{\alpha\beta}^{(10)}(\mathbf{r}) &= \int f_{\alpha\beta}^{(s)}(\mathbf{r}, \mathbf{r} + \boldsymbol{\rho}) d\boldsymbol{\rho}, \\ \gamma_{\alpha\beta\nu}(\mathbf{r}) &= \frac{1}{2} \int f_{\alpha\beta}^{(a)}(\mathbf{r}, \mathbf{r} + \boldsymbol{\rho}) \rho_\nu d\boldsymbol{\rho}, \\ Q_{\alpha\beta\nu\delta}(\mathbf{r}) &= \frac{1}{2} \int f_{\alpha\beta}^{(s)}(\mathbf{r}, \mathbf{r} + \boldsymbol{\rho}) \rho_\nu \rho_\delta d\boldsymbol{\rho}. \end{aligned}$$

Here $f_{\alpha\beta}^{(s)}$ and $f_{\alpha\beta}^{(a)}$ are the symmetric and antisymmetric (with respect to $\alpha\beta$) parts of the tensor $f_{\alpha\beta}$. If the medium is gyrotropic, then the tensor $\gamma_{\alpha\beta\nu}(\mathbf{r})$ is nonzero, and it is possible to restrict expansion (11) to terms that contain the first derivatives with respect to the coordinates

$$D_\alpha(\mathbf{r}) = [\varepsilon_{\alpha\beta}^{(0)}(\mathbf{r}) + \nabla_\nu \gamma_{\alpha\beta\nu}(\mathbf{r})] E_\beta(\mathbf{r}) + 2\gamma_{\alpha\beta\nu}(\mathbf{r}) \nabla_\nu E_\beta(\mathbf{r}). \quad (12)$$

The coupling equations were used in such a form in Refs. 17 and 18. In a spatially inhomogeneous medium $\nabla_\nu \gamma_{\alpha\beta\nu}(\mathbf{r}) = 0$, and we arrive at the usual form of the material equation for a gyrotropic medium.¹

Frequently, along with $\gamma_{\alpha\beta\nu}$, use is made of the gyration tensor of second rank

$$\kappa_{\nu\rho}(\mathbf{r}) = \frac{1}{2} e_{\nu\alpha\beta} \gamma_{\alpha\beta\rho}(\mathbf{r}), \quad (13)$$

where $e_{\nu\alpha\beta}$ is a unit antisymmetric tensor of third rank. The tensors $\varepsilon_{\alpha\beta}^{(0)}$ and $\kappa_{\alpha\beta}$ in turn can be resolved into their irreducible parts

$$\begin{aligned} \kappa_{\alpha\beta}(\mathbf{r}) &= \tau_{\alpha\beta}(\mathbf{r}) + e_{\alpha\beta\gamma} \mu_\gamma(\mathbf{r}) + \nu(\mathbf{r}) \delta_{\alpha\beta}, \\ \varepsilon_{\alpha\beta}^{(0)}(\mathbf{r}) &= \varphi_{\alpha\beta}(\mathbf{r}) + \varepsilon^{(0)}(\mathbf{r}) \delta_{\alpha\beta}. \end{aligned} \quad (14)$$

Here $\hat{\tau}(\mathbf{r})$ and $\hat{\varphi}(\mathbf{r})$ are symmetric traceless tensors, $\boldsymbol{\mu}(\boldsymbol{\tau})$ is a vector, and $\nu(\mathbf{r})$ and $\varepsilon^{(0)}(\mathbf{r})$ are scalars.

3. LIGHT SCATTERING IN AN ISOTROPIC GYROTROPIC MEDIUM

If the medium undergoes fluctuations, then the tensors $\hat{\varepsilon}^{(0)}(\mathbf{r})$ and $\hat{\gamma}(\mathbf{r})$ in Eq. (12) are random quantities. The fluctuating tensor $U_{\alpha\beta}(\mathbf{r}, \mathbf{r} - \mathbf{r}_1)$, according to Eq. (12), is equal to¹⁾

$$\begin{aligned} U_{\alpha\beta}(\mathbf{r}, \mathbf{r} - \mathbf{r}_1) &= \left[\delta \varepsilon_{\alpha\beta}(\mathbf{r}) + \frac{\partial \delta \gamma_{\alpha\beta\nu}(\mathbf{r})}{\partial r_\nu} \right. \\ &\left. + 2\delta \gamma_{\alpha\beta\nu}(\mathbf{r}) \frac{\partial}{\partial r_\nu} \right] \delta(\mathbf{r} - \mathbf{r}_1). \end{aligned} \quad (15)$$

Assuming the medium to be isotropic on the average, the effective dielectric tensor $\varepsilon_{\alpha\beta}$ in the (\mathbf{k}, ω) representation can be written in the form

$$\varepsilon_{\alpha\beta}(\mathbf{k}, \omega) = \varepsilon \delta_{\alpha\beta} + 2i\nu k_\mu e_{\alpha\beta\mu}, \quad (16)$$

where ε and ν are constants. Below we will confine ourselves to the case of real ε and ν . In this case two transverse waves are natural waves of the medium: one with polarization $\mathbf{e}^{(1)} = \mathbf{m}(\mathbf{p})$ and wave vector $\mathbf{k}_{(1)} = \mathbf{k}_{+1}$ (circular wave with right-handed polarization), and the other with polarization $\mathbf{e}^{(2)} = \mathbf{m}^*(\mathbf{p})$ and wave vector $\mathbf{k}_{(2)} = \mathbf{k}_{-1}$ (circular wave with left polarization), where

$$\mathbf{k}_{\pm 1} = \frac{\omega}{c} \left\{ \left[\varepsilon + \left(\frac{\nu\omega}{c} \right)^2 \right]^{1/2} \mp \frac{\nu\omega}{c} \right\} \mathbf{p}, \quad (17)$$

and \mathbf{p} is the direction of propagation of the wave.¹

The quantities $\hat{N}^{(j)}$ and $t^{(j)}$ in Eq. (5) are in the present case equal to

$$\begin{aligned} N_{\alpha\beta}^{(j)} &= (-1)^j \delta_{\alpha\beta} \nu \omega^2 / k_{(j)} c^2, \\ t^{(j)} &= \omega k_{(j)} / \{ c [\varepsilon + (\nu\omega/c)^2]^{1/2} \}, \quad j=1, 2 \end{aligned}$$

and the Green's function (5) takes the form

$$\begin{aligned} T_{\alpha\beta}(\mathbf{r}, \omega) &= \frac{\omega}{4\pi r c [\varepsilon + (\nu\omega/c)^2]^{1/2}} [\mathbf{k}_{+1} m_\alpha m_\beta^* e^{i\mathbf{k}_{+1}\cdot\mathbf{r}} \\ &+ \mathbf{k}_{-1} m_\alpha^* m_\beta e^{i\mathbf{k}_{-1}\cdot\mathbf{r}}]. \end{aligned} \quad (18)$$

Here it has been taken into account that $\mathbf{m}^*(\mathbf{p}) = \mathbf{m}(-\mathbf{p})$.

Since the electric field is transverse the tensor $\langle E'_\alpha(\mathbf{r}) E'^*_\beta(\mathbf{r}) \rangle$ is determined by four independent parameters, and in a gyrotropic medium it is convenient to take as such parameters

$$\langle |E_{+1}'(\mathbf{r})|^2 \rangle, \langle |E_{-1}'(\mathbf{r})|^2 \rangle, \langle E_{+1}'(\mathbf{r}) E_{-1}'^*(\mathbf{r}) \rangle, \langle E_{-1}'(\mathbf{r}) E_{+1}'^*(\mathbf{r}) \rangle.$$

Here $E_{+1}'(\mathbf{r})$ and $E_{-1}'(\mathbf{r})$ are the amplitudes of the right and left circularly polarized waves in the expansion of the scattered wave

$$E_\alpha'(\mathbf{r}) = m_\alpha(\mathbf{s}) E_{+1}'(\mathbf{r}) + m_\alpha^*(\mathbf{s}) E_{-1}'(\mathbf{r}), \quad (19)$$

and \mathbf{s} is its direction of propagation. For the Poynting vector of the scattering wave, Eq. (8) gives in the present case

$$\mathbf{S}_{\pm 1}' = \frac{c^2}{8\pi\omega} \left[\varepsilon + \left(\frac{v\omega}{c} \right)^2 \right]^{1/2} \langle |E_{\pm 1}'(\mathbf{r})|^2 \rangle \mathbf{s}. \quad (20)$$

We call attention to the fact that the cross correlators $\langle E_{+1}' E_{-1}'^* \rangle$ and $\langle E_{-1}' E_{+1}'^* \rangle$ do not contribute to the energy flux.

Calculating $\langle |E_{+1}'|^2 \rangle$ and $\langle |E_{-1}'|^2 \rangle$ from Eq. (6), we find with the help of Eq. (20) expressions for the intensities of the left- and right-circularly polarized components of the scattered light for the various circular polarizations of the incident light

$$I(\alpha, \beta) = I_0 C_{(\alpha)} \left(-i\beta \varepsilon_{\gamma\lambda} p_\lambda + \delta_{\gamma\lambda} p_\gamma p_\lambda \right) \left(-i\alpha \varepsilon_{\nu\mu\sigma} \delta_{\nu\mu} - s_\nu s_\mu \right) \left\{ \langle \delta \varepsilon_{\mu\tau} \delta \varepsilon_{\nu\kappa} \rangle_{\mathbf{q}} + i g_{\mathbf{q}} \left(\langle \delta \gamma_{\mu\tau} \delta \varepsilon_{\nu\kappa} \rangle_{\mathbf{q}} - \langle \delta \varepsilon_{\mu\tau} \delta \gamma_{\nu\kappa} \rangle_{\mathbf{q}} \right) + g_{\mathbf{q}} g_{\mathbf{q}} \langle \delta \gamma_{\mu\tau} \delta \gamma_{\nu\kappa} \rangle_{\mathbf{q}} \right\}, \quad (21)$$

where

$$C_{(\alpha)} = \frac{\omega^2 V k_{(\alpha)}^2}{64\pi^2 r^2 c^2 [\varepsilon + (v\omega/c)^2]}, \quad g_{\mathbf{q}} = k_{(\alpha) s_{\mathbf{q}}} + k_{(\beta) p_{\mathbf{q}}}.$$

Here I_0 is the intensity of the incident light, β and α are numbers which characterize the polarizations of the incident and scattered light (+1—right, and -1—left circular polarization), and $\mathbf{q} = k_{(\alpha)} \mathbf{s} - k_{(\beta)} \mathbf{p}$ is the scattering vector. The subscript \mathbf{q} of the fluctuating quantities denotes the corresponding Fourier components

$$f_{\mathbf{q}} = \int d\mathbf{r} e^{-i\mathbf{q}\cdot\mathbf{r}} f(\mathbf{r}).$$

Note that in general, independently of the type of polarization of the incident light, waves of both polarizations are formed in the scattering. Since the point group of a gyrotropic medium does not contain an inversion center, a simultaneous reversal of the polarization of the incident and scattered light ($\alpha \rightarrow -\alpha, \beta \rightarrow -\beta$) can result in a change in the intensity of the scattered light.

4. ANALYSIS OF PARTICULAR CASES

Both fluctuations of the dielectric tensor $\delta \varepsilon_{\alpha\beta}$ (symmetric scattering) and fluctuations of the gyrotropy $\delta \gamma_{\alpha\beta\lambda}$ (antisymmetric scattering) can contribute to the intensity of the scattered light (21). The main contribution to the intensity in Eq. (21) comes from the first term inside the braces, which is not associated with fluctuations of the gyrotropy. The other two terms have additional coefficients respectively of the order of ka and $k^2 a^2$, and their contributions are, as a rule, small. However, taking the fluctuations of the gyro-

tropy into account can in some cases lead to qualitative changes in the light scattering picture, for example, to a difference in the intensities $I(+1, -1)$ and $I(-1, +1)$. The fluctuations of the gyrotropy can be important when the contribution of the fluctuations of the dielectric tensor to the scattering are for some reasons small.

The nature of the gyrotropy fluctuations can vary. They can be spontaneous as well as induced by fluctuations of other thermodynamic quantities. Let us first consider the case of induced fluctuations. It is most interesting to study them near second-order phase transition points, where fluctuations of the order parameter $\hat{\eta}$ are strongly developed. If the dielectric tensor $\hat{\varepsilon}$ and the gyration tensor $\hat{\kappa}$ are functionally related with this parameter, i.e., $\partial \hat{\varepsilon} / \partial \hat{\eta} \neq 0, \partial \hat{\kappa} / \partial \hat{\eta} \neq 0$, then fluctuations of $\hat{\eta}$ can lead to significant fluctuations of the dielectric tensor and of the gyrotropy of the medium. Fluctuations only of those irreducible parts of $\hat{\varepsilon}$ and $\hat{\eta}$ in Eq. (14) which have the same tensor dimension as $\hat{\eta}$ will be induced in this case. We will analyze separately the case in which the order parameter is a scalar, a vector, and a traceless symmetric tensor of second rank.

In liquid mixtures, for example, close to the stratification point, the order parameter is a scalar (the concentration) and if one of the components is chiral, the fluctuations of η can cause fluctuations not only of $\delta \hat{\varepsilon}$, but also of $\delta \hat{\nu}$, the scalar part of the gyration tensor κ , thus

$$\delta \varepsilon_{\mathbf{q}} = A_{(s)} \delta \eta_{\mathbf{q}}, \quad \delta \nu_{\mathbf{q}} = B_{(s)} \delta \eta_{\mathbf{q}}. \quad (22)$$

Here and below, the subscript of A and B indicates the type of order parameter η : (s)—scalar, (v)—vector, and (t)—tensor. Allowing in Eq. (21) only for the contributions of the correlators of the strongly fluctuating quantities $\langle \delta \varepsilon^2 \rangle_{\mathbf{q}}$, $\langle \delta \varepsilon \delta \nu \rangle_{\mathbf{q}}$, and $\langle \delta \nu^2 \rangle_{\mathbf{q}}$, we obtain for the scattering intensities

$$I(\alpha, \beta) = I_0 C_0 \langle \delta \eta^2 \rangle_{\mathbf{q}} [A_{(s)}^2 f_{(s\varepsilon)} - k A_{(s)} B_{(s)} f_{(s\nu)} + k^2 B_{(s)}^2 f_{(v\nu)}], \quad (23)$$

where the functions $f_{(s\varepsilon)}$, $f_{(s\nu)}$, and $f_{(v\nu)}$ determine the contributions of the corresponding correlators

$$f_{(s\varepsilon)} = 2(\alpha\beta \cos \theta + 1 - 1/2 \sin^2 \theta), \quad f_{(s\nu)} = 8(\alpha + \beta) \cos^4(\theta/2), \\ f_{(v\nu)} = 2 \sin^2 \theta (1 - \alpha\beta + 2 \cos^2(\theta/2)), \quad (24)$$

and θ is the scattering angle. In the derivation of Eq. (23), in view of the smallness of ν it was assumed that

$$k_{(+1)} = k_{(-1)} = k, \quad q = 2k \sin(\theta/2), \\ C_{(\alpha)} = C_0 = \omega^2 V k^2 / (64\pi^2 r^2 c^2 \varepsilon).$$

We will also use this approximation below. Note that the dependence of the intensity on the polarization types (β, α) of the incident and scattered light are completely determined by the functions f , and that the dependence on the scattering angle θ is determined also by the correlation function in expression (23).

If we consider the usual situation, in which one need only take account of fluctuations of the dielectric tensor, then, as follows from Eqs. (23) and (24), scattering without change of polarization type takes place preferentially forward, and scattering with change of polarization type, preferentially backward. We emphasize that the somewhat unusual character of such a picture is connected not with the gyrotropy of the medium, but with the fact that the descrip-

tion is given in terms of circular polarizations.

If the gyrotropy fluctuations are taken into account, then an anomaly is introduced into the scattering by the fact that the contribution of the correlator $\langle \delta \varepsilon \delta \nu \rangle$, in contrast with the contributions of $\langle \delta \varepsilon \delta \varepsilon \rangle$ and $\langle \delta \nu \delta \nu \rangle$, changes sign upon simultaneous reversal of the polarizations of the incident and scattered light. Such an asymmetry in the case of a scalar order parameter can therefore serve as a basis for the detection of fluctuations of the gyrotropy of the medium.

Let us consider the case of the vector order parameter $\boldsymbol{\eta}$ in the symmetric phase. A situation is frequently encountered in which the linear dependence of $\delta \hat{\varepsilon}$ on $\boldsymbol{\eta}$ is absent. In particular, in the majority of ferroelectrics, where the role of $\boldsymbol{\eta}$ is played by the polarization, the dependence of $\hat{\varepsilon}$ on $\boldsymbol{\eta}$ is only quadratic (a linear relation can prevail only for ferroelastics¹⁹). In this case in the symmetric phase the contribution of the fluctuations of the dielectric tensor to the scattering is proportional to $\delta \eta^4$ and is a small quantity.^{19,20} Along with this, the tensor $\kappa_{\alpha\beta}$, in contrast with $\varepsilon_{\alpha\beta}$, has an antisymmetric part determined by the vector $\boldsymbol{\mu}$ in Eq. (14), and a linear relation $\boldsymbol{\mu} = \mathbf{B}_{(v)} \boldsymbol{\eta}$ is possible.²¹

If the crystal is optically isotropic, then it is possible to use Eq. (21) to calculate the scattering intensities. Allowing in it only for fluctuations of the vector part of the gyration tensor, we obtain

$$I(\alpha, \beta) = I_0 C_0 B_{(v)}^2 k^2 [\langle \eta^{\perp 2} \rangle_{\mathbf{q}} f_{(\mu\mu)}^{\perp} + \langle \eta^{\parallel 2} \rangle_{\mathbf{q}} f_{(\mu\mu)}^{\parallel}], \quad (25)$$

where $f_{(\mu\mu)}^{\parallel}$ and $f_{(\mu\mu)}^{\perp}$ correspond to the contributions of the longitudinal $\boldsymbol{\eta}_{\mathbf{q}}^{\parallel} = \mathbf{q}(\mathbf{q} \cdot \boldsymbol{\eta})/\mathbf{q}$ and the transverse $\boldsymbol{\eta}_{\mathbf{q}}^{\perp} = \boldsymbol{\eta}_{\mathbf{q}} - \boldsymbol{\eta}_{\mathbf{q}}^{\parallel}$ components of $\boldsymbol{\eta}_{\mathbf{q}}$ and are given by the expressions

$$\begin{aligned} f_{(\mu\mu)}^{\parallel} &= 4 \sin^2 \theta \cos^2(\theta/2), \\ f_{(\mu\mu)}^{\perp} &= 2 \sin^2 \theta (1 - \alpha\beta). \end{aligned} \quad (26)$$

Note that in this case the intensity of scattering by the longitudinal fluctuations does not depend on the polarization types of the incident and scattered light, and that scattering by the transverse fluctuations takes place only with reversal of the polarization.

If the order parameter $\hat{\eta}$ is a traceless symmetric tensor, as, for example, in the isotropic phase of a CLC, where it is possible to choose as the order parameter the anisotropic part $\varphi_{\alpha\beta}$ of the dielectric tensor, then in expression (21) it is sufficient to take into account only the fluctuations of $\varphi_{\alpha\beta}$ and $\tau_{\alpha\beta}$, putting $\tau_{\alpha\beta, \mathbf{q}} = \mathbf{B}_{(t)} \varphi_{\alpha\beta, \mathbf{q}}$ and $A_{(t)} = 1$.

The tensors $\varphi_{\alpha\beta}$ and $\tau_{\alpha\beta}$ can be expanded over the tensor basis $\sigma_{\alpha\beta}^{(l)}$ ($l = -2, -1, 0, 1, 2$) [Ref. 14], constructed with the help of the only preferred $\mathbf{n} = \mathbf{q}/q$:

$$\begin{aligned} \sigma_{\alpha\beta}^{(2)} &= m_{\alpha}(\mathbf{n}) m_{\beta}(\mathbf{n}), & \sigma_{\alpha\beta}^{(-2)} &= \sigma_{\alpha\beta}^{(2)*}, \\ \sigma_{\alpha\beta}^{(1)} &= (1/2)^{1/2} i [m_{\alpha}(\mathbf{n}) n_{\beta} + m_{\beta}(\mathbf{n}) n_{\alpha}], & \sigma_{\alpha\beta}^{(-1)} &= -\sigma_{\alpha\beta}^{(1)*}, \\ \sigma_{\alpha\beta}^{(0)} &= (3/2)^{1/2} (n_{\alpha} n_{\beta} - 1/3 \delta_{\alpha\beta}). \end{aligned} \quad (27)$$

In this case the following relations should hold:

$$\sigma_{\alpha\beta}^{(s)} \sigma_{\beta\alpha}^{(p)} = \delta_{sp}, \quad \sigma_{\alpha\beta}^{(l)}(\mathbf{n}) = \sigma_{\alpha\beta}^{(l)*}(-\mathbf{n}).$$

The order parameter is represented in the form of a sum

$$\varphi_{\alpha\beta, \mathbf{q}} = \sum_{l=-2}^2 \xi_{\mathbf{q}}^{(l)} \sigma_{\alpha\beta}^{(l)}, \quad (28)$$

where the expansion coefficients are $\xi_{\mathbf{q}}^{(l)} = \varphi_{\alpha\beta, \mathbf{q}} \sigma_{\alpha\beta}^{(l)*}$, with $\xi_{\mathbf{q}}^{(l)} = \xi_{\mathbf{q}}^{(l)*}$. In an isotropic medium $\langle \xi_{\mathbf{q}}^{(l)} \xi_{\mathbf{q}}^{(s)*} \rangle_{\mathbf{q}} = \langle \xi^{(l)2} \rangle_{\mathbf{q}} \delta_{ls}$ and the scattering intensity can be broken down into contributions of the individual modes $\xi_{\mathbf{q}}^{(l)}$, each of which has its own angular and polarizational dependence

$$I(\alpha, \beta) = I_0 C_0 \sum_{l=-2}^2 (f_{(\varphi\varphi)}^{(l)} - k B_{(t)} f_{(\tau\tau)}^{(l)} + k^2 B_{(t)}^2 f_{(\tau\tau)}^{(l)}) \langle \xi^{(l)2} \rangle_{\mathbf{q}}, \quad (29)$$

where

$$\begin{aligned} f_{(\varphi\varphi)}^{(2)} &= 1/4 \{ 4\alpha\beta \sin^2(\theta/2) + [2(\alpha+\beta) \sin(\theta/2) + 1 + \sin^2(\theta/2)] \\ &\quad \times [1 + \sin^2(\theta/2)] \}, \\ f_{(\varphi\varphi)}^{(1)} &= 1/2 (1 - \alpha\beta) \cos^2(\theta/2), & f_{(\varphi\varphi)}^{(0)} &= 1/6 [(1 + \cos^2(\theta/2)) \\ &\quad \times (1 - \alpha\beta + \cos^2(\theta/2)) + 1], \\ f_{(\tau\tau)}^{(2)} &= - [(2\alpha\beta + 1 + \sin^2(\theta/2)) 2 \sin^2(\theta/2) \\ &\quad + (\alpha + \beta) (1 + 3 \sin^2(\theta/2))] \cos^2(\theta/2), \\ f_{(\tau\tau)}^{(1)} &= (\alpha\beta - 1) \sin \theta \cos(\theta/2), & f_{(\tau\tau)}^{(0)} &= -2/3 (\alpha + \beta) \cos^2(\theta/2), \\ f_{(\tau\tau)}^{(2)} &= 2 [1 + \alpha\beta + 2(\alpha + \beta) \sin(\theta/2) + 2 \sin^2(\theta/2)] \cos^4(\theta/2), \\ f_{(\tau\tau)}^{(1)} &= 1/2 (1 - \alpha\beta) \sin^2 \theta, & f_{(\tau\tau)}^{(0)} &= 1/3 (1 + \alpha\beta) \cos^4(\theta/2), \end{aligned}$$

and the remaining functions are determined by the relations

$$\begin{aligned} f_{(\varphi\varphi)}^{(-l)}(\alpha, \beta) &= f_{(\varphi\varphi)}^{(l)}(-\alpha, -\beta), & f_{(\tau\tau)}^{(-l)}(\alpha, \beta) &= -f_{(\tau\tau)}^{(l)}(-\alpha, -\beta), \\ f_{(\tau\tau)}^{(-l)}(\alpha, \beta) &= f_{(\tau\tau)}^{(l)}(-\alpha, -\beta). \end{aligned}$$

Let us investigate in more detail the angular and temperature dependences of the light-scattering intensity. This requires a knowledge in Eqs. (23), (25), and (29) of the correlators as functions of temperature and wave vector. We will restrict the discussion to the case of scattering in the isotropic phase of a CLC near the transition point to the ordered phase. Here, in the Gaussian approximation

$$\langle \xi^{(l)2} \rangle_{\mathbf{q}} = \{ a_0 (T - T^*) + l d q + [b + (c/6) (4 - l^2)] q^2 \}^{-1}, \quad (30)$$

where a_0 , b , c , d , and T^* are constants, and T is the absolute temperature.¹⁴ Near the transition point to the ordered phase the modes with $l = 2$ and $l = -2$ fluctuate the most strongly as functions of the sign of d , i.e., depending on whether the helix after the transition is right- or left-handed. The contributions of these modes strongly depend on the polarizations of the incident and scattered light. Thus, for example, the contribution to the intensity of the mode with $l = 2$, associated with the fluctuations of the dielectric tensor when the incident and scattered waves have right circular polarization, for almost all scattering angles exceeds the intensity for the other polarization by an order of magnitude, and in this case the scattering is preferentially backward.²⁾ To these same modes is due the circular dichroism which is connected with the difference of the extinction coefficients of the right and left circularly polarized waves.^{22,23} A typical angular dependence of the scattering intensity, calculated according to Eq. (29), is shown in Fig. 1. Here the contribution of the first term in braces in Eq. (29) (a) and that of the sum of the second and third terms (b) are shown separately.

The temperature dependence of the scattering intensity is mainly determined by the correlation functions $\langle \xi^{(l)2} \rangle_{\mathbf{q}}$

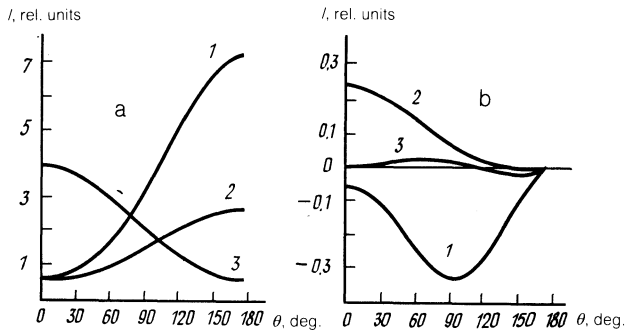


FIG. 1. The sum of the contributions of all modes to the scattered light intensity $I(\alpha, \beta)$ in the isotropic phase of a *dextro*-CLC as a function of scattering angle θ , calculated on the basis of Eq. (20): a) scattering from fluctuations of the dielectric tensor and b) contribution of the gyrotropy fluctuations. 1) $\alpha = 1, \beta = 1$; 2) $\alpha = -1, \beta = -1$; 3) $\alpha = 1, \beta = -1$. Calculation was carried out for the CLC CE2, the parameters of the correlation function $\langle \xi^{(D)2} \rangle_q$, a, b, c, d , and T^* taken from Ref. 27. Wavelength of the light $\lambda = 6238 \text{ \AA}$, temperature difference $T - T^* = 1 \text{ K}$. For ease of visualization the value $kB_{(i)}$ = 0.5 has been deliberately exaggerated.

[Eq. (30)]. We call attention to the fact that far from the phase transition, when the term $a_0(T - T^*)$ in the denominator in expression (30) dominates, the inverse intensity depends linearly on temperature, thus $I^{-1}(\alpha, \beta) \propto (T - T^*)$. However, close to the transition point the terms in Eq. (30) which contain \mathbf{q} become important, and the character of this dependence becomes complicated. If account is taken of the fact that the terms in Eq. (30) that are quadratic in \mathbf{q} are small, then the deviation of the temperature dependence of $I^{-1}(\alpha, \beta)$ from a linear one can serve as an independent method of determining the ratio d/a_0 . Note that here the situation markedly differs from the one that exists in the isotropic phase of a nematic liquid crystal, where $d = 0$, and the inverse intensity depends linearly on temperature in the Gaussian approximation. The experimentally observable deviation from linearity near the transition point is connected with non-Gaussian fluctuational corrections.^{4,24} In principal, similar corrections also exist in CLC's,²⁵ and must be taken into account in any analysis of the experimental data, along with the terms that are linear in q .³⁰

As the phase transition is approached, in addition to the growth of the scattering intensity in the CLC, there is also observed a critical growth of the effective coefficient of gyration.¹⁵ This effect is manifested in an anomalously large rotation of the polarization plane, and has been studied in detail both theoretically²⁶ and experimentally.²⁷ Significant optical activity hinders the description of the scattering in terms of the commonly used linear polarizations, since it is necessary here to take into account the difference in the polarization at different points in the scattering volume. By using circular polarizations such problems are avoided, and the effective coefficient of gyration ν in the expression for the scattering intensity (21) enters only through the quantity $C_{(\alpha)}$ and the vectors $\mathbf{k}_{(\alpha)}$ and $\mathbf{k}_{(\beta)}$. In this case, since the critical growth of ν is limited by virtue of the abrupt change to a first-order phase transition, the actual value of ν throughout the region of existence of the isotropic phase are such that $\nu\omega/c \ll 1$ and the quantity ν can be neglected in $C(\alpha)$, $\mathbf{k}_{(\alpha)}$, and $\mathbf{k}_{(\beta)}$, as was done in Eq. (29).

If we consider the case of spontaneous fluctuations, in

which there is no strongly fluctuating order parameter η , then the correlation radius of the tensors $\hat{\epsilon}$ and $\hat{\gamma}$ is much smaller than the wavelength of the light. In this case the correlation functions entering into Eq. (31) can be calculated at $\mathbf{q} = 0$. In this case only those parts of $\hat{\epsilon}$ and $\hat{\gamma}$ which have identical tensor dimension will correlate. The scattering intensity in this case has the form

$$I(\alpha, \beta) = I_0 C_0 \left[\sum_i (x_{(qq)} f_{(qq)} + k x_{(qr)} f_{(qr)} + k^2 x_{(rr)} f_{(rr)}) + k^2 y_{(\mu\mu)} (f_{(\mu\mu)}^\perp + f_{(\mu\mu)}^\parallel) + z_{(ee)} f_{(ee)} + k z_{(ev)} f_{(ev)} + k^2 z_{(vv)} f_{(vv)} \right],$$

where the scalars $x_{(i)}$, $y_{(i)}$, and $z_{(i)}$ determine the contributions of the correlators of the corresponding quantities.

5. CONCLUSION

We note some peculiar features in the investigation of light scattering in gyrotropic media. First of all, in the presence of significant optical activity plane-polarized wave scattering experiments become ineffective since the measurements results depend to a strong degree on the geometry of the experiment: the path of the transmitted light through the medium, the size of the scattering volume, the transverse dimensions of the rays, etc. This hinders an unambiguous interpretation of the experimental results and lowers the accuracy of determination of the parameters of the medium from the light-scattering data.

The use of circular polarizations has none of these difficulties. At the same time, as follows from the calculations which we have presented here, the determination of the parameters is no more complicated than in the usual analysis of scattering of plane-polarized waves in nongyrotropic media. In this case, instead of the four intensities I_V^V, I_H^V, I_V^H , and I_H^H used in the case of linear polarizations, there here arise the four intensities $I(\pm 1, \pm 1)$. The only notable difference between the scattering in the circular and the linear polarizations is the unusual angular dependence.

Another peculiar feature of scattering in gyrotropic media is the possibility of the manifestation of gyrotropy fluctuations (antisymmetric scattering). Let us discuss the question of the experimental observation of gyrotropy fluctuations. As can be seen from the results of Sec. 4, gyrotropy fluctuations should manifest themselves most clearly in the scattering of light in an intrinsic ferroelectric, where there are no lower order contributions of $\delta\eta$ and $\delta\epsilon$. It should however be mentioned that experimentally such an observation can be hindered by scattering from point defects peculiar to ferroelectric crystals.

Assuming that the fluctuations of all the components of the tensor \hat{f} are of the same order of magnitude, we find that the relative contribution kB/A to the scattering from the gyrotropy fluctuations in Eqs. (23) and (29) is of the order of a/λ . The ratio of a/λ is usually estimated on the basis of the molecular size and ranges between 10^{-3} and 10^{-2} [Ref. 10]. If the gyrotropy has a structural origin, this ratio can be significantly larger. Thus, for example, close to the transition point to the gyrotropic ordered phase there arise in the isotropic phase fluctuational seeds, which have the structure of an asymmetric phase and whose dimensions can significantly exceed the molecular. Therefore, for a linear relation between $\delta\epsilon$ and $\delta\eta$ investigations of light scattering in the isotropic phase of CLC's in the vicinity of the phase transi-

tion to the ordered phase are promising for the detection of gyrotropy fluctuations. In this case, as can be seen from Fig. 1, the most appropriate experimental geometries are those with polarizations $(-1, -1)$ (for *dextro*-CLC's) at small scattering angles or with polarizations $(1, 1)$ at a scattering angle $\sim 90^\circ$.

In conclusion, let us elucidate the physical mechanisms which can give rise to gyrotropy fluctuations. If the medium is gyrotropic, i.e., it contains chiral molecules, then the gyrotropy fluctuations in it can be thought of as regions in which the axes of the gyration tensors of the molecules have a common preferred-orientation direction. Gyrotropy fluctuations can also be manifested in a nongyrotropic medium, for example, in a racemic mixture, where $\langle \gamma \rangle = 0$ and the fluctuations $\delta\gamma$ are nonzero due to local fluctuations of the concentrations of the molecules of the right-handed and left-handed forms. It follows hence, in particular, that the magnitude of the gyrotropy fluctuations $\delta\gamma$ can significantly exceed its mean value $\langle \gamma \rangle$. And finally, gyrotropy fluctuations of structural type are possible, i.e., the formation of asymmetric fluctuational macroscopic regions, etc.

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¹Here and below we omit the superscript (0) of $\delta\hat{\epsilon}^{(0)}(\mathbf{r})$.

²The latter effect is in some sense analogous to the well-known effect²⁸ of reflection of a right-polarized wave from a *dextro*-cholesteric in the ordered phase.

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