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Light scattering in cholesteric liquid crystals

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Light scattering in a cholesteric liquid crystal is investigated. Uniaxial fluctuations of the director are considered with a wave vector that has a small component perpendicular to the axis of the cholesteric crystal. It is shown that the spectrum of the scattered light should reveal maxima at $\mathbf{q} = n\mathbf{t}_0$, $n = 0, \pm 1$, ..., with intensities that decrease with increasing number n.

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We investigate here the scattering of light in cholesteric liquid crystals. The optical properties of liquid crystals were dealt with in many papers. Kats[1] and Belyakov and Dmitrienko[2] investigated the propagation of light in cholesteric crystals. Kats[1] solved Maxwell's equations for the case of normal incidence of light and for incidence at a small angle to the crystal axis. Belyakov and Dmitrienko[2] considered the passage and selective reflection of light from a cholesteric light crystal with the aid of dynamic diffraction theory, and obtained expressions for the amplitudes and polarizations of the reflected and transmitted waves at an arbitrary angle of incidence of the light on the crystal.

De Gennes[3] and Brazovskiĭ and Dmitriev[4] found the correlation function in the high-temperature phase of a cholesteric crystal. The case of the low-temperature phase, when spontaneous spiral anisotropy is present, was considered in the review of Stephen and Straley,[3] who investigated the scattering of light by long-wave fluctuations. They, however, introduced fluctuating rotation angles of the directors and assumed that these angles vary slowly compared with the period of the spiral. It will be shown, however, that the nonzero harmonics of these functions are also not small.

We consider scattering by fluctuations of arbitrary wavelength. In contrast to Stratonovich,[6] who investigated biaxial fluctuations, we obtain results for wavelengths located directly in the vicinities of the maxima of $\mathbf{q} = \pm n\mathbf{t}_0$, where the terms of the series (3.3) of the Stratonovich paper[6] turn out to be large (since the values of the corresponding τ go through zero) and the expansion turns out to be incorrect.

We investigate uniaxial fluctuations of the director $\delta n_x = -\varphi\sin\varphi_0$, $\delta n_y = \varphi\cos\varphi_0$, δn_z, where $\varphi_0 = t_0 z$, and the z axis is directed along the spiral axis; $n_x = \cos\varphi_0$, $n_y = \sin\varphi_0$, $n_z = 0$.

The assumption that the elastic constants are equal leads to the following expression for the change of the thermodynamic potential:

$$\Delta G = \frac{1}{2} K \int_V d\mathbf{r} \left\{ (\nabla\varphi)^2 + 2t_0 \delta n_z \left[\cos\varphi_0 \left(\frac{\partial\varphi}{\partial x}\right) + \sin\varphi_0 \left(\frac{\partial\varphi}{\partial y}\right) \right] + t_0^2 \delta n_z^2 + (\nabla \delta n_z)^2 \right\}$$

or

$$\Delta G = \frac{1}{2} K \left\{ \sum_{\mathbf{q}} q^2 \varphi_q \varphi_q^* + t_0 \sum_{\mathbf{q}} i q_x \varphi_q (\psi_{q+t_0}^* + \psi_{q-t_0}^*) + t_0 \sum_{\mathbf{q}} q_y \varphi_q (\psi_{q+t_0}^* - \psi_{q-t_0}^*) + t_0^2 \sum_{\mathbf{q}} \psi_q \psi_q^* + \sum_{\mathbf{q}} q^2 \psi_q \psi_q^* \right\},$$

where $\varphi_q \equiv \varphi(\mathbf{q})$, $\psi_q \equiv \delta n_z(\mathbf{q})$ are the Fourier components of the spatial quantities $\varphi(\mathbf{r})$ and $\delta n_z(\mathbf{r})$.

To diagonalize this quadratic form we must solve the system of equations

$$\begin{aligned} \frac{\partial \Delta G}{\partial \varphi_q^*} &= q^2 \varphi_q - i t_0 q_x (\psi_{q+t_0} + \psi_{q-t_0}) + t_0 q_y (\psi_{q+t_0} - \psi_{q-t_0}) = \lambda \varphi_q, \\ \frac{\partial \Delta G}{\partial \psi_q^*} &= (q^2 + t_0^2) \psi_q + i t_0 q_x (\varphi_{q+t_0} + \varphi_{q-t_0}) - t_0 q_y (\varphi_{q+t_0} - \varphi_{q-t_0}) = \lambda \psi_q, \end{aligned} \tag{1}$$

which assumes in the case $q_x = 0$ and $q_y = 0$ the simplest form

$$q^2 \varphi_q = \lambda \varphi_q, \qquad (q^2 + t_0^2) \psi_q = \lambda \psi_q.$$

Then $\lambda_1 = q_z^2$, $\lambda_2 = q_z^2 + t_0^2$,

$$\Delta G = \frac{1}{2} K \sum_{\mathbf{q}} \{ (t_0^2 + q^2) |\psi_q|^2 + q^2 |\varphi_q|^2 \},$$

$$\langle |\varphi_q|^2 \rangle = T/K q_z^2, \qquad \langle |\psi_q|^2 \rangle = T/K (q_z^2 + t_0^2).$$

We shall show that at small $q_\perp$, i.e., $q_\perp / t_0 \ll 1$, the solution can be found in the form of an expansion in this small parameter. The zeroth approximation corresponds to the case $q_\perp = 0$.

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We shall henceforth direct, for convenience the y axis along $\mathbf{q}_\perp$; then $q_\perp = q_y$, $q_x = 0$. We determine first a value of λ close to q_z^2 at $q_z/t_0 \ll 1$. Since the components of the eigenvector corresponding to this λ decrease in proportion to powers of q_y, i.e., $\psi(\varphi)_{q+nt_0} \sim q_y^n$, in the first approximation in $(q_y/t_0)^2$ we need retain in the system (1) only φ_q and $\psi_{q\pm t_0}$. We obtain a system of three equations:

$$[(\mathbf{q}-\mathbf{t}_0)^2+t_0^2]\psi_{q-t_0}-t_0q_y\varphi_q=\lambda\psi_{q-t_0},$$
$$q^2\varphi_q+t_0q_y(\psi_{q+t_0}-\psi_{q-t_0})=\lambda\varphi_q,$$
$$[(\mathbf{q}+\mathbf{t}_0)^2+t_0^2]\psi_{q+t_0}+t_0q_y\varphi_q=\lambda\psi_{q+t_0}.$$

The eigenvalue of interest to us

$$\lambda_1^{(0)}=q_z^2-q_z^2q_y^2/t_0^2$$

has a normalized eigenvector

$$\mathbf{e}_1=\begin{pmatrix}x_1\\ y_1\\ z_1\end{pmatrix},\quad x_1^2\approx z_1^2=\frac{q_y^2}{4t_0^2},\quad y_1^2\approx 1.$$

To determine the $\lambda_2^{(0)}$ close to $q_z^2+t_0^2$ at small q_z we proceed in similar fashion, but it is now necessary to retain $\psi_q, \varphi_{q\pm t_0}, \psi_{q\pm 2t_0}$, and we obtain a system of five equations, whose eigenvalue and solution are the following at $q_z = 0$:

$$\lambda_2^{(0)}=q_y^2+t_0^2,$$

$$\mathbf{e}_2=\begin{pmatrix}a_2\\ x_2\\ y_2\\ z_2\\ b_2\end{pmatrix}\leftarrow\begin{pmatrix}\psi_{q-2t_0}\\ \varphi_{q-t_0}\\ \psi_q\\ \varphi_{q+t_0}\\ \psi_{q+2t_0}\end{pmatrix},\quad a_2^2\approx b_2^2\approx\frac{1}{2},\quad x_2^2\approx y_2^2\approx z_2^2\approx\frac{q_y^2}{32t_0^2}.$$

It remains now to note that when $\mathbf{q}$ is replaced in the initial infinite system (1) by $\mathbf{q}\pm n\mathbf{t}_0$, the system remains unchanged. Consequently, under this substitution $\lambda(\mathbf{q})$ goes over into $\lambda(\mathbf{q}\pm n\mathbf{t}_0)$ and from the obtained $\lambda_1^{(0)}$ and $\lambda_2^{(0)}$ at $q_z \approx 0$ we determine also all the remaining $\lambda_1^{(\pm n)}$, $\lambda_2^{(\pm n)}$ at $q_z \pm nt_0 \approx 0$ by replacing $\mathbf{q}$ with $\mathbf{q}\pm n\mathbf{t}_0$ (and the same for the eigenvectors).

In the first approximation in $(q_y/t_0)^2$ we obtain for $\langle|\psi_q|^2\rangle$ an expression that has at $q_z = 0$ and $q_z = \pm 2t_0$ respectively the finite maxima

$$\frac{T}{K(q_y^2+t_0^2)}\frac{q_y^2}{32t_0^2},\quad \frac{T}{K(q_y^2+t_0^2)}\frac{1}{2}$$

and divergences of the form

$$a/(q_z\mp t_0)^2$$

at $q_z \approx \pm t_0$, where $a = Tq_y^2/4Kt_0^2$. For $\langle|\varphi_q|^2\rangle$ we obtain a divergence of the type

$$\frac{b}{q_z^2+q_z^2q_y^2/t_0^2}$$

as $q_z \to 0$, where $b = T/K$.

To find the solution in the next approximations in $(q_y/t_0)^2$, it is necessary to introduce each time into the system two new terms, and correspondingly increase the number of equations in these systems by two. We then obtain ever new terms with maxima at $q_z = \pm nt_0$ and with coefficients $\sim (q_y/t_0)^{2n}$. These coefficients can be very easily determined by retaining in them only the lowest powers of q_y/t_0, since knowledge of λ accurate to $(q_y/t_0)^2$ makes it possible to find its eigenvector with relative accuracy $(q_y/t_0)^2$. Thus, in the next approximation for $\langle|\varphi_q|^2\rangle$ we obtain additional divergences of the form

$$C/(q_z\pm 2t_0)^2$$

at $q_z \approx \mp 2t_0$, where $C = Tq_y^4/64Kt_0^4$, etc.

The result is valid for light scattering with change of the wave vector $\mathbf{q}$. We use the known formula for the differential cross section of light scattering in the interval of frequencies $d\omega$ and solid angles $d\Omega$:

$$\frac{d\sigma}{d\omega\, d\Omega}=\frac{\omega^4}{32\pi^3}\langle\delta\varepsilon_{\alpha\beta}\delta\varepsilon_{\gamma\delta}\rangle p_\alpha p_\gamma p_\beta' p_\delta',$$

where $\delta\varepsilon_{\alpha\beta}$ is the fluctuation of the dielectric constant; p and p' are the polarization vectors of the incident and scattered light, and the speed of light is $c = 1$.

By virtue of symmetry, $\delta\varepsilon_{\alpha\beta}$ is connected with the fluctuation $\delta S_{\alpha\beta}$ of the order parameter by the relation $\delta\varepsilon_{\alpha\beta} = M\delta S_{\alpha\beta}$. As a result we get

$$\frac{d\sigma}{d\omega d\Omega}=\frac{\omega^4M^2}{32\pi^3}\{A[I_\varphi(\mathbf{q}+2\mathbf{t}_0)+I_\varphi(\mathbf{q}-2\mathbf{t}_0)]+B[I_\psi(\mathbf{q}+\mathbf{t}_0)+I_\psi(\mathbf{q}-\mathbf{t}_0)],$$

where

$$I_\varphi(\mathbf{q})=\langle|\varphi_q|^2\rangle,\quad I_\psi(\mathbf{q})=\langle|\psi_q|^2\rangle,$$
$$A={}^1/_4\varepsilon_a^2(p_x^2+p_y^2)(p_x'^2+p_y'^2),\quad B={}^1/_4\varepsilon_a^2[(p_xp_z'+p_x'p_z)^2+(p_yp_z'+p_y'p_z)^2].$$

Thus, in the case of $q_\perp = 0$, depending on the polarization of the incident and scattered light, different maxima will be observed in the spectrum of the scattered light. When the incident and scattered light is polarized in the equatorial plane ($B = 0$), maxima will be observed at $q_z = \pm 2t_0$. In the case of polarization along the axis of the cholesteric crystal, A vanishes and accordingly maxima are observed at $q_z = \pm t_0$. At arbitrary polarization of the incident and scattered light all four maxima will be observed.

At nonzero $q_\perp$, the scattering picture becomes more complicated. Now the maxima in the spectrum of the scattered light will be observed at $q_z = \pm nt_0$, $n = 0, 1, 2, \ldots$, and will decrease with increasing number n. Just as in the case $q_\perp = 0$, the values of the maxima depend on the polarization of the incident and scattered light.

The described approximation provides a clear physical picture of the nature of the light scattered by fluctuations of the dielectric-tensor. It can be used for quantitative description only in situations when the dimensions of the regions that scatter the light coherently are small so that the attenuation of the light due to scattering in the coherently scattering region can be neglected. This situation is realized either in very thin planar samples ($L\delta/p \ll 1$, where L is the sample thickness), or in polycrystalline samples of cholesteric crystals with small individual domains. In a typical experimental situation the parameter $L\delta/p$ is not small compared with unity, so that a more rigorous analysis of the interaction of the light with cholesteric liquid crystals is necessary.

Belyakov and Dmitrienko[2] have shown that in the two-wave approximation of the dynamic theory of diffraction, light waves with wave vectors $\mathbf{k}_0^j$ and $\mathbf{k}_\tau^j = \mathbf{k}_0^j + \tau$ can propagate in a cholesteric crystal:

$$\mathbf{E}_j(\mathbf{r})=\mathbf{e}_j^0\exp[-i\mathbf{k}_0^j\mathbf{X}\mathbf{r}]+b_j\mathbf{e}_j^\tau\exp[-i\mathbf{k}_\tau^j\mathbf{X}\mathbf{r}],$$

where $(j=1, 2, 3, 4)$

$$\mathbf{k}_0^j=\varkappa+\frac{\varkappa\delta(p+m+q_j)(1+m)}{2\sin\theta_B}\frac{\mathbf{z}}{z};$$

$$m=\frac{\cos^2\theta_B}{1+\sin^2\theta_B},\quad q=\frac{\mathbf{k}_\tau^2-\mathbf{k}_0^2}{\delta\varkappa^2(1+\sin^2\theta_B)},\quad p=\frac{2\varkappa^2-k_0^2-k_\tau^2}{\delta\varkappa^2(1+\sin^2\theta_B)}-m.$$

To describe the passage of light through a cholesteric crystal it is necessary to use the solution

$$\mathbf{E}(\mathbf{r})=\sum_{j=1}^4\xi_j\mathbf{E}_j(\mathbf{r})$$

in conjunction with the boundary conditions.

We shall now consider scattering by fluctuations of the tensor of the same waves, propagating in the sample in accordance with Maxwell's equations. We shall use for each wave the results obtained for the scattering of a monochromatic wave by the fluctuations. Thus, each scattered wave yields an aggregate of peaks, and the resultant picture of the scattering with small change of $q_{\perp j}$, where $\mathbf{q}_{\perp j}=\mathbf{k}'-\mathbf{k}_{0j}$, will consist of maxima located in directions corresponding to the wave vectors

$$\mathbf{k}_j'=\mathrm{Re}(\mathbf{k}_{0j})\pm n\tau=\varkappa+\mathrm{Re}\left(\frac{\varkappa\delta(m+p+q_j)(1+m)}{2\sin\theta_B}\frac{\mathbf{z}}{z}\right)\pm n\tau.$$

The relative values of each series of peaks will be determined by the boundary conditions and by the damping of the waves in the sample, i.e.,

$$\mathrm{Im}\frac{\varkappa\delta(p+m+q_j)(1+m)}{2\sin\theta_B}\frac{\mathbf{z}}{z}.$$

The results can be illustrated by considering the case of normal incidence of light.[1] The fundamental solution of Maxwell's equations then takes the form

$$\mathbf{E}=\sum_{j=1,2,3,4}A_+^j\exp\{i[\omega t+(\beta_j+\alpha)z]\}\mathbf{n}_++A_-^j\exp\{i[\omega t+(\beta_j-\alpha)z]\}\mathbf{n}_-,$$

$$\mathbf{n}_\pm=(\mathbf{x}\pm i\mathbf{y})/2^{1/2},\quad \alpha=t_0/2,\quad \beta_j=\pm[k^2+\alpha^2\pm k(4\alpha^2+k^2\delta^2)^{1/2}]^{1/2},$$

$$A_+^j/A_-^j=k^2\delta/[(\beta_j+\alpha)^2-k^2].$$

Assume that a circularly polarized wave is incident on a cholesteric liquid crystal and does not experience selective reflection from the sample:

$$2^{-1/2}E_0(\mathbf{x}\pm i\mathbf{y})\exp[i(\omega t-\mathbf{kr})]$$

(the plus or minus sign depends on the sign of the spirality of the cholesteric liquid crystal). Solving the boundary-value problem, we see that this wave excites in the crystal only a solution at $j=1$ or $j=4$ (depending on the direction from which it is incident on the crystal):

$$\mathbf{E}=A_1^+\mathbf{n}_+\exp\{i[\omega t+(\beta_1+\alpha)z]\}+A_1^-\mathbf{n}_-\exp\{i[\omega t+(\beta_1-\alpha)z]\}.$$

To find the scattering cross section we must determine the quantity[7]

$$\langle G_iG_k^*\rangle=\left\langle\iint E_iE_m^*\alpha_{il}\alpha_{km}dV_1dV_2\right\rangle$$

$$=E_i^+E_m^{+*}\iint\langle\alpha_{il}\alpha_{km}^*\rangle\exp[-i(\beta_1+\alpha)(\mathbf{r}_1-\mathbf{r}_2)]dV_1dV_2$$

$$+E_i^-E_m^{-*}\iint\langle\alpha_{il}\alpha_{km}^*\rangle\exp[-i(\beta_1-\alpha)(\mathbf{r}_1-\mathbf{r}_2)]dV_1dV_2$$

$$+E_i^+E_m^{-*}\iint\langle\alpha_{il}\alpha_{km}^*\rangle\exp[-i\beta_1(\mathbf{r}_1-\mathbf{r}_2)-i\alpha(\mathbf{r}_1+\mathbf{r}_2)]dV_1dV_2$$

$$+E_i^-E_m^{+*}\iint\langle\alpha_{il}\alpha_{km}^*\rangle\exp[i\beta_1(\mathbf{r}_1-\mathbf{r}_2)+i\alpha(\mathbf{r}_1+\mathbf{r}_2)]dV_1dV_2,$$

$$\mathbf{E}^+=A_1^+\mathbf{n}_+,\quad \mathbf{E}^-=A_1^-\mathbf{n}_-.$$

As usual, since the dimensions of the scattering sample are much larger than the pitch of the spiral $2\pi/t_0$, the terms contained in the integrand $\exp[\pm i\alpha(\mathbf{r}_1+\mathbf{r}_2)]$ turn out to be negligibly small. As a result we get

$$\frac{d\sigma}{d\omega d\Omega}=\frac{\omega^4M^2}{32\pi^3}\Bigg\{\left(\frac{A_1^+}{E_0}\right)^2\Big\{a^+[I_\varphi(\beta+\alpha+2t_0)+I_\varphi(\beta+\alpha-2t_0)]$$

$$+b^+[I_\varphi(\beta+\alpha+t_0)+I_\varphi(\beta+\alpha-t_0)]\Big\}$$

$$+\left(\frac{A_1^-}{E_0}\right)^2\Big\{a^-[I_\varphi(\beta-\alpha-2t_0)+I_\varphi(\beta-\alpha+2t_0)]$$

$$+b^-[I_\varphi(\beta-\alpha-t_0)+I_\varphi(\beta-\alpha+t_0)]\Big\}\Bigg\},$$

where

$$a^\pm={}^1/_2\varepsilon_a^2(p_x'^2+p_y'^2),\quad b^\pm={}^1/_2\varepsilon_a^2p_z'^2,\quad \varepsilon_a=\delta.$$

Thus, each of the two waves $\mathbf{E}^+$ and $\mathbf{E}^-$, the superposition of which constitutes the wave $\mathbf{E}$ propagating in the crystal, is independently scattered and produces in the spectrum of the scattered light an aggregate of maxima located at wave-vector values $\mathbf{q}$ that differ from one another by an amount $\pm nt_0, n=0, 1, 2, \ldots$, and whose intensities decrease with increasing number n.

This example illustrates well the statement made above that in a more general situation of oblique light incidence each monochromatic component of the wave propagating in the liquid crystal

$$\mathbf{E}(\mathbf{r})=\sum_{j=1}^4\xi_j\mathbf{E}_j(\mathbf{r})$$

will produce a series of maxima in the spectrum of the scattered light.

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