Orientational effect of a light wave on a cholesteric mesophase

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The orientational effect of a light wave on the initial helical structure of the mesophase is treated theoretically within the framework of the continuum theory of a cholesteric liquid crystal (CLC). The effect with the shortest build-up time corresponds to a variation of the director profile over the period of the CLC with preservation of the pitch of the helix (a record of the space-time bulk lattice). The inverse effect of this perturbation of the orientation by light consists in a change of the effective permittivity tensor. For monochromatic fields, this manifests itself as lattice optical nonlinearity; for biharmonic fields, as stimulated scattering of light. The change of the pitch of the CLC helix under the action of light is calculated. It is shown that this change is caused by a torque that twists the helix and that is transmitted by the spin angular momentum of the radiation reflected with change of sign of the circularity. Gigantic optical nonlinearity, which occurs on change of the liquid-crystal orientation with a nonuniformity scale of the order of the whole thickness of the cell, is discussed. For a cell of CLC with at least one orienting surface, the gigantic optical nonlinearity should not occur.

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1. INTRODUCTION

One of the most important properties of a liquid-crystal (LC) mesophase consists in the anisotropy of the local permittivity tensor at an optical frequency, $\hat{\varepsilon}(\omega) \neq \varepsilon_0 \hat{I}$. In consequence of this, the dipole moment of unit volume, $\mathbf{P} = (4\pi)^{-1}(\hat{\varepsilon} - \hat{1})\mathbf{E}_b$, induced in the medium by the light field

$$\mathbf{E}_b = 0.5(\mathbf{E}e^{-i\omega t} + \mathbf{E}^* e^{i\omega t}), \tag{1}$$

is not collinear with the field $\mathbf{E}$. As a result, unit volume of the LC is subject to a torque $\mathbf{M} = [\mathbf{P} \times \mathbf{E}]$ that does not vanish on averaging over time and that is proportional to the intensity of the light. Nonlinear optical effects caused by reorientation of the LC director under the action of this torque were recently predicted theoretically[1-7] and detected experimentally.[6,8] At present there is a burgeoning grown in the number of papers, both theoretical and experimental, devoted to orientational optical nonlinearity of LC. The present paper discusses and calculates in detail various effects of the action of light fields on the cholesteric liquid crystal (CLC) mesophase.

2. EXPRESSION FOR THE FREE ENERGY AND THE EQUATIONS FOR THE DIRECTOR AND THE FIELD

We shall describe the state of the CLC by the director unit vector $\mathbf{n}(\mathbf{r})$. For the unperturbed helical structure (the Grandjean texture), the $\mathbf{n}(\mathbf{r})$ relation has the form

$$\mathbf{n}(\mathbf{r}) = \mathbf{e}_x \cos q_0 z + \mathbf{e}_y \sin q_0 z. \tag{2}$$

We shall take the free energy (or the Lagrangian function taken with a minus sign) of unit volume in the form

$$F = \frac{1}{2}[K_{11}(\operatorname{div}\mathbf{n})^2 + K_{22}(\mathbf{n}\operatorname{rot}\mathbf{n} + q_0)^2 + K_{33}[\mathbf{n}\times\operatorname{rot}\mathbf{n}]^2]$$
$$-\left[\frac{\varepsilon_\perp}{16\pi}(\mathbf{E}\mathbf{E}^*) + \frac{\varepsilon_a}{16\pi}(\mathbf{n}\mathbf{E})(\mathbf{n}\mathbf{E}^*)\right] + \frac{c^2}{16\pi\omega^2}\left[\frac{\partial E_i}{\partial x_k}\frac{\partial E_i^*}{\partial x_k} - \frac{\partial E_i}{\partial x_k}\frac{\partial E_k^*}{\partial x}\right]. \tag{3}$$

The first group of terms in (3) is due to the distortion of the CLC structure. It is not difficult to show that in general this group of terms is nonnegative and that the structure $\mathbf{n}(\mathbf{r})$ of (2) gives its absolute minimum, equal to zero. The second term describes the energy of interaction of the CLC with the light field. The permittivity tensor at the frequency of the light is taken in the form

$$\varepsilon_{ik}(\omega, \mathbf{r}) = \varepsilon_\perp(\omega)\delta_{ik} + \varepsilon_a(\omega) n_i(\mathbf{r}) n_k(\mathbf{r}), \tag{4}$$

where $\varepsilon_a(\omega) = \varepsilon_\parallel(\omega) - \varepsilon_\perp(\omega)$ is optical anisotropy.

To calculate build-up processes, we need also an expression for the dissipative function R ($\mathrm{erg\,cm^{-3}\,s^{-1}}$), which we shall take in the form

$$R = 0{,}5\gamma(\partial\mathbf{n}/\partial t)^2, \tag{5}$$

where the relaxation constant γ has the dimension poise. The director vector $\mathbf{n}(\mathbf{r}, t)$ obeys the variational equations

$$\Pi_{ik}\left[\frac{\delta F}{\delta n_k} - \frac{\partial}{\partial x_l}\frac{\delta F}{\delta(\partial n_k/\partial x_l)}\right] = -\frac{\delta R}{\delta \dot{n}_i}, \quad \Pi_{ik} = \delta_{ik} - n_i n_k. \tag{6}$$

The operator Π_{ik} is projected on the plane perpendicular to the local direction of the director $\mathbf{n}(\mathbf{r}, t)$; this guarantees satisfaction of the equality $|\mathbf{n}(\mathbf{r}, t)| = 1$. With allowance for the perturbation of the structure $\mathbf{n}(\mathbf{r})$, the field $\mathbf{E}(\mathbf{r})$ may change its value as compared with the original wave. But in the variation of F in (3) and (6), we must suppose that what is given is the electric field of the light wave $\mathbf{E}(\mathbf{r})$ (and not, for example, the induction $\mathbf{D} = \hat{\varepsilon}\mathbf{E}$).

Variation of (3) with respect to the components $E_i(\mathbf{r})$ at fixed $\mathbf{n}(\mathbf{r})$ gives the vectorial variational equation

$$\operatorname{rot}\operatorname{rot}\mathbf{E} - \omega^2 c^{-2}\hat{\varepsilon}\mathbf{E} = 0, \tag{7}$$

which agrees exactly with the consequence of Maxwell's equations for a monochromatic field $\mathbf{E}(\mathbf{r})$. The specific form of writing of equations (5) and (7) depends on the problem being considered (see below).

3. EFFECTS DUE TO DISTORTION OF THE PROFILE OF THE CLC HELIX

For typical CLC, the pitch $h = 2\pi/q_0$ of the helix amounts to $h \sim 10^{-4}$ cm, and the value of ε_a lies in the in-

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terval from +0.03 to +0.3. Far from the Bragg resonance and far from the Mauguin limit,[9,10] the change of the state of polarization of a light wave over the length of the pitch h is small. Therefore we shall in this section calculate the distortion of the CLC structure under the action of light waves by considering, in a first approximation, one or several plane light waves with a given direction of propagation and polarization unit vector.[1] We shall consider the actual change of the state of polarization, the overall phase, and the amplitude of the waves in the next stage, with a more accurate solution of Maxwell's equations.

Furthermore, in this section we shall consider processes with the shortest build-up time, in which the orientation of the axis of the helix and the value of the pitch remain as before throughout the whole cell, and all that changes is the specific behavior of the director within the limits of a period.

3a. Lattice optical nonlinearity (LON) and nonlinear birefringence

We assume that a single plane monochromatic wave $\mathbf{E}(z)=\mathbf{E}\exp(ikz)$ is propagated along the CLC in the direction of the axis of the helix, and by virtue of the transversality condition $(\mathbf{e}_z\cdot\mathbf{E})=0$. Then the tensor $E_iE_k^*$ that occurs in $F_E=-\varepsilon_a(\mathbf{n}\cdot\mathbf{E})(\mathbf{n}\cdot\mathbf{E}^*)/16\pi$ may be considered practically constant in space. We shall seek the perturbed distribution of the director in the form

$$\mathbf{n}(z,t)=\mathbf{n}_0(z)+[\mathbf{e}_z\times\mathbf{n}_0(z)]\alpha(z,t), \tag{8}$$

where $\alpha\ll1$ is a small distortion of the phase of the helix. Then the variational Eqs. (6) give

$$\gamma\frac{\partial\alpha}{\partial t}-K_{22}\frac{\partial^2\alpha}{\partial z^2}=\frac{\varepsilon_a}{16\pi}\{\sin 2q_0z(|E_y|^2-|E_x|^2)+\cos 2q_0z(E_xE_y^*+E_x^*E_y)\}. \tag{9}$$

It is natural to write the solution of Eq. (9) as

$$\alpha(z,t)=\alpha_c(t)\cos 2q_0z+\alpha_s(t)\sin 2q_0z, \tag{10}$$

$$\frac{d\alpha_c}{dt}+\Gamma\alpha_c(t)=\frac{\varepsilon_a}{16\pi\gamma}(E_xE_y^*+E_x^*E_y), \tag{11a}$$

$$\frac{d\alpha_s}{dt}+\Gamma\alpha_s(t)=\frac{\varepsilon_a}{16\pi\gamma}(|E_y|^2-|E_x|^2). \tag{11b}$$

In (11) we have introduced a constant $\Gamma(\mathrm{s}^{-1})$ that characterizes the rate of establishment of the lattice (10) of the perturbed director:

$$\Gamma=K_{22}q_0^2/\gamma. \tag{11c}$$

In the stationary mode,

$$\alpha_c=\frac{\varepsilon_a}{16\pi K_{22}q_0^2}(E_xE_y^*+E_x^*E_y),\quad \alpha_s=\frac{\varepsilon_a}{16\pi K_{22}q_0^2}(|E_y|^2-|E_x|^2). \tag{12}$$

As is seen from this expression, for circular polarization of the light $\alpha_s=\alpha_c=0$.

It is evident from formulas (10)–(12) that the resultant nonlinearity is due to those perturbations of the director that form a "lattice" $\alpha(z)$ of the form (10) with a small spatial period $2\pi/q_0$. We shall call nonlinearity of this type lattice optical nonlinearity (LON). A characteristic feature of LON consists in the fact that its constant is proportional to $(K_{ii}q_0^2)^{-1}$, where q_0 is the wave vector of the lattice. For nematics, LON was considered in Ref. 3.

As a result of the reorientation, there appears in the dielectric permittivity tensor of the medium a perturbation of the form $\delta\varepsilon_{ik}(z)=\varepsilon_a(n_{0i}\delta n_k+n_{0k}\delta n_i)$, in which there are rapidly varying terms $\sim\cos 4q_0z$ and $\sin 4q_0z$ and also terms $\delta\bar{\varepsilon}$ that are constant in space. Retaining only these latter, we get

$$\delta\varepsilon_{ik}={}^1/_2\varepsilon_2[E_i^*E_k+E_iE_k^*-E_l^*E_l\delta_{ik}^{(2)}],\quad \varepsilon_2=\varepsilon_a^2/16\pi K_{22}(2q_0)^2 \tag{13}$$

and $\delta_{ik}^{(2)}=\delta_{ik}-(\mathbf{e}_z)_i(\mathbf{e}_z)_k$ is the two-dimensional Kronecker δ symbol. Thus a strong linearly polarized wave $\mathbf{E}=\mathbf{e}_xE$ produces in the medium a birefringence $\delta\bar{\varepsilon}_{xx}=0.5\varepsilon_2|E|^2$, $\delta\bar{\varepsilon}_{yy}=-0.5\varepsilon_2|E|^2$. The presence of such birefringence can be recorded by means of a weak auxiliary wave, in general of an altogether different frequency and also, perhaps, of opposite direction. But if we are interested in the effects of self-interaction of a strong wave, then

$$(\delta\mathbf{D})_i=\delta\varepsilon_{ik}E_k={}^1/_2\varepsilon_2(\mathbf{E}^*)_i(\mathbf{E}\mathbf{E}). \tag{14}$$

Here we mention the following. Because of the rapid periodic nonuniformity of the CLC, it is the "lattice" perturbations (10) of the director that give nonvanishing contributions to the space-averaging tensor $\delta\hat{\varepsilon}$. We shall estimate the orders of magnitude of ε_2 and Γ. Let $n_e-n_0=0.06$; then $\varepsilon_a\approx 2n\Delta n\approx 0.2$. Taking $K_{22}=5\cdot10^{-7}$ dyn, the pitch $h=10^{-4}$ cm, $q=2\pi/h\approx 6\cdot10^4$ cm^{-1}, we get the estimate $\varepsilon_2\approx 0.4\cdot10^{-6}$ cm^3/erg. For light wavelength $\lambda_{\mathrm{vac}}=2\pi c/\omega$ the nonlinear advance of phase is

$$\delta\varphi=\frac{\omega}{4cn}\varepsilon_2|E|^2L=APL, \tag{15}$$

where P is the power density (W/cm^2), L is the thickness of the layer (cm), and the constant A has the dimensions cm/W. This constant is proportional to $\lambda_{\mathrm{vac}}^{-1}$; and for $\lambda_{\mathrm{vac}}=0.5$ μm, we have $A=5\cdot10^{-5}$ cm/W. The build-up time is $\tau\sim\Gamma^{-1}\approx 5\cdot10^{-4}$ s for $\gamma\approx 1$ P. The nonlinearity (13)–(14) has very remarkable polarization properties: it is greatest for linearly polarized light and vanishes for both circular polarizations, right and left. Furthermore, the nonlinearity (14) leads to an effect well known in nonlinear optics,[11] self-rotation of the polarization ellipse. In fact, if

$$\mathbf{E}=E[\cos\beta(\mathbf{e}_x\cos\psi+\mathbf{e}_y\sin\psi)+i\sin\beta(\mathbf{e}_y\cos\psi-\mathbf{e}_x\sin\psi)],$$

then the angle ψ varies according to the law

$$d\psi/dz=AP\sin 2\beta. \tag{16}$$

Experimental study of nonlinearity of the form (14) can be carried out either on the basis of the effect of self-focusing of light [the advance of phase (15)] or on the basis of the self-rotation of the polarization ellipse (16). Furthermore, distortions of the CLC spiral of the form (8), (10) should lead to the appearance of resonance Bragg reflection of the second order for normal incidence of the exploratory light wave.

Still another interesting feature consists in the fact that in the approximation considered, all effects of the nonlinearity (13)–(14) are identical for right and left CLC.

3b. Stimulated forward scattering

We suppose that two waves are propagated in the positive direction of the z axis: a strong one $\mathbf{E}_1\exp(-i\omega_1t)$

and a weak one $\mathbf{E}_2 \exp(-i\omega_2 t)$. First of all, the strong wave $\mathbf{E}_1$ produces for itself a nonlinear response of the form (14). Furthermore, the weak wave $\mathbf{E}_2$ is propagated in the medium with the birefringence (13) produced by the strong wave $\mathbf{E}_1$. But when $\Delta\omega = \omega_1 - \omega_2 \neq 0$, there is still another specific effect, corresponding to a process of stimulated scattering (SS). The point is that the interference term $\propto E_{1i}^* E_{2k}$ in F_E leads to the appearance of perturbations δn of the director and $\delta\varepsilon \propto \delta n$ of the permittivity tensor, of the form $\delta\varepsilon \propto E_1^* E_2$. Scattering of the strong field by these perturbations gives an addition to the induction at frequency ω_2, $\delta D_2 = \text{const}\,|E_1|^2 E_2 \exp(-i\omega_2 t)$. As a result, we can speak of still another, interference mechanism of correction to $\varepsilon(\omega_2)$ because of the field $\mathbf{E}_1$. It is easy to understand that these additional, interference terms in $\delta\varepsilon_{ik}$ are obtained from the expression (13) by the substitutions $\mathbf{E} \to \mathbf{E}_2$, $\mathbf{E}^* \to \mathbf{E}_1$, $\varepsilon_2 \to \varepsilon_2(1 + i\Delta\omega/\Gamma)^{-1}$. As a result we have

$$\delta\mathbf{D}_2 = \frac{1}{2}\varepsilon_2 \frac{1 - i\Delta\omega/\Gamma}{1 + (\Delta\omega/\Gamma)^2}\{\mathbf{E}_2(\mathbf{E}_1\mathbf{E}_1^*) + \mathbf{E}_1^*(\mathbf{E}_1\mathbf{E}_2) - \mathbf{E}_1(\mathbf{E}_1^*\mathbf{E}_2)\}. \quad (17)$$

If $\Delta\omega = \omega_1 - \omega_2 > 0$, i.e., if the weak wave $\mathbf{E}_2$ is shifted in frequency into the Stokes range with respect to the strong one $\mathbf{E}_1$, then the effective permittivity at frequency ω_2 acquires a negative imaginary part. In other words, scattering of the wave $\mathbf{E}_1$ by the interference perturbation leads to an exponential enhancement of the wave $\mathbf{E}_2$. The maximum of this enhancement is attained when $\Delta\omega = \Gamma$. As regards the polarization features, the maximum of the enhancement will be for circular polarizations of opposite signs of rotation; for example, $\mathbf{e}_1 = (\mathbf{e}_x + i\mathbf{e}_y)/\sqrt{2}$, $\mathbf{e}_2 = (\mathbf{e}_x - i\mathbf{e}_y)/\sqrt{2}$. This absolute maximum corresponds to gain g (cm^{-1}, with respect to the intensity)

$$g = GP; \quad G = 4\pi^2\varepsilon_2/\lambda c\bar{\varepsilon} = A. \quad (18)$$

The gain for two linear polarizations is twice as small; and, interestingly, it does not depend on the mutual orientation of the polarizations of the pump $\mathbf{E}_1$ and of the signal $\mathbf{E}_2$.

We note that in the present treatment we have not taken into account the effect of terms $\propto E_1 E_2^*$, corresponding to so-called four-wave parametric interaction.[12]

3c. Stimulated backward scattering

Let a wave $\mathbf{E}_1 \exp(ikz - i\omega_1 t)$ be propagated in the positive direction of the z axis, and a wave $\mathbf{E}_2 \exp(-ikz - i\omega_2 t)$ in the negative. As before, the strong wave $\mathbf{E}_1$ produces birefringence of the form (13), which affects both its own propagation and the propagation of the wave $\mathbf{E}_2$, and this effect is no different from the case of propagation in the same direction. But the interference process that causes backward SS has peculiarities as compared with the case of forward SS.

To calculate backward SS, we must write the perturbation $\alpha(z,t)$ in the form

$$\alpha(z,t) = \alpha_-(t)e^{-2i(k-q_0)z} + \alpha_+(t)e^{-2i(k+q_0)z}. \quad (19a)$$

It is found that

$$\alpha_\pm(t) \propto \frac{e^{i\Delta\omega t}E_{1i}^* E_{2k}}{1 + i\Delta\omega/\Gamma_\pm}, \quad \Gamma_\pm = \frac{K_{22}(2q_0 \pm 2k)^2}{\gamma}. \quad (19b)$$

Scattering of the strong wave $E_1 \exp(ikz - i\omega_1 t)$ by the perturbations $\delta\varepsilon \propto \alpha_+$, α_- gives terms in the induction with spatial variation $\propto \exp(-ikz - 4iq_0 z)$, $\exp(-ikz + 4iq_0 z)$ and finally of the form $\exp(-ikz)$. Retaining only terms of the last type (because only they satisfy the wave equation), after calculations that are straightforward in principle but extremely cumbersome, we get for the interference contribution to $\delta\mathbf{D}_2$

$$\delta\mathbf{D}_2 = \frac{1}{4}\left(\frac{\varepsilon_2^{(-)}}{1 + i\Delta\omega/\Gamma_-} + \frac{\varepsilon_2^{(+)}}{1 + i\Delta\omega/\Gamma_+}\right)\{\mathbf{E}_2(\mathbf{E}_1\mathbf{E}_1^*) \quad (20a)$$

$$+ \mathbf{E}_1^*(\mathbf{E}_1\mathbf{E}_2) - \mathbf{E}_1(\mathbf{E}_1^*\mathbf{E}_2)\} + \frac{i}{4}\left(\frac{\varepsilon_2^{(-)}}{1 + i\Delta\omega/\Gamma_-} - \frac{\varepsilon_2^{(+)}}{1 + i\Delta\omega/\Gamma_+}\right) \quad (20b)$$

$$\times\{[\mathbf{e}_z \times \mathbf{E}_2](\mathbf{E}_1\mathbf{E}_1^*) + [\mathbf{e}_z \times \mathbf{E}_1^*](\mathbf{E}_1\mathbf{E}_2) - [\mathbf{e}_z \times \mathbf{E}_1](\mathbf{E}_1^*\mathbf{E}_2)\},$$

$$\varepsilon_2^\pm = \varepsilon_a^2/16\pi K_{22}(2q_0 \pm 2k)^2.$$

We shall discuss the structure of these expressions, When $q \gg k$, even the interference picture of the fields, $E_1^* E_2 \exp(2ikz)$, may be considered constant within the limits of a single period of the helix. In this case the properties of forward and backward SS are the same, as is easily verified by direct comparison of formulas (13), (17), and (20a), (20b) when $q_0 \gg k$, since $\varepsilon_2^{(+)} \approx \varepsilon_2^{(-)} \approx \varepsilon_2$.

In the other limiting case $k \gg q_0$ (but such as not to enter the Mauguin regime), the process of backward SS again has the same polarization structure as for forward SS, and only the nonlinearity constant and the buildup time are different; they are obtained from the formula for forward SS by the simple change $K_{22}(2q)^2 \to K_{22}(2k)^2$. As in Sections 3a and 3b, both these cases ($q_0 \gg k$ and $q_0 \ll k$) give results independent of the sign the CLC helix.

In the intermediate case $q_0 \sim k$, there are two constants $\Gamma_\pm = K_{22}(2k \pm 2q_0)^2/\gamma$, and the polarization properties are determined by the expression (20), which is conveniently written in circular unit vectors

$$\mathbf{E} = \mathbf{e}_R E_R^* + \mathbf{e}_L E_L, \quad \mathbf{e}_R = 2^{-1/2}(\mathbf{e}_x + i\mathbf{e}_y), \quad \mathbf{e}_L = 2^{-1/2}(\mathbf{e}_x - i\mathbf{e}_y); \quad (21)$$

$$\delta\mathbf{D}_2 = \frac{\varepsilon^{(-)}}{2(1 + i\Delta\omega/\Gamma_-)}\mathbf{e}_R E_{2R} 2|E_{1L}|^2 + \frac{\varepsilon_2^{(+)}}{2(1 + i\Delta\omega/\Gamma_+)}\mathbf{e}_L E_{2L} 2|E_{1R}|^2. \quad (20c)$$

Since the waves $\mathbf{E}_1$ and $\mathbf{E}_2$ are propagated in opposite directions, we may say that in backward SS the clockwise-polarized pumping wave (R) enhances only the clockwise-polarized Stokes wave (L) (and correspondingly for counterclockwise polarizations), each with its own enhancement coefficient. For forward SS, a given circular polarization of the pump (R) enhances the opposite (L) Stokes polarization of, and in this case the two enhancement coefficients coincide. Such coincidence occurs also for backward SS in the cases $q_0 \gg k$ and $q_0 \ll k$.

Finally, in the vicinity of resonance, $q_0 \approx k$, it is the term proportional to $\varepsilon_2^{(-)}$ that has the largest buildup time, and with it the largest enhancement coefficient.

Here, however, it is assumed that we are not so close to resonance that it is necessary to take into account the actual change of field because of Bragg reflection. The effects of a field change because of virtual reflection are considered in the next section.

We may say that when $q_0 \approx k$, the interference term of the two fields, $E_1 E_2^* \exp(2ikz)$, enters into spatial resonance with the helical structure of the cholesteric. The asymmetry between $\varepsilon_2^{(+)}$ and $\varepsilon_2^{(-)}$ is determined by the sign of q_0, i.e. by the sign of the CLC helix. Furthermore, for $\Delta\omega \to 0$ and near Bragg resonance, $q_0 \sim k$, the interference term $\delta \mathbf{D}_2$ of (20) should lead to an additional rotation of the plane of polarization of the wave $\mathbf{E}_2$, proportional to the intensity of the opposite wave $\mathbf{E}_1$ (nonlinear gyrotropy[13]).

Numerical estimates for backward SS when $k \gg q$ give $A_{\rm back} = (q/k)^2 A_{\rm forw}$, $\Gamma_{\rm back} = (k/q)^2 \Gamma_{\rm forw}$: we leave to the reader the substitution of numerical values. Furthermore, when $k \ll q$ we have $A_{\rm back} = A_{\rm forw}$, $\Gamma_{\rm back} = \Gamma_{\rm forw}$.

4. CHANGE OF THE PITCH OF THE SPIRAL UNDER THE ACTION OF A LIGHT FIELD

For consideration of the effects of Sections 3a to 3c, it was sufficient to determine the oscillatory (in its dependence on z) component of the torque $K_{22}(d\theta/dz - q_0)$ (dyn $\cdot$ cm^{-1}) transmitted across unit area normal to the z axis. For the problem of the change of pitch of the helix, it is necessary to know the value averaged over space of the specific torque. For its determination, it is convenient to use a well-known Noether theorem.[14] Namely, for a problem in which both the director $\mathbf{n}(\mathbf{r})$ and the field $\mathbf{E}(\mathbf{r})$ depend only on z, there is symmetry of the Lagrangian (3) with respect to rotation through an arbitrary angle about the z axis. As a result, this stationary system of variational equations has the integral

$$dJ(z)/dz = 0,$$

where

$$J(z) = -K_{22}\left(\frac{d\theta}{dz} - q_0\right) + \frac{c^2}{16\pi\omega^2} e_{ik}\left(E_k \frac{\partial E_i^*}{\partial z} + E_k^* \frac{\partial E_i}{\partial z}\right) \qquad (22)$$

is the torque transmitted across 1 cm^2 (dyn $\cdot$ cm^{-1}). Here $e_{ik} \equiv e_{ikl}(\mathbf{e}_z)_l$ is the unit two-dimensional antisymmetric tensor, and $\mathbf{E} = \mathbf{e}_x E_x(z) + \mathbf{e}_y E_y(z)$. It is easy to demonstrate the validity of the relation $J(z) = \text{const}$ by direct differentiation of (22) and substitution in the result of the stationary $d^2\theta/dz$ from (6) and $d^2\mathbf{E}/dz^2$ from (7). Outside the liquid crystal, i.e. in any other transparent dielectric or in vacuum, the term $\propto K_{22}$ of course is absent. The expression (22) corresponds to transmission of a spin moment $+\hbar$ or $-\hbar$ by each quantum with energy $\hbar\omega$ with clockwise or counterclockwise circular polarization, respectively. In the dielectric outside the CLC let there be a light flux $P^{(z)} = P_+^{(z)} + P_-^{(z)}$ (erg/cm$^2 \cdot$ s), in the positive direction of the z axis, consisting of clockwise-polarized (+) and counterclockwise-polarized (−) waves, i.e., respectively $(\mathbf{e}_x + i\mathbf{e}_y) \times \exp(ikz)$ and $(\mathbf{e}_x - i\mathbf{e}_y)\exp(ikz)$. Let the same separation $P^{(-z)} = P_+^{(-z)} + P_-^{(z)}$, into waves of the form $(\mathbf{e}_x + i\mathbf{e}_y)\exp(-ikz)$ and $(\mathbf{e}_x - i\mathbf{e}_y)\exp(-ikz)$, be made for light traveling in the direction $(-z)$. Then direct calculation with (22) shows that

$$J = \omega^{-1}(P_+^{(z)} - P_-^{(z)} - P_+^{(-z)} + P_-^{(-z)}), \qquad (23)$$

the constancy of $I(z)$ ensures the constancy of each of the quantities $P_i^{(k)}$.

Very important is the question of the behavior of the integral J on passage across the CLC boundary from other media or from vacuum (air). If the surface holds the angle θ of the director $\mathbf{n}$ rigidly in the (x, y) plane, then the axial symmetry is lost, and nothing can be said about the behavior on passage across the boundary. In other words, a rigid boundary can receive or give up an arbitrary torque. But if the angle θ is not restrained by the boundary (for example, by a CLC-air boundary), then it is natural to suppose that the value of the transmitted specific torque $J(z)$ does not change on passage across the boundary. Thus it follows from (22) that

$$\frac{d\theta}{dz} = q_0 + \frac{c^2}{16\pi K_{22}\omega^2} e_{ik}\left(E_k \frac{\partial E_i^*}{\partial z} + E_k^* \frac{\partial E_i}{\partial z}\right) - \frac{J_0}{K_{22}}, \qquad (24)$$

where the value of J_0 can be calculated from (23) outside the CLC on the side of the cell where boundary is free.

Thus we arrive at the important conclusion that the change of the pitch of the CLC helix in a light field is determined not only by the field $\mathbf{E}(z)$ locally present in the medium [the term $\propto EE^*$ in (24)], but also by the process of reflection of fields of different circularities from the CLC helix [the term J_0 in (24), expressed in terms of (23)]. With respect to calculation of the torque due to actually reflected waves, this approach differs both from the approach of Dmitriev[15] and from the approach that we followed in the original version of the present work. The key feature in obtaining this new derivation was the use of a single Lagrangian for the derivation both of Maxwell's equations and of the equations for the dielectric.

The actual field distribution in a CLC, especially near Bragg resonance, has a quite complicated form.[16] Here, therefore, we shall consider only the case when the effect $|\Delta q/q|$ has its largest value. Specifically, let the wave incident on the CLC be circularly polarized, with such a sign of the rotation that total reflection of it occurs at a thickness Δz much less than the cell thickness L. We must further consider two possibilities.

In the first of these, this wave enters the CLC from a surface that rigidly maintains the angle θ. In this case the other (free) surface transmits no torque, radiation across it does not occur, $J_0 = 0, \mathbf{E} = 0$, and as a result it follows from (24) that in the greater part of the cell volume, the pitch of the spiral remains unchanged.

The second possibility corresponds to passage of reflected light through a free surface. If $P^{(+z)} = P_+^{(-z)} = P_{\rm inc}$, i.e., 100% reflection occurs from a spiral with $q_0 > 0$, then $J_0 = -2\omega^{-1} P_{\rm inc}$, and in the main volume of the cell, to which the field does not penetrate,

$$d\theta/dz = q_0 + 2P_{\rm inc}/\omega K_{22}. \qquad (25)$$

This means that the torque transmitted by the reflected field additionally twists the helix, i.e., decreases its pitch.

Of course limitation of the beam with respect to a transverse coordinate may play a very important role in the experiment; because of it, the part of the CLC that is located outside the beam will tend to preserve the previous behavior of the helix.

For numerical estimates, we take $\omega=3\cdot10^{15}\ s^{-1}$ (this corresponds to wavelength in vacuum $\lambda=0.6\ \mu m$), $K_{22}\sim10^{-7}$ dyn. Then for a power density of the wave $P_{inc}=1\ kW/cm^2$, we get from (25) $\Delta q=0.6\cdot10^2\ cm^{-1}$ and $\Delta\theta=\Delta qL\approx0.6$ rad for a cell thickness $L=10^{-2}$ cm.

The change considered above in the pitch of the CLC helix should manifest itself in a whole series of optical effects. As is well known, the rotation of the plane of polarization in a CLC at normal incidence depends substantially on the relation between the pitch $h=2\pi/q$ of the helix and the wavelength of the light.

Exact solutions of Maxwell's equations for normal incidence of a wave on a CLC have been obtained.[17] For consideration of the effects of rotation of the plane of polarization and of the change of phase velocity in a strictly sinusoidal CLC helix of the form (2), it is sufficient to iterate Maxwell's equations twice with respect to powers of ε_a. Then for slowly varying amplitudes $R(z)$ and $L(z)$ we have

$$\frac{dR}{dz}=i\left(\delta\bar{k}-\frac{d\psi}{dz}\right)R,\quad \frac{dL}{dz}=i\left(\delta\bar{k}+\frac{d\psi}{dz}\right)L, \tag{26a}$$

$$\delta\bar{k}=\frac{\varepsilon_a^2}{32}\left(\frac{\omega}{c}\right)^3\varepsilon^{-1/2}\frac{1}{q^2-k^2},\quad \frac{d\psi}{dz}=\frac{\varepsilon_a^2}{32}\left(\frac{\omega}{c}\right)^4\frac{1}{q(q^2-k^2)}. \tag{26b}$$

Here $\delta\bar{k}$ is the correction to the mean of the wave vector over the two polarizations, and $d\psi/dz$ is the specific rotation of the plane of polarization. We denote by φ_p and ψ_p the mean phase and angle of rotation of the polarization, respectively, of an exploratory beam with frequency ω_p. Then because of the change of pitch Δq of the spiral over the layer thickness L, we get

$$\begin{aligned}\Delta\varphi&=-\frac{\varepsilon_a^2}{32}\left(\frac{\omega_p}{c}\right)^3\varepsilon^{-1/2}\frac{2q\Delta q}{(q^2-k_p^2)^2}L,\\ \Delta\psi&=-\frac{\varepsilon_a^2}{32}\left(\frac{\omega_p}{c}\right)^4\frac{3q^2-k_p^2}{(q^2-k_p^2)^2q^2}\Delta qL.\end{aligned} \tag{27}$$

The quantity $\Delta\psi$ of (34) corresponds to nonlinear rotation of the plane of polarization of the light. In the case when the powerful beam is itself also the exploratory beam, the quantities $\Delta\psi\propto|E|^2$ and $\Delta\varphi\propto|E|^2$ near Bragg resonance are proportional to $(q-k)^{-2}$. For experimental observation of the nonlinear rotation $\Delta\psi$, it is most convenient to use linearly polarized incident light, for which the effect of the self-rotation (16) on the induced birefringence (13) is absent. On the contrary, the self-focusing effect ($\Delta\varphi$) of the change of pitch is more conveniently investigated with circularly polarized incident light, when the birefringence (13) is altogether absent. Furthermore, the change of pitch of the helix can be recorded on the basis of the shift of the wavelength of resonance reflection.

5. ABSENCE OF GIGANTIC OPTICAL NONLINEARITY IN CLC

In all the cases considered, the spatial scale of the perturbations of the director corresponded to the pitch of the helix, $l\sim q_0^{-1}$, or to an even smaller quantity [for example, $l\sim(2k)^{-1}$]. For this reason, the dimensionless amount of the perturbation was determined by the parameter $\varepsilon_a|E|^2/K_{22}l^{-2}$, where $l\lesssim q_0^{-1}$. But for liquid crystals perturbations are in principle possible whose inhomogeneity scale corresponds to the whole thickness L of the cell. Since L can take quite large values, $l\sim10^{-2}$ cm, the corresponding nonlinearity has a value larger by 8 to 10 orders of magnitude than the optical nonlinearity of carbon disulfide (CS_2). Such nonlinearity was predicted and observed[2,6] for nematics and received the name "gigantic optical nonlinearity" (GON).

The same GON should exist also for smectics C.[4] As regards smectics A and cholesterics, however, for a cell with a rigidly prescribed orientation on at least one of the surfaces the GON should be absent. We shall illustrate this statement for CLC as an example.

We introduce a unit vector $\mathbf{l}$ along the axis of the cholesteric spiral. The three-dimensional permittivity tensor has the form $\varepsilon_{ik}=\varepsilon_\perp\delta_{ik}+\varepsilon_a n_i n_k$. If we average this expression over the harmonic oscillations of $\mathbf{n}(\mathbf{r})$ (i.e., over the period of the spiral), then we get for $\bar{\varepsilon}_{ik}$

$$\bar{\varepsilon}_{ik}=\bar{\varepsilon}\delta_{ik}-0.5\varepsilon_a l_i l_k. \tag{28}$$

As a result, the averaged energy of interaction of the light field with the CLC can be written in the form $\delta F_E=(32\pi)^{-1}\varepsilon_a(\mathbf{E}\cdot\mathbf{l})(\mathbf{E}^*\cdot\mathbf{l})$; here we have omitted terms independent of $\mathbf{l}$.

In order to characterize the averaged Frank energy of smooth deformation of the CLC, besides the vector $\mathbf{l}(\mathbf{r})$ another vector $\mathbf{V}_s(\mathbf{r})$ is introduced (for the definition, see Ref. 18); in terms of them, the free energy takes the form[18]

$$\begin{aligned}F=\frac{\varepsilon_a}{32\pi}(\mathbf{E}\mathbf{l})(\mathbf{E}^*\mathbf{l})+{}^1/_{16}(K_{22}+3K_{33})(\nabla\mathbf{l})^2+{}^1/_2K_{22}(\mathbf{l}\mathbf{V}_s)\\ +{}^1/_4(K_{11}+K_{33})[\mathbf{l}\times\mathbf{V}_s]^2\end{aligned} \tag{29}$$

and the condition

$$(\operatorname{rot}\mathbf{l})_i=\frac{1}{q_0}(\operatorname{rot}\mathbf{V}_s)_i-\frac{1}{2q_0}e_{ijk}\mathbf{l}[\nabla^j\mathbf{l}\times\nabla^k\mathbf{l}] \tag{30a}$$

is imposed. From (30a) we have, to terms of order $\sim q_0^{-1}$,

$$\operatorname{rot}\mathbf{l}=O(1/q_0)\approx0. \tag{30b}$$

We assume that on at least one of the cell walls (for example, $z=0$) the orientation $\mathbf{l}$ of the director is rigidly prescribed along the normal. Hence follows

$$l_x(x,y,z=0),\quad l_y(x,y,z=0)=0. \tag{31}$$

The solution of Eq. (30b) has the form

$$\mathbf{l}=\operatorname{grad}\varphi(\mathbf{r}),\quad |\mathbf{l}|=|\operatorname{grad}\varphi|=1. \tag{32}$$

The second of Eqs. (32), under the boundary condition $\varphi(x,y,z=0)=\text{const}$ (pinned azimuth) has the unique solution $\mathbf{l}(\mathbf{r})\equiv\mathbf{e}_z$. This means absence of GON for a CLC with the director pinned on at least one of the surfaces. For a CLC with two free surfaces, GON apparently can occur; in the present paper we do not concern ourselves with this question.

6. CONCLUSION

Thus in the present paper a whole series of effects of action of a light beam on a CLC mesophase have been predicted and calculated. We note that for the existence of many of these effects, monochromaticity of the light (so characteristic of lasers) is not required.

Thus the induced birefringence (Section 3a) and the change of pitch of the helix, together with the resultant change of the nonlinear optical activity (Section 4), are determined by the combined action of the intensities of each of the spectral components.

From our point of view, it would be extremely important to detect experimentally the effects predicted and calculated in the present paper. In particular, it would be interesting to verify the conclusion about the absence of GON in a wall-oriented layer of CLC and about the change of pitch of the spiral in a light field.

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[1)] From an insufficiently attentive reading of the literature on LC, one can get the erroneous impression that the plane of polarization of light, in a cell containing CLC, can turn through many revolutions because of the huge gyrotropy characteristic of CLC. Actually, for $h \sim 1\ \mu m$ the specific rotation is large, $d\psi/dz \sim (10^1 - 10^2)$ rad/cm; but for cell thickness $L \sim 50\ \mu m$, the rotation is small, $\psi \sim (0.05\text{–}0.5)$ rad.

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