High-intensity electromagnetic wave in a cholesteric liquid crystal

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The behavior of a cholesteric liquid crystal in the field of a circularly polarized light wave of high intensity is investigated. It is found that the pitch of the cholesteric spiral increases under the action of such a wave. The corresponding formulas are presented.

1. Cholesteric liquid crystals (CLC) possess highly distinctive optical properties[1]. Several papers[2–4] have been devoted to the study of some of these properties—in particular, the anomalously strong rotation of the plane of polarization and the phenomenon of almost total reflection of circularly polarized light from a cholesteric medium. For example, it has been established[3,4] that the right- or left-polarized component (depending on whether the cholesteric spiral has right- or left-handed helicity) of a light wave with wave-vector k close to q_0 (where q_0 is the wave vector of the cholesteric spiral) passes almost totally into the cholesteric medium, while the other (left- or right-polarized) component is almost totally reflected.

The results obtained in the papers indicated are valid if the energy of the light wave in the liquid-crystalline medium is much smaller than the characteristic elastic energies of the LC, i.e., for low-intensity light waves. In the case of a sufficiently intense light wave (see below), the situation is changed and, in the solution of typical optical problems, it is necessary to take into account the change of structure of the LC under the action of the light. In particular, we should expect a shift of the range of wave vectors k in which almost total reflection occurs.

In the present paper, we consider an intense circularly polarized light wave propagating in an infinite cholesteric medium along its axis. In this case, as will be shown below, the pitch of the cholesteric spiral is increased.

2. As is well-known[5,6], to study the behavior of a liquid crystal under the action on it of an oscillating electromagnetic field, it is necessary to solve simultaneously the equations of motion of the director (the unit vector $\mathbf{n}(\mathbf{r})$ describing the preferred orientation of the axes of the LC molecules at the point $\mathbf{r}$), the hydrodynamic equations, and Maxwell's equations. However, in the case of a high-frequency field, when $\tau \gg 1/\omega$ (τ is the characteristic relaxation time of the LC, and ω is the frequency of the field), which is valid for a light wave, the problem of the propagation of light in a liquid-crystalline medium without conduction reduces to seeking the minimum of the time-averaged free energy of the LC and to solving Maxwell's equations in a medium with an average distribution of the director. We remark that the region of applicability of the results of [4] corresponds to the situation when the changes of the LC structure that arise when the energy of the light is taken into account are sufficiently small, and it is therefore possible to confine oneself to solving Maxwell's equations with the assumption that the dielectric permittivity tensor is not changed by the action of the field.

We now take into account the effect of the field on the distribution of the director. We shall consider a transverse electromagnetic wave propagating along the axis of an infinite CLC (the z axis). In view of the symmetry of the problem, we shall assume that all the functions under consideration depend only on z and the time t. We introduce the notation

$$n_x=\cos\theta, \quad n_y=\sin\theta, \quad \theta=\theta(z),$$
$$\mathbf{E}=\mathbf{E}(z)e^{i\omega t}, \quad \mathbf{E}\perp z, \tag{1}$$

where $\mathbf{E}$ is the electric field intensity, and n_x and n_y are components of the director.

The dielectric permittivity tensor of the CLC has the form

$$\varepsilon_{ik}=\begin{pmatrix} \varepsilon+{}^1/_2\delta\cos 2\theta & {}^1/_2\delta\sin 2\theta \\ {}^1/_2\delta\sin 2\theta & \varepsilon-{}^1/_2\delta\cos 2\theta \end{pmatrix} \quad (i,k=x,y),$$
$$\varepsilon={}^1/_2(\varepsilon_\parallel+\varepsilon_\perp), \quad \delta=\varepsilon_\parallel-\varepsilon_\perp, \quad \varepsilon_\parallel=\varepsilon_\parallel(\omega), \quad \varepsilon_\perp=\varepsilon_\perp(\omega), \quad \varepsilon_{zz}=\varepsilon_\perp(\omega), \tag{2}$$

where $\epsilon_\perp$ and $\epsilon_\parallel$ are respectively the dielectric permittivities of the CLC in the directions perpendicular and parallel to the symmetry axis of the CLC molecules (cf. [4]); we shall take the magnetic permeability equal to unity. Eliminating the magnetic field from the Maxwell equations in the usual way, we obtain equations for the electric components of the light wave. In the notation indicated above, these equations can be written in the following form:

$$E_\pm''+\frac{1}{2}\delta\frac{\omega^2}{c^2}E_\mp\exp\{\pm 2i\theta\}+\frac{\omega^2}{c^2}\varepsilon E_\pm=0, \tag{3}$$

where $E_\pm = E_x \pm iE_y$.

The free energy of the CLC in the presence of the field is given, as is well-known[7–9], by the expression*

$$H=\int {}^1/_2[k_{11}(\operatorname{div}\mathbf{n})^2+k_{22}(\mathbf{n}\operatorname{rot}\mathbf{n}+q_0)^2+k_{33}[\mathbf{n}\operatorname{rot}\mathbf{n}]^2]dV+H_f,$$

where k_{11}, k_{22} and k_{33} are elastic constants, and H_f is the energy of the field. For our problem,

$$H=\int {}^1/_2k_{22}(\theta'-q_0)^2\,dV+\overline{H}_f. \tag{4}$$

The line above H_f denotes time averaging.

Thus, to solve the problem it is necessary to solve Maxwell's equations (3) simultaneously with the Euler equations obtained from (4).

3. To solve in general form the problem formulated above is clearly difficult. However, a number of considerations suggest that the solution

$$\theta=pz, \tag{5}$$

is realized, where the quantity p must be determined from the condition for the minimum of the energy (4). First, we shall convince ourselves that (5) is indeed a solution of our problem. In fact, the solution of (3) in this case can be written in the form

$$E_x=E[\varkappa\cos(\omega t-\varphi-rz-pz)+\cos(\omega t-\varphi-rz+pz)],$$
$$E_y=E[-\varkappa\sin(\omega t-\varphi-rz-pz)+\sin(\omega t-\varphi-rz+pz)], \qquad (6)$$

where E and φ are respectively the amplitude and phase of the field, r is the real solution of the equation

$$\det\begin{Vmatrix} -(p+r)^2+\omega^2\varepsilon/c^2 & \delta\omega^2/2c^2 \\ \delta\omega^2/2c^2 & -(p-r)^2+\omega^2\varepsilon/c^2 \end{Vmatrix}=0, \qquad (7)$$

and the parameter κ is equal to

$$\varkappa=\left[(p-r)^2-\frac{\omega^2}{c^2}\varepsilon\right]\Big/\frac{\delta}{2}\frac{\omega^2}{c^2}.$$

Clearly, Eq. (7) is not changed on replacement of r by −r and, consequently, has two pairs of solutions, differing in sign. In correspondence with this, Eq. (3) has two linearly independent solutions of the type (6) with different r. We note that solutions with the other sign of ωt describe waves propagating in the opposite direction and give nothing new.

We shall consider one of the solutions. As is well known[10], the change of free energy of a system in a field for given temperature and for zero work by the field sources is equal to

$$\delta H=\delta H_0-\frac{1}{2}\int\delta\varepsilon_{ik}\overline{E_iE_k}\,dV, \qquad (8)$$

where H_0 is the free energy in the absence of the field. From (4) and (8) it is easy to obtain the Euler equation

$$\theta''-(\delta\varkappa E^2/k_{22})\sin[2(\theta-pz)]=0. \qquad (9)$$

It is now clear that (5) is indeed a solution of our problem, in which p must be found from the condition for the minimum of (4).

We note further that the problem of seeking the minimum of the free energy of the CLC leads to the solution (5) or (cf., e.g., the paper by de Gennes[11]) the solution

$$\sin\theta=\mathrm{sn}(z/\xi,\ k) \qquad (10)$$

for θ. Here, $\mathrm{sn}(z/\xi, k)$ is the elliptic sine (cf. [12]), and the constants appearing in the solution depend on the field and should be determined from the condition for the minimum of the free energy of the CLC.

In weak fields, the solution (10) can be expanded in a series (cf. [12]) in the field, and the leading term of the expansion has the form (5). Taking into account what has been said, it is natural to assume that the expansion of the required solutions in a series in the field starts from the term (5), and to attempt, by replacing θ by (5) in (3), to find solutions which, when substituted into (4), will lead to the correct dependence, of the form (5), of θ on z in the first non-vanishing order in the field. This program as we have seen, leads to Eq. (9) for θ and to the solution (5), (6).

We shall now find p. We note that, for this, we cannot confine ourselves to consideration of structures of the type (5); this is clear, if only from the fact that the variation of the free energy of the CLC with respect to p vanishes for these structures. As can be seen without difficulty, the variation procedure is not legitimate for the structures (5), since small changes of p in this case necessarily lead to changes in the distribution of the director that are not small. Therefore, to determine this variation, it is necessary to consider CLC structures of a more general type than (5), which would, on variation of the parameters describing these structures, translate the spirals (5) continuously into each other. A tensor of the form

$$\varepsilon_{ik}=\varepsilon_{ik}^0(q_0,\ \delta(1-\lambda))+\varepsilon_{ik}^c(p,\ \delta\lambda), \qquad (11)$$

clearly satisfies the requirements formulated. In (11), ϵ_{ik}^0 is the tensor (2), and λ is a parameter, on variation of which from 0 to 1 the tensor (11) changes from $\epsilon_{ik}^0(q_0, \delta)$ to $\epsilon_{ik}^0(p, \delta)$.

It is difficult to solve Maxwell's equations with the tensor (11) in general form. We shall calculate the change in pitch of the CLC spiral on switching on of the field in the case of small anisotropy $|\delta/\epsilon|\ll 1$. In addition, we shall assume that $p^2\sim q_0^2\sim\epsilon\omega^2/c^2$ and that the change of pitch of the spiral is small.

For this, we solve Eqs. (3) with the tensor (11) to accuracy δ/ϵ and, by substituting the solution into (8), find to the same accuracy the change in the free energy of the CLC on variation of λ from 0 to 1, which corresponds to going from q_0 to p in (5). As a result, we obtain

$$\delta H=\int\left\{\frac{1}{2}k_{22}(p-q_0)^2+\frac{1}{8}\delta^2\frac{\omega^2}{c^2}E_0^2\times\left[\frac{1}{k_\varepsilon^2-(k_\varepsilon+2p)^2}-\frac{1}{k_\varepsilon^2-(k_\varepsilon+2q_0)^2}\right]\right\}dV,$$

where $k_\epsilon^2=\epsilon\omega^2/c^2$, and E_0 is the field of the light wave in the medium with no anisotropy. Hence, equating δH to zero (which is the condition for the minimum of the free energy), we obtain

$$k_{22}(p-q_0)=-\frac{\delta^2E_0^2}{32}\frac{\omega^2}{c^2}\left(\frac{\omega}{c}\sqrt{\varepsilon}+2q_0\right)\Big/\left[q_0^2\left(\frac{\omega}{c}\sqrt{\varepsilon}+q_0\right)^2\right]. \qquad (12)$$

4. In conclusion, we observe that the pitch of the CLC structure is increased in the field of the light wave, the magnitude of the change being described by the corresponding formula (12). The region of applicability of the results is confined to sufficiently high fields, when nonlinear effects, i.e., dependence of the dielectric permittivity on the field, may become important. In addition, no account is taken in this work of absorption effects, which may make it difficult to study the above field dependence of the pitch of the CLC spiral experimentally.

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*[n rot n] ≡ n × curl n.

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