Photoinduced gyrotropy of the mesophase of liquid crystals

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A nonlinear optical activity is predicted in the mesophase of smectic C liquid crystals and of cholesteric liquid crystals in a cell with only a single orienting surface. In the smectic C crystals the nonlinear optical activity results from a rotation of the C-director by the light, while in the cholesteric crystals it results from a change in the pitch of the helix caused by light which is propagating along the axis of the helix. For cell thicknesses $\sim 50\ \mu$m, the magnitude of the nonlinear optical activity is determined by the constant $\eta \sim 0.1$ deg·cm/W and is ten orders of magnitude greater than the typical values of the electronic nonlinear optical activity.

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1. The extreme optical nonlinearity of the mesophase of liquid crystals,[1–5] which results from a reorientation of the director by light, makes it possible to observe several nonlinear optical effects at extremely low light intensities. In this letter we predict a photoinduced gyrotropy in a smectic C liquid crystal. We also discuss the nonlinear rotation of the polarization plane of the light in a cholesteric liquid crystal.

2. A smectic C liquid crystal has a multilayer structure in which the molecules in the layers are oriented along some direction $\mathbf{n}$ which makes a constant angle ϕ with the normal to the layers, $\mathbf{m}$, so that the scalar product of $\mathbf{n}$ and $\mathbf{m}$ is a constant: $(\mathbf{nm}) = \cos\phi = \text{const}$ (Fig. 1). We define the C-director as a unit vector in the plane of the layers which characterizes the projection of the director $\mathbf{n}$ on this plane: $\mathbf{c} = \{\mathbf{n} - \mathbf{n}(\mathbf{nm})\}/\sin\phi$, $|\mathbf{c}| = 1$. The vector $\mathbf{c}$ generally depends on the coordinates. For a smectic C liquid crystal we know that the only possible configuration, which is not accompanied by the appearance of a large elastic energy, is that with strictly plane layers.[6] We will accordingly examine the response of a smectic C liquid crystal to light in the approximation $\mathbf{m} = \mathbf{m}^0 = \text{const}$. In this approximation the free energy

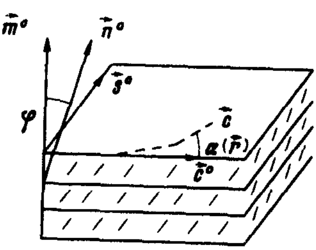

FIG. 1.

of a unit volume depends on the inhomogeneity of the C-director orientation. Introducing the angle α between the C-director and some fixed reference direction in the plane of the layers, we can write the free energy F (in ergs per cubic centimeter) of a unit volume of the liquid crystal subjected to the light as follows:

$$F = {}^1/_2\, B_1\,(\mathbf{c}\,\vec{\nabla}\alpha)^2 + {}^1/_2\, B_2\,([\mathbf{m}^\circ \times \mathbf{c}\,]\vec{\nabla}\alpha)^2 + {}^1/_2\, B_3\,(\mathbf{m}^\circ\vec{\nabla}\alpha)^2$$

$$+ B_{13}\,(\mathbf{c}\,\vec{\nabla}\alpha)\,(\mathbf{m}^\circ\vec{\nabla}\alpha) - \frac{\epsilon_a}{8\pi}\sin\phi\,(\mathbf{s}^\circ\mathbf{e})(\mathbf{n}^\circ\mathbf{e})\,|E|^2\alpha. \tag{1}$$

Here B_1, B_2, B_3, and B_{13} have the same dimensionality (dynes) and are of the same order of magnitude as the Frank constant for nematic or cholesteric liquid crystals[7]; $\epsilon_a = \epsilon_{\parallel} - \epsilon_{\perp}$ is the anisotropy of the dielectric constant of the liquid crystal at the frequency of the light; $\mathbf{s}_0 = [\mathbf{m}^0 \times \mathbf{c}^0]$; and $\mathbf{e} = \mathbf{e}^*$ is the polarization vector of the monochromatic light wave with complex amplitude $\mathbf{E}$. Since we intend to examine the effects in the first approximation in the intensity of the light wave, we have linearized the energy of the light-medium interaction with respect to α in (1): $F_E = \epsilon_a\,(\mathbf{nE})(\mathbf{nE}^*)/17\,\pi$.

We consider a cell holding a liquid crystal in which the layers of the crystal run parallel to the plates of the cell. We direct the z axis of the coordinate system along the normal to the plates, i.e., along $\mathbf{m}^0$. A linearly polarized wave, which is propagating at an oblique angle with respect to the z axis such that $(\mathbf{s}^0\mathbf{e}) \neq 0$, $(\mathbf{n}^0\mathbf{e}) \neq 0$, then leads to a rotation of the C-director, causing an extremely pronounced optical nonlinearity of the smectic C liquid crystal.[2] If the light beam is quite broad in comparison with the cell thickness L, and if the director is initially oriented homogeneously (through an appropriate treatment of the surfaces of the cell plates at $z = 0$ and $z = L$), we may assume that the perturbations of the C-director field depend on z along, i.e., $\alpha = \alpha(z)$. Retaining only those terms in the Frank energy which are quadratic in α, we then find from (1)

$$F = {}^1/_2\, B_3\left(\frac{\partial\alpha}{\partial z}\right)^2 - \frac{\epsilon_a \sin\phi\,(\mathbf{s}^\circ\mathbf{e})\,(\mathbf{n}^\circ\mathbf{e})\,|E|^2}{8\pi}\,\alpha\,. \tag{2}$$

Substituting (2) in the equation

$$\frac{\delta F}{\delta\alpha} - \frac{d}{dz}\,\frac{\delta F}{\delta\,(d\alpha/dz)} = 0$$

we find an equation which determines the equilibrium distribution of the director field:

$$\frac{d^2\alpha}{dz^2} = -\,\frac{\epsilon_a \sin\phi\,(\mathbf{s}^\circ\mathbf{e})(\mathbf{n}^\circ\mathbf{e})\,|E|^2}{8\pi B_3}, \tag{3}$$

$$\alpha(z) = -\,\frac{\epsilon_a \sin\phi\,(\mathbf{s}^\circ\mathbf{e})\,(\mathbf{n}^\circ\mathbf{e})\,|E|^2}{16\pi B_3}\,z^2 + az + b\,. \tag{4}$$

The constants a and b are determined from the boundary conditions. If the cell is constructed in such a manner that the orientation of the director is rigidly fixed $[\alpha(0)=0]$ at only one surface (the front surface) of the cell plate, and no boundary conditions of any sort are specified at the other surface, then $\alpha(L)$ is nonzero and is determined by Eq. (4) with $b=0$. The constant a is found by minimizing the free-energy functional:

$$\alpha(L) = \frac{\epsilon_a \sin\phi\,(\mathbf{s}^\circ \mathbf{e})(\mathbf{n}^\circ \mathbf{e})\,|E|^2 L^2}{16\pi B_3}. \tag{5}$$

As shown in Ref. 2, these distortions of the director field, which are proportional to the light intensity, lead to a nonlinear phase shift of the wave, i.e., to a self-focusing of bounded beams.

Two new effects arise in this situation, in which one of the cell surfaces is a free surface. First, the nonlinearity turns out to be four times that in the case in which the cell (of the same thickness) holding the smectic C liquid crystal has two orienting surfaces. Second, and particularly important in our case, the angle α at the exit is nonzero, $\alpha(L)\neq 0$, in a cell with a free exit surface. The polarization of the light wave propagating through such a medium adiabatically follows the rotation of the director and exits from the cell rotated through an angle $\alpha(L)$; i.e., the medium becomes optically active in a sense. A beam, which is propagating precisely along the z axis, exhibits the gyrotropic properties of the medium, and there is no nonlinear phase shift [in other experimental arrangements, also, there is no focusing of the light; the nonlinear phase shift is proportional to $\delta\epsilon \sim (\mathbf{s}^0 \mathbf{e}_p)(\mathbf{n}^0 \mathbf{e}_p)$, where $\mathbf{e}_p$ is the polarization of the probing wave[2]]. For some numerical estimates, we use for the smectic C liquid crystals some typical values for liquid crystals: $\epsilon_a \sim 0.5$, $B_3 \sim 5 \times 10^{-7}$ dyn, $L \sim 5 \times 10^{-3}$ cm, $(\mathbf{s}^0 \mathbf{e}) = (\mathbf{n}^0 \mathbf{e}) = 1/\sqrt{2}$, and $\sin\phi \approx 0.5$. We then find from (5) the result $\alpha(L) = 0.125\ |E|^2$, where $|E|^2$ is in electrostatic units. A nonlinear rotation of the polarization plane through an angle ~ 1 rad is reached at a light intensity $P \approx 1.5$ kW/cm^2.

3. We now consider the planar texture of a cholesteric liquid crystal in a cell of the type described above, with a single orienting plate. The director of the cholesteric liquid crystal in such a cell rotates in accordance with $\mathbf{n}(z) = \{n_x, n_y\} = \{\cos qz, \sin qz\}$, where $q = 2\pi/h$, and h is the pitch of the cholesteric helix. In the Mauguin limit, i.e., under the condition $\lambda \ll (n_e = n_o)h$, the polarization of an extraordinary wave (or an ordinary wave), which is incident on the crystal along the z axis, adiabatically follows the rotation of the director, and as the wave leaves the cell the polarization makes an angle qL with the x axis. On the other hand, if the anisotropy ϵ_a of the cholesteric liquid crystal is small, and if conditions are far from the Bragg-reflection region, a light wave propagating along the axis of the helix causes a change in the pitch of the helix described by[2)]

$$q - q_0 = -\left(\frac{\omega}{c}\right)^2 \frac{\epsilon_a^2}{128\pi K_{22}} \left\{ |E_R|^2 \frac{2q_0 + k}{q_0^2 (q_0 + k)^2} + \right.$$

$$|E_L|^2 \frac{2q_0 - k}{q_0^2 (q_0 - k)^2}\Bigg\}, \tag{6}$$

where k_{22} is the Frank constant, $k^2 = (\omega/c)^2 (\epsilon_{\parallel} + \epsilon_{\perp})/2$, and $|E_R|^2$ and $|E_L|^2$ are the intensities of the right-hand and left-hand circularly polarized components of the light wave **E** as it enters the medium:

$$\mathbf{E} = \frac{\mathbf{e}_x + i\mathbf{e}_y}{\sqrt{2}} E_R + \frac{\mathbf{e}_x - i\mathbf{e}_y}{\sqrt{2}} E_L .$$

In particular, for a linearly polarized wave we find from (1)

$$\delta q = q - q_0 = -\left(\frac{\omega}{c}\right)^2 \frac{\epsilon_a^2 |E|^2}{64\pi K_{22}} \frac{q_0}{(q_0^2 - k^2)^2}. \tag{7}$$

This change in the pitch of the helix leads to a nonlinear rotation of the polarization plane of a wave satisfying the Mauguin condition, through an angle $\alpha(L) = \delta qL$. For the values $q_0 = 2\pi/h = 2\pi \times 10^3\ \mathrm{cm}^{-1}$, $K_{22} = 5 \times 10^{-7}$ dyn, $\lambda = 0.5\ \mu$m, $\epsilon_{\parallel} \sim 3$, $\epsilon_{\perp} \sim 2$, and $L = 10^{-2}$ cm, we find $\alpha \sim 6 \times 10^{-6} |E|^2$. At a light intensity $P \sim 3\ \mathrm{kW/cm^2}$ the nonlinear rotation of the polarization plane of a Mauguin beam reaches values $\alpha(L) = \delta qL \sim 10^{-4}$ rad. Obviously, this effect may be greatly enhanced near resonance; here the increase in $\alpha(L)$ will be $\sim (q_0 - k)^{-2}$.

In summary, an orientational nonlinearity can lead to an extremely pronounced nonlinear optical activity of liquid crystals. For smectic C liquid crystals, this activity has a value of 0.1 deg • cm/W (for a cell 50 μm thick) and is ten orders of magnitude greater than the electronic nonlinear optical activity in $LiIO_3$ (Ref. 8).

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