Interaction of light waves with nonuniformly oriented liquid crystals

B. Ya. Zel'dovich and N. V. Tabiryan

Institute of Problems in Mechanics, Academy of Sciences of the USSR, and Special Design Bureau, Academy of Sciences of the Armenian SSR

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The orientation interaction is considered for light waves and liquid crystals with a uniform initial director orientation (such crystals include twisted nematics and cholesterics, and crystals with "hybrid" or "hyperhybrid" structures). Conditions are found for critical behavior to occur for a nonthreshold interaction. A light-induced Frederiks transition is predicted for cells with a hybrid nematic orientation, in which radiation absorption may reorient the director.

1. INTRODUCTION

Much recent work has been done on the interaction of light waves with the mesophases of liquid crystals (LC). Most of these results (particularly the experimental data) are limited to liquid crystals with a uniform initial orientation. Only a few papers have dealt with light wave interaction with initially nonuniformly oriented mesophases. Specifically Refs. 1–4 investigated the deformation or change in pitch of spiral cholesteric LC structures in response to a light field, and the associated nonlinear optical phenomena were considered; self-focusing of light in a cell containing a nematic LC with a hybrid orientation was considered theoretically in Refs. 5 and 6; finally, light-induced bleaching of an unconfined nematic liquid crystal (NLC) was observed experimentally in Ref. 7 (the bleaching was accompanied by a straightening out of the initially nonuniform directors).

In this paper we theoretically predict and analyze several novel phenomena specific to the interaction of light with nonuniform mesophases. We also discuss how the twisting of the director field influences the light-induced Frederiks transition (LFT).

2. FUNDAMENTAL EQUATIONS

The equations governing the interaction of external fields with LC's can be derived by minimizing the free energy: $\delta\int F dV = 0$. The free energy density F [erg/cm^3] of a nematic or cholesteric LC in a light field is given by

$$F=\frac{1}{2}K_1(\operatorname{div}\mathbf{n})^2+\frac{1}{2}K_2(\mathbf{n}\operatorname{rot}\mathbf{n}+q)^2+\frac{1}{2}K_3[\mathbf{n}\operatorname{rot}\mathbf{n}]^2-\frac{\varepsilon_{ik}}{16\pi}E_iE_k^*, \qquad (1)$$

where the K_i (in dynes) are the Franck constants, $\mathbf{n}=\mathbf{n}(\mathbf{r})$ is the director, $\mathbf{E}$ is the complex amplitude of the (monochromatic) light wave, and

$$\mathbf{E}_{\text{real}}(\mathbf{r},t)=0.5[\mathbf{E}\exp(-i\omega t+i\mathbf{kr})+\mathbf{E}^*\exp(i\omega t-i\mathbf{kr})];$$

$\varepsilon_{ik}=\varepsilon_\perp\delta_{ik}+\varepsilon_a n_i n_k$ is the dielectric permittivity tensor of the NLC at the light frequency. We have $q=0$ and $q=2\pi/h$ for nematics and cholesterics, respectively, where h is the equilibrium pitch of the cholesteric helix.

For situations when the director (unperturbed by the external fields) is uniformly oriented ($n=n^0=\text{const}$), it is helpful to write the variational equations in the form

$$\Pi_{ik}\left[\frac{\delta F}{\delta n_i}-\frac{\partial}{\partial x_j}\frac{\delta F}{\delta(\partial n_i/\partial x_j)}\right]=-\frac{\delta R}{\delta(\partial n_i/\partial t)}, \qquad (2)$$

where the factor $\Pi_{ik}=\delta_{ik}-n_i n_k$ ensures the normalization $n^2=1$, $\eta[\Pi]$ is the orientation viscosity coefficient, and R [erg/cm$^3\cdot$s] is the dissipation function density. The right-hand side of (2) describes the relaxation processes.[2] To estimate the relaxation time it suffices to take R of the form

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

i.e., we may neglect the effects of hydrodynamic motion in the nematic LC on the director orientation.

However, in the general case when $\mathbf{n}^0=\mathbf{n}^0(\mathbf{r})$ even the linearized equations (including only terms of first order in the director perturbation $\delta\mathbf{n}=\mathbf{n}-\mathbf{n}^0$) are quite complicated. It is simpler to write out the equilibrium equations in terms of two variable angles specifying the director orientation—i.e., we write

$$\mathbf{n}(\mathbf{r})=[\mathbf{e}_x\cos\varphi(z)+\mathbf{e}_y\sin\varphi(z)]\sin\theta(z)+\mathbf{e}_z\cos\theta(z), \qquad (3)$$

where $\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z$ are the unit vectors of a cartesian coordinate system with z axis normal to the walls of the NLC cell. If we assume that the system is uniform in the x,y plane then the variational equations take the form

$$(K_1\sin^2\theta+K_3\cos^2\theta)\frac{\partial^2\theta}{\partial z^2}-\frac{1}{2}(K_3-K_1)\sin 2\theta\left(\frac{\partial\theta}{\partial z}\right)^2-\sin\theta\cos\theta[K_3-2(K_3-K_2)\sin^2\theta]\left(\frac{\partial\varphi}{\partial z}\right)^2+K_2q\sin 2\theta\frac{\partial\varphi}{\partial z}$$
$$+\frac{\varepsilon_a}{16\pi}\{\sin 2\theta[\cos^2\varphi|E_x|^2+\sin^2\varphi|E_y|^2-|E_z|^2+\sin\varphi\cos\varphi(E_xE_y^*+E_x^*E_y)]$$
$$+\cos 2\theta[\cos\varphi(E_xE_z^*+E_x^*E_z)+\sin\varphi(E_yE_z^*+E_y^*E_z)]\}=\eta\frac{\partial\theta}{\partial t}, \qquad (4a)$$

$$\sin^2\theta(K_2\sin^2\theta+K_3\cos^2\theta)\frac{\partial^2\varphi}{\partial z^2}$$
$$+\sin 2\theta[K_3-2(K_3-K_2)\sin^2\theta]\frac{\partial\theta}{\partial z}\frac{\partial\varphi}{\partial z}$$
$$-K_2q\sin 2\theta\frac{\partial\theta}{\partial z}+\frac{\varepsilon_a}{16\pi}\{\sin^2\theta[\sin 2\varphi(|E_y|^2-|E_x|^2)$$
$$+\cos 2\varphi(E_xE_y^*+E_x^*E_y)]$$
$$+\sin\theta\cos\theta[\cos\varphi(E_yE_z^*+E_y^*E_z)-\sin\varphi(E_xE_z^*+E_x^*E_z)]\}$$
$$=\eta\sin^2\theta\frac{\partial\varphi}{\partial t}. \tag{4b}$$

Equations (4) must be solved simultaneously with the wave equation, which we write in the form

$$\Delta\mathbf{E}+(\omega/c)^2\hat{\varepsilon}(z)\mathbf{E}-\operatorname{grad}\operatorname{div}\mathbf{E}=0. \tag{5}$$

They simplify considerably in the "one-constant" approximation, i.e., if $K_1=K_2=K_3=K$. However, in this approximation Eq. (2) becomes

$$K\frac{\partial^2 n_i}{\partial z^2}-Kn_i\mathbf{n}\frac{\partial^2\mathbf{n}}{\partial z^2}-2Kq(\operatorname{rot}\mathbf{n})_i+2Kqn_i\mathbf{n}\operatorname{rot}\mathbf{n}$$
$$+\frac{\varepsilon a}{16\pi}\{\mathbf{nE}[E_i^*-n_i\mathbf{nE}^*]+\mathbf{nE}^*[E_i-n_i\mathbf{nE}]\}=\eta\frac{\partial n_i}{\partial t}, \tag{6}$$

which is also relatively tractable. In what follows we will use whichever form of the equilibrium equation happens to be more convenient.

When no external fields are present, Eqs. (4) and (6) describe how the equilibrium director configuration $\mathbf{n}^0(\mathbf{r})$ depends on the boundary conditions. In particular, if the director on the surface $z=0$ is rigidly oriented normal to the plate (i.e., along the z axis) but points along the x axis on the plane $z=L$, $\mathbf{n}^0(z)$ will lie in the x,z plane everywhere inside the NLC if no external fields are present. Setting $\varphi=0$ and $E=0$ in Eqs. (4), we get the simple expression

$$\theta^{(0)}(z)=(\pi/2)(z/L) \tag{7}$$

for $\theta^{(0)}(z)$ in the one-constant approximation. Cells of this type are said to be "hybrid." Cells for which $\theta^{(0)}(z)=pz$, $p>\pi/2L$, will be called hyperhybrid cells.

3. NONTHRESHOLD FREDERIKS EFFECT IN NONUNIFORM NEMATICS

The nonthreshold Frederiks effect occurs when the extraordinary wave (e-wave) makes an oblique angle with the director. It was shown theoretically and experimentally in Ref. 8 that this effect can be detected even in extremely weak light fields.

In this section we analyze how light interacts with an LC with a twisted initial director orientation and find configurations for which the effects of director nonuniformity are particularly pronounced.

1. Twisted nematics. We have

$$\mathbf{n}^{(0)}(z)=\mathbf{e}_x\cos\varphi(z)+\mathbf{e}_y\sin\varphi(z),\quad \varphi=pz \tag{8}$$

for an unperturbed nematic. If we assume that an e-wave propagates with wave vector

$$\mathbf{k}=(\omega/c)(\mathbf{e}_x\sin\alpha\cos\beta+\mathbf{e}_y\sin\alpha\sin\beta+\mathbf{e}_z\cos\alpha)\varepsilon_e^{1/2} \tag{9}$$

in the cell and make the approximation

$$(\omega/c)(\varepsilon_e^{1/2}-\varepsilon_\perp^{1/2})L\gg 1,\qquad \varepsilon_a/\varepsilon_\perp\ll 1,$$

we can write the wave polarization as

$$\mathbf{e}=\mathbf{E}/E=[\mathbf{k}[\mathbf{kn}]]/|[\mathbf{k}[\mathbf{kn}]]|, \tag{10}$$

where $[\mathbf{kn}]$ denotes the vector product. Substitution of Eqs. (8)–(10) into (4) yields an expression for the magnitude of the light-induced component $nz=\cos\theta$ of the director, where $\theta=\pi/2-\gamma$, $\gamma\ll 1$. To first order in the wave intensity, the steady-state equation for γ takes the form

$$\frac{d^2\gamma}{dz^2}+\xi\gamma-\frac{\varepsilon_a|E|^2\sin\alpha\cos\alpha}{8\pi K_1}\cos(\beta-pz)=0, \tag{11}$$

where $\xi=(2K_2-K_3)p^2/K_1$. The behavior is particularly interesting for $\xi>0$, because in this case the system becomes unstable to perturbations which bend the director out of the x,y plane by twisting the NLC director by the angle

$$\varphi_k=\pm\pi[K_1/(2K_2-K_3)]^{1/2}$$

(see, e.g., Ref. 9). Thus, if we take

$$2K_2>K_3\quad\text{and}\quad \varphi(z=L)\equiv\varphi_L=\varphi_k(1-\Delta/\pi),$$

where $\Delta\ll 1$, the solution of (11) satisfying the boundary conditions $\gamma(z=0)=\gamma(z=L)=0$ is found to be

$$\gamma=\left[\varepsilon_a|E|^2\sin 2\alpha\cos\frac{\varphi_L}{2}\cos\left(\beta-\frac{\varphi_L}{2}\right)L^2\sin\frac{\pi z}{L}\right]$$
$$\times[8\pi K_1\Delta(\varphi_L^2-\pi^2)]^{-1}. \tag{12}$$

The magnitude of the response γ depends in an extremely complicated way on the interaction geometry. In general, γ becomes anomalously large as $\Delta\to 0$. It is easy to check that in the one-constant case ($K_1=K_2=K_3$), for which instability develops when the director is twisted by an angle $\varphi_k=\pi$, the behavior of γ exhibits no anomalies at $\beta=0$. We must have $\beta\neq 0$ for instability to be observable in the orientation interaction of a light wave with a nematic liquid crystal.

2. Hyperhybrid cells. We take the unperturbed director distribution to be

$$\mathbf{n}^{(0)}=\mathbf{e}_x\sin\theta(z)+\mathbf{e}_z\cos\theta(z),\quad \theta(z)=pz \tag{13}$$

and consider the linearized equation for the y-component n_y. Using (9) and (13), we find that

$$\frac{d^2n_y}{dz^2}+p^2n_y-\frac{\varepsilon_a|E|^2\sin\alpha\sin\beta}{8\pi K}$$
$$\times(\cos\alpha\cos pz+\sin\alpha\cos\beta\sin pz)=0 \tag{14}$$

from (6) in the steady-state case. The solution of (14) satisfying $n_y(z=0)=n_y(z=L)=0$ is

$$n_y=\frac{\varepsilon_a|E|^2\sin\alpha\sin\beta}{16\pi Kp}\{(z-L)\cos\alpha\sin pz$$
$$+\sin\alpha\cos\beta(L\operatorname{ctg}pL\sin pz-z\cos pz)\}. \tag{15a}$$

As $\Delta=\pi-pL\to 0$, the component n_y again exhibits critical behavior:

$$n_y \approx -\frac{\varepsilon_a |E|^2 L^2 \sin^2\alpha \sin\beta \cos\beta}{16\pi^2 K\Delta} \sin\frac{\pi z}{L}. \qquad (15b)$$

We must have $\cos\beta \neq 0$ in order for criticality to occur (the latter arises because as $pL \to \pi$, the deformed NLC structure becomes unstable to fluctuations that take the director out of the x,z plane).

The deformed states of an NLC with $pL \lesssim \pi$ correspond to local minima of the free energy. These states can relax to an absolute energy minimum through the generation of disclinations. These questions are both interesting and extremely difficult to analyze; in particular, it would be of interest to see if light-induced generation of disclinations can occur in the cases considered above. Moreover, the types of instability mentioned above for nonuniform nematic liquid crystals are not exhaustive—other instabilities involving reversible adhesion of molecules on the substrate surfaces can also occur.[10]

We also mention the following interesting effect, which occurs, e.g., in planar-homeotropic (hybrid) oriented NLC's and is not directly related to nonlinear optical effects. According to (10), in the adiabatic approximation with $\beta \neq 0$, the polarization vector of the light wave transmitted by the cell will be rotated by 90° relative to the incident wave polarization. A wave with polarization $\mathbf{e} = \mathbf{e}_x$ incident in the z,y plane will be an ordinary wave (o-wave) if it is incident from the homeotropic wall ($z = 0$) and an e-wave if it is incident from the planar wall ($z = L$).

We also observe that in contrast to uniform LC's, in general the perturbation of the director in nonuniformly oriented cells is not greatest at the center of the cell.

3. Deformed cholesteric. Assume that Eq. (8) describes the initial structure of the director, but that the cell is filled with a cholesteric liquid crystal with equilibrium pitch $h = 2\pi/q$. For $p = q$ we have an ordinary Grandjean structure. For $p \neq q$ elastic stresses are present which in principle should be revealed when the cholesteric interacts with light. In the Mauguin limit, the equations describing this interaction are given by (11) except that

$$\xi = \frac{2K_2 - K_3}{K_1} p^2 - \frac{2K_2}{K_1} qp. \qquad (16)$$

We can use (16) to find conditions for $\xi > 0$, i.e., for instability to be possible. Thus, for $p \ll q$ the condition $\xi > 0$ requires that sign $(qp) < 0$, i.e., the direction of the mechanical twisting must be opposite to the cholesteric twisting. The critical twisting angle $\varphi_{cr} = p_{cr} L$ is

$$|\varphi_{cr}| = \pi^2 K_1 / 2K_2 |q| L.$$

The magnitude of the light-induced component n_z and its dependence on the experimental configuration are given by an expression analogous to (12) with ξ given by (16).

When the Mauguin condition is violated, it is much more difficult to describe the lightwave-cholesteric interaction because the natural modes of the LC are elliptically polarized waves whose properties depend in a complicated way on the angle of incidence of the wave and on the ratio of the wavelength divided by the pitch of the helices. However, this problem can be solved without difficulty by using the results in Ref. 11.

Under our assumptions, the phase shift of the e-wave can be found from the expression

$$\delta\Phi = \frac{\omega}{2c\varepsilon_e^{1/2}\cos\alpha} \int_0^L \delta\varepsilon_{ik} e_i e_k \, dz = -\frac{\omega\varepsilon_a}{c\varepsilon_e^{1/2} k^2 \cos\alpha} \int_0^L (\mathbf{k}\mathbf{n}^{(0)})(\mathbf{k}\delta\mathbf{n}) \, dz. \qquad (17)$$

The minus sign in (17) does not mean that the nonlinear phase shift is negative—indeed, the orientational nonlinearity always results in self-focusing of the light, i.e., $\delta\Phi > 0$. This can be seen by calculating $\delta\Phi$ for some specific cases by substituting the corresponding expressions for $\mathbf{n}^{(0)}(z)$ and $\delta\mathbf{n}(z)$ into (17).

4. FREDERIKS TRANSITION IN HYBRID-ORIENTED NEMATICS

If the light wave is incident normally on a homeotropically oriented cell, the director will become reoriented if the light intensity exceeds a certain threshold (this is called a light-induced Frederiks transition, or LFT). There may also be a threshold intensity for director reorientation when an o-wave is incident on a cell with a uniform planar orientation, but only if the adiabatic condition is violated.[12] This leads naturally to the question of what happens when an o-wave is normally incident on a hybrid cell (Fig. 1).

In order to find the threshold intensity for director reorientation, we must linearize and solve Eq. (6) to first order in the perturbation of the director n_y. In the one-constant approximation, we obtain

$$\eta \frac{\partial n_y}{\partial t} + K \frac{\partial^2 n_y}{\partial z^2} + K p^2 n_y + \frac{\varepsilon_a}{16\pi} [2n_y |E_y|^2 + (n_x E_x + n_z E_z) E_y^* + (n_x E_x^* + n_z E_z^*) E_y] = 0, \qquad (18)$$

where $p = \pi/2L$ and E_y is the strength of the light field incident on the nematic liquid crystal. Because of the perturbation of the director, the field components E_x and E_z are nonzero: $E_x, E_z \propto \varepsilon_a n_y$.

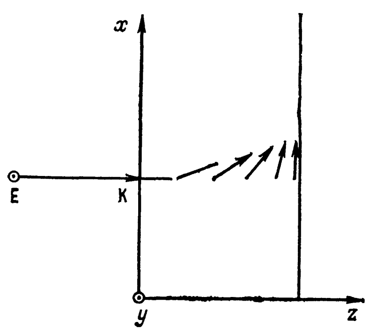

FIG. 1. Light-induced Frederiks transition in a hybrid cell with director initially aligned in the x, z plane. The light wave is incident normally on the cell (along the z axis) and is polarized along the y axis, i.e., perpendicular to the plane of the figure.

The field amplitudes E in (17) can be found from the Maxwell equations

$$\frac{d^2E_x}{dz^2}+\left(\frac{\omega}{c}\right)^2[(\varepsilon_\perp+\varepsilon_a n_x^2)E_x+\varepsilon_a n_x n_y E_y+\varepsilon_a n_x n_z E_z]=0, \quad (19a)$$

$$\frac{d^2E_y}{dz^2}+\left(\frac{\omega}{c}\right)^2[\varepsilon_a n_x n_y E_x+(\varepsilon_\perp+\varepsilon_a n_y^2)E_y+\varepsilon_a n_y n_z E_z]=0, \quad (19b)$$

$$\varepsilon_a n_x n_z E_x+\varepsilon_a n_z n_y E_y+(\varepsilon_\perp+\varepsilon_a n_z^2)E_z=0. \quad (19c)$$

Using (19c) to eliminate E_z, we can recast (19a) and (19b) to first order in the director perturbation as

$$\frac{d^2E_y}{dz^2}+k_\perp^2E_y=0, \quad (20a)$$

$$\frac{d^2E_x}{dz^2}+\left(\frac{\omega}{c}\right)^2\frac{\varepsilon_\parallel\varepsilon_\perp}{\varepsilon_\perp+\varepsilon_a n_z^2}E_x=-\left(\frac{\omega}{c}\right)^2\frac{\varepsilon_a\varepsilon_\perp n_x n_y}{\varepsilon_\perp+\varepsilon_a n_z^2}E_y, \quad (20b)$$

where $k_\perp=(\omega/c)\varepsilon_\perp^{1/2}$. The solution of (20a) is $E_y=E_0\times\exp(ik_\perp z)$.

We assume a solution of the form

$$E_x=E_0A(z)e^{ik_\perp z} \quad (21)$$

for (20b), where the function $A(z)$ varies slowly over a characteristic length $2k_\perp^{-1}$. Equation (20b) then gives

$$\frac{dA}{dz}-\frac{i}{2}k_\perp\frac{\varepsilon_a n_x^2}{\varepsilon_\perp+\varepsilon_a n_z^2}A=\frac{i}{2}k_\perp\frac{\varepsilon_a n_x n_y}{\varepsilon_\perp+\varepsilon_a n_z^2}E_0. \quad (22)$$

If A is smooth even over distances $(\varepsilon_a k_\perp)^{-1}$, then (22) and (19c) imply that $A\sim -n_y/n_x$ and $E_z\approx 0$.

The o-wave thus adiabatically follows the rotation of the director and the right-hand side of (18) is equal to zero. On the other hand, if $A(z)$ is not assumed to satisfy the above condition, (22) has the solution

$$A(z)=\frac{i}{2}\varepsilon_a k_\perp\int_0^z\frac{n_x(z')n_y(z')}{\varepsilon_\perp+\varepsilon_a n_z^2(z')}\exp[i\psi(z)-i\psi(z')]dz',$$

$$\psi(z)=\frac{1}{2}k_\perp\varepsilon_a\int_0^z\frac{n_x^2}{\varepsilon_\perp+\varepsilon_a n_z^2}dz. \quad (23)$$

From (19c) and (23),

$$E_z\approx-\frac{\varepsilon_a n_z E_0}{\varepsilon_\perp+\varepsilon_a n_z^2}[n_y+n_xA(z)]e^{ik_\perp z}, \quad (24)$$

and Eq. (18) takes the form

$$\eta\frac{\partial n_y}{\partial t}=K\frac{\partial^2 n_y}{\partial z^2}+\left[Kp^2+\frac{\varepsilon_a|E_0|^2}{8\pi}\left(1-\frac{\varepsilon_a n_z^2}{\varepsilon_\perp+\varepsilon_a n_z^2}\right)\right]n_y$$

$$-\frac{\varepsilon_a^2|E_0|^2k_\perp}{16\pi}\left(1-\frac{\varepsilon_a n_z^2}{\varepsilon_\perp+\varepsilon_a n_z^2}\right)n_x\int_0^z\frac{n_x(z')n_y(z')}{\varepsilon_\perp+\varepsilon_a n_z^2(z')}\times\sin[\psi(z)-\psi(z')]dz'. \quad (25)$$

If no field is present ($|E_0|^2=0$), Eq. (25) describes the damping of the small perturbations:

$$n_y(z,t)=\sum_{m=1}^{\infty}\sin\frac{m\pi z}{L}\exp(-\Gamma_m t), \quad (26a)$$

$$\Gamma_m=\frac{K}{\eta}\left(\frac{\pi}{L}\right)^2\left(m^2-\frac{1}{4}\right). \quad (26b)$$

The term proportional to the coefficient $-1/4$ in (26b), and the corresponding term in (25), describes the destabilizing influence of the initial nonuniform structure. It is easy to show that when

$$Ld\theta/dz=\theta(L)-\theta(0)\geqslant\pi$$

the planar structure is unstable (see also Ref. 9). In our case $\theta(L)-\theta(0)=\pi/L$ and this destabilization is offset by stabilization at the walls $z=0$ and $z=L$, where the director is rigidly oriented.

We will discuss the light-induced Frederiks transition for the case when the light is incident from the homeotropically orienting boundary; then $\theta(z)=\pi z/2L$. The lightwave-LC interaction is strongest where the phase difference $\psi(\Delta z)$ between the o- and the e-waves is $\lesssim 1$. The width Δz of the strong-interaction region is typically a small fraction of the total cell width L, so that for $z\lesssim\Delta z$ we need only retain the first nonvanishing term in the expression for $\psi(z)$:

$$\psi(z)=\left(\frac{d\theta}{dz}\right)^2\frac{k_\perp\varepsilon_a}{6\varepsilon_\parallel}z^3=\frac{\pi^2}{24}\frac{k_\perp\varepsilon_a}{\varepsilon_\parallel L^2}z^3\equiv Bz^3. \quad (27)$$

For $z\lesssim\Delta z$ we can also set $n_z\approx 1$ and $n_x\approx\pi z/2L$.

To simplify the notation the subscript y in $n_y(z,t)$ will be omitted in what follows. We define the dimensionless coordinate v by

$$v=B^{1/3}z,\quad \psi(z)=v^3, \quad (28)$$

so that $\psi(z)=1$ corresponds to $v=1$ and $\psi(z)=2\pi$ to $v=(2\pi)^{1/3}\approx 1.85$. The cell boundary $z=L$ corresponds to the following (large) dimensionless value:

$$v(z=L)=M=(\pi^2\varepsilon_a k_\perp L/24\varepsilon_\parallel)^{1/3}.$$

For example, we have $M\approx 6$ for radiation of wavelength $\lambda=0.6\ \mu$m in vacuum for a cell with $\varepsilon_\parallel\approx 3$, $\varepsilon_a\approx 1$, and $L=100\ \mu$m.

We also introduce the parameter ρ which gives the incident power density relative to the LFT threshold for an ordinary homeotropic cell of the same width L:

$$|E_0|^2=\rho(8\pi\varepsilon_\parallel K_3/\varepsilon_a\varepsilon_\perp). \quad (29)$$

In these variables the linearized small-perturbation equation takes the form

$$\frac{\partial n(v,t)}{\partial t}=\frac{K}{\eta}\left(\frac{\pi}{L}\right)^2\left(\frac{M}{\pi}\right)^2\left\{\frac{\partial^2 n}{\partial v^2}+\left(\frac{\pi}{2M}\right)^2(1+4\rho)n-3\rho\left(\frac{\pi}{M}\right)^2v\int_0^v v'n(v')\sin(v^3-v'^3)dv'\right\}\equiv\hat{L}n, \quad (30a)$$

$$n(v=0,\ t)=n(v=M,\ t)=0. \quad (30b)$$

We solve (30) by separation of variables by substituting $n(v,t)=n(v)\exp(-\Gamma t)$ into (30a). The problem then reduces to an eigenvalue problem for Γ. Because the operator on the right-hand side of (30a) is nonhermitian, the eigenvalues Γ are in general complex, $\Gamma = \Gamma' + i\Gamma''$. At threshold, the incident wave intensity p is such that the real part of Γ becomes negative: $\Gamma' \leqslant 0$; since Γ'' may be nonzero, the director perturbation may oscillate. The situation here is similar to the Frederiks transition in a uniform NLC cell induced by o-wave light.[12,13]

We have not yet succeeded in solving Eqs. (30) in closed form. Some qualitative estimates for the threshold LFT intensity in a hybrid cell can be derived as follows. We first estimate the width Δz of the strong-interaction region; taking $\psi(2\pi) \approx 2\pi$, we find from (27) that $\Delta z = (2\pi/B)^{1/3}$, or $\Delta z/L \approx (2\pi)^{1/3}/M$. The LFT threshold in a hybrid cell of width L is of course lower than for a homeotropic cell of width Δz:

$$P < \frac{c\varepsilon_\parallel K_3}{\varepsilon_a \varepsilon_\perp^{1/2}} \frac{\pi^2}{(\Delta z)^2} = \left(\frac{\pi}{L}\right)^2 \frac{c\varepsilon_\parallel K_3}{\varepsilon_a \varepsilon_\perp^{1/2}} \frac{M^2}{(2\pi)^{2/3}}. \qquad (31)$$

For the above parameter values, $M \approx 6$ and $M^2/(2\pi)^{2/3} \approx 10$.

In fact, in some cases the quantity $(\Delta z L)^{1/2}$ can be regarded as the "effective width" of the hybrid cell—this is true, e.g., for the light-induced Frederiks transition in the field of a wave localized within a distance Δz from the NLC surface.

When the light wave is incident from the plane-orienting boundary of the cell, we must take $\theta(z) = (\pi/2L)(L - z)$ in (25). The form of the resulting equation then differs considerably from (30). This implies that the LFT thresholds may differ for light incident from opposite directions on a cell with a nonuniform initial NLC orientation.

In some cases the integral in (30), or even the entire field term, can be regarded as a small perturbation, as is the case when $\theta(z) = pz$, $pL \ll 1$. In the opposite case when $\theta(z) \lesssim pz$, $p \lesssim \pi/L$, the director distribution becomes very sensitive to perturbations out of the x,z plane. Clearly, the LFT threshold is low in this case, $\rho \ll 1$. However, there is little point in analyzing the various cases enumerated above in the absence of specific experimental data. Moreover, in addition to the hybrid cell it would also be of great interest to investigate light-induced effects in cells in which a static magnetic field, say, is used to produce a nonuniform director orientation. This would make it possible to study these effects under conditions when the initial director field can be changed continuously.

5. THERMAL ORIENTATION EFFECTS IN A HYBRID CELL

The liquid crystal mesophases provide various opportunities for thermal reorientation; for instance, heating alters the pitch of cholesteric helices. Reference 14 considered nonlinear optical properties of C smectics associated with changes in the molecular orientation angle during heating. In this section we show that thermal orientation effects are also present in nonuniformly oriented nematics.

We assume that an o-wave of intensity below the LFT threshold is incident on a hybrid cell. It turns out that thermal effects can reorient the director even in this case. We will analyze this effect by using the equation

$$(K_1 \sin^2\theta + K_3 \cos^2\theta)\frac{d^2\theta}{dz^2} - \frac{1}{2}(K_3 - K_1)\sin 2\theta\left(\frac{d\theta}{dz}\right)^2 = 0 \qquad (32)$$

for the equilibrium director distribution in a hybrid cell. This equation follows from (4a) by setting $\alpha = 0$ and using $\mathbf{E}\cdot\mathbf{n} \equiv \mathbf{E}\mathbf{n} = 0$. We assume that the light field affects the nematic LC through the temperature-dependence of the Franck constants. Equation (32) has the implicit solution

$$\mathrm{E}(\theta, u^{1/2}) = \mathrm{E}\left(\frac{\pi}{2}, u^{1/2}\right)\left(1 - \frac{z}{L}\right), \qquad (33)$$

which satisfies the boundary conditions $\theta(z=0) = \pi/2$ and $\theta(z=L) = 0$; here E is the elliptic integral of the second kind and $u = 1 - K_1/K_3$. We can obtain an explicit expression for $\theta(z)$ if $u \ll 1$; to first order in u,

$$\theta(z) = \frac{\pi}{2}\left(1 - \frac{z}{L}\right) - \frac{u}{8}\sin\frac{\pi z}{L}. \qquad (34)$$

The change in θ due to heating is given by

$$\delta\theta = -\frac{\delta u}{8}\sin\frac{\pi z}{L}. \qquad (35)$$

Particularly large changes $\delta u = -\delta(K_1/K_3)$ should be expected near the nematic-smectic-A transition, for which the constant K_3 increases rapidly.[15]

If an e-wave is incident on a hybrid NLC cell, all three interaction mechanisms (orientation, thermal, and thermo-orientation) will occur simultaneously. We will use Eq. (17) and the corresponding expressions for the perturbation of the permittivity tensor to estimate the relative contributions of these mechanisms to the nonlinear phase shift. For the orientation and thermo-orientation mechanisms, $\delta\varepsilon_{ik} e_i e_k = \varepsilon_a \sin 2\theta^{(0)}\delta\theta$. We readily find an expression for $\delta\theta$ for the orientation mechanism from Eq. (4a) by setting $\varphi = 0$, $E_x \approx E = \mathrm{const}$, and $E_z \approx 0$ (for definiteness we consider a normally indicent e-wave). For the thermal mechanism of nonlinearity,

$$\delta\varepsilon_{ik} e_i e_k = [(\partial\varepsilon_\perp/\partial T) + (\partial\varepsilon_a/\partial T)(\mathbf{n}^{(0)}\mathbf{e})^2]\delta T.$$

The heating δT in the light field can be estimated as $\delta T \approx \sigma P \tau_t / C_p$, where σ is the absorption coefficient in cm^{-1}, τ_t [s] is the relaxation time, and C_p [erg/cm^3·deg] is the specific heat per unit volume. If the width of the beam is greater than the cell width L then $\tau_t \approx (L/\pi)^2/\chi$, where χ [cm^2/s] is the thermal diffusivity.

The above discussion leads to the following expressions for the nonlinear phase shifts $\delta\Phi_0$, $\delta\Phi_{t0}$, and $\delta\Phi_t$ due to the orientation, thermo-orientation, and thermal mechanisms:

$$\delta\Phi_{T0} = \frac{\partial(K_1/K_3)}{\partial T}\frac{\omega\varepsilon_a \sigma L^3 |E|^2}{256\pi^3 C_P \chi}, \qquad (36)$$

$$\delta\Phi_0 = \frac{\omega\varepsilon_a^2 L^3 |E|^2}{64\pi^3 c n_e K}, \qquad (37)$$

$$\delta\Phi_T = \left(\frac{\partial\varepsilon_\parallel}{\partial T} + \frac{\partial\varepsilon_\perp}{\partial T}\right)\frac{\omega\sigma L^3 |E|^2}{32\pi^3 C_P \chi}. \qquad (38)$$

We see at once that the phase shifts depend differently on the anisotropy of the permittivity: $\delta\Phi_0 \propto \varepsilon_a^2$, $\delta\Phi_{t0} \propto \varepsilon_a^1$, $\delta\Phi_t \propto \varepsilon_a^0$. We obtain $|\delta\Phi_{t0}/\delta\Phi_0| \approx |\delta\Phi_{t0}/\delta\Phi_t| \approx 6$ if we take $|\partial(K_1/K_3)/\partial T| \sim 10^{-1}\ \mathrm{deg}^{-1}$, $\sigma \sim 5\ \mathrm{cm}^{-1}$, $C_p \sim 10^7$

erg/cm^3·deg, $\chi \sim 10^{-4}$ cm^2/s, $n_e \sim 1.6$, $\varepsilon_a \sim 0.5$, $K \sim 5\cdot 10^{-7}$ dyn, and $|\partial\varepsilon_\parallel/\partial T + \partial\varepsilon_\perp/\partial T| \sim 10^{-3}$ deg^{-1}.

The thermal relaxation time ($\tau_t \approx 2.5\cdot 10^{-2}$ s for $L = 50$ μm) is significantly less than the relaxation time for the director orientation τ_{0r}. The latter can be estimated from Eqs. (4) or (6) and is $\tau_{0r} \sim \eta L^2/\pi^2 K \sim 5$ s for $\eta \sim 1$ Poise (see, e.g., Ref. 8).

"Giant" optical nonlinearity of an NLC mesophase caused by light-induced changes in the molecular conformation was discovered in Ref. 16. These changes alter the molecular polarizabilities and the macroscopic properties of the nematic (the order parameter Q and the phase transition temperature[17]). Presumably, the Franck constants are also affected. Well away from the nematic-smectic-A phase transition, the Franck constants depend on temperature as $K_i \propto Q^2$, so that $K_i/K_j = \text{const}$. On the other hand, it is not clear *a priori* how the ratios K_i/K_j will behave during light-induced conformational changes. A study of the absorption and orientation nonlinearity might shed some light on this question.

6. THRESHOLD THERMO-ORIENTATION NONLINEARITY

Cholesteric LC's in cells with a rigid homeotropic orientation at the walls were considered in Refs. 18–21, where it was shown that for cholesteric pitches h exceeding a critical value $h_{cr} = 2LK_2/K_3$, the LC assumes a uniform homeotropic orientation with director $n_x = n_y = 0$, $n_z = 1$. When h is less than h_{cr} (equivalently, if the wave vector q of the cholesteric LC exceeds the critical value $q_{cr} = \pi K_3/LK_2$), the director takes on a nonuniform orientation and the resulting structure is similar to that for a spring which is stretched at both ends.[21] Let us now examine what happens when $q < q_{cr}$ and q increases with T. Clearly, the lightwave intensity must exceed a threshold value if the LC is to be heated enough so that $q > q_{cr}$ and the director becomes realigned. If we take the change in the wave vector of the cholesteric helix to be

$$\delta q = (\partial q/\partial T)\,\delta T = (\partial q/\partial T)\,\sigma L^2 P/\pi^2 C_P \chi$$

(see Sec. 5), we get the result

$$P_{\text{thr}} = \frac{q_{cr} - q}{q_{cr}}\,\frac{\pi^2 \chi C_P}{\sigma L^2}\left(\frac{1}{q}\frac{\partial q}{\partial T}\right)^{-1} \qquad (39)$$

for the threshold intensity. If $L = 10$ μm, $(q_{cr} - q)/q_{cr} \sim 10^{-1}$, $q^{-1}(\partial q/\partial T) \sim 10$ K^{-1}, and the remaining parameter values are as above, we obtain $P_{\text{thr}} \approx 2$ W/cm^2.

The resulting structure can be shown to be optically active, so that we are in fact dealing with nonlinear optical activity. By suitably selecting the absorption coefficient of the cholesteric, the difference $q - q_{cr}$, and the polarization of the light wave, it is quite easy to ensure that the direct orientational effects within the light beam have no influence on the director orientation.[22] In general the magnitude of the thermal reorientation can be calculated numerically by using the formulas derived in Ref. 21. It was shown there that if the Franck constants satisfy $K_1 - 3(K_3 - K_2) < 0$, then hysteresis is present in the dependence $\theta_m(q/q_{cr} - 1)$, where θ_m is the angle of greatest director deflection (this occurs at the center of the cell) and $q/q_{cr} - 1$ is the relative increase above threshold. In addition to heating, light-induced conformational changes in the LC may also influence q.

7. CONCLUSIONS

We have considered several examples which illustrate the diversity of the interesting effects associated with lightwave-liquid-crystal interaction in cells with a nonuniform initial director distribution. Many such cells can be constructed. We have already seen that their qualitative properties depend not only on the boundary conditions but also on the specific physical properties of the LC material. This fact should be stressed, since in cells with a uniform director orientation the differences in the Franck constants, say, from one LC to another lead merely to quantitative differences. In many cases, even the description of the equilibrium structure for "nonuniform" cells encounters serious mathematical difficulties. Conservation laws[23] may provide a powerful technique for solving problems of this type. For instance, a theorem of E. Noether was used in Ref. 23 to derive analytic expressions for the equilibrium structure of complex configurations such as homeotropic-planar oriented cholesterics and cholesterics in magnetic fields with a homeotropic orientation at the walls.

Bistability is quite common in nonuniform cells (see, e.g., Ref. 9), and even the linear optical properties of such cells are far from trivial.[24] It is here that the qualitative differences between the effects of lightwaves and static fields on LC's are most pronounced.

We hope that the above discussion will stimulate more experimental work on the interaction of light with mesophases in cells with nonuniform LC orientations.

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