The Fréedericksz transition in a light field with spatially modulated intensity undergoing absorption

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(Submitted 24 December 1994)
Zh. Éksp. Teor. Fiz. **107**, 1921–1932 (June 1995)

It is assumed that as a result of the absorption of light in a nematic liquid crystal (NLC) there arise relatively long-lived photoconverted states of the molecules. The dependence of the density of the photoconverted molecules on the orientation of the director of the NLC relative to the polarization of the light field and the difference between the interaction with the environment of the photoconverted molecules and that of the ground-state molecules are shown to change the magnitude of the threshold for the light-induced Fréedericksz transition. For photoconverted dipole molecules the threshold can vary by several orders of magnitude. The distribution of the director in the cell for moderate overshoot of the threshold field as well as the dependence of the magnitude of the threshold field on the spatial period of the incident light intensity, the energy of detachment of the director from the cell surface, and the parameters of the photoconverted molecules are found. Homeotropic and planar NLC cells are considered. © *1995 American Institute of Physics.*

I. INTRODUCTION

Recently, intense study has been devoted to threshold reorientation of the director of nematic liquid crystals (NLC) in light fields, which is to say, the light-induced Fréedericksz transition (LIFT), discovered experimentally in Ref. 1. At present, phenomena associated with this transition are well studied and well understood in the case of transparent NLC's.[2,3] At the same time, recently noted properties of the LIFT in the field of a light wave whose frequency falls within the absorption region of the NLC have still not been completely explained.[4,5] Thus, the authors of Ref. 4 considered light-induced reorientation of the director in a NLC containing light-absorbing impurity molecules of anthroquinone dyes. They found that the threshold of the LIFT in this case is depressed significantly (by several orders of magnitude) in comparison with typical values in non-light-absorbing NLC's.

In Ref. 5 it was shown that different impurities influence the reorientation of the director due to the field of the light wave in a substantially different way, even if their absorption coefficients are approximately the same. This made it possible to establish that the net decrease of the threshold is not connected with the liberation of heat by the light-absorbing impurity. It was conjectured that the observed effect is possibly due to the appearance in the photo-excited impurity molecules of quite large electric dipoles whose orientational distribution can create an additional torque (in addition to that of the external field), which would effectively correspond to a decrease of the LIFT threshold. Note that such an idea was used in Ref. 6 to explain the lowering of the LIFT threshold in nonabsorbing NLC's containing chiral impurity molecules.

On the other hand, it is well known that in a number of NLCs, as a result of the absorption of light, the molecules change their conformation, thereby forming a relatively long-lived photoconverted state. The physical parameters of such photoconverted molecules differ from those of the NLC molecules.[7,8] This phenomenon is used, in particular, to write dynamic holographic arrays.[8,9]

In the present work we assume that photoconverted molecules form in a NLC under the action of an incident light field. They can arise as a result of photoconversion both of the ground-state molecules of the NLC and of the specially introduced impurity molecules (see Ref. 4 and 5).

The interaction of the photoconverted molecules with their environment differs from that of the ground-state molecules and thus gives rise to an additional contribution to the free energy of the NLC, proportional to the difference of these interactions and the concentration of the photoconverted molecules. Since the concentration of the photoconverted molecules depends on the orientation of the director of the NLC molecules relative to the polarization of the incident light, the contribution of the photoconverted molecules to the free energy will also depend on this orientation. As a result, the energetic advantage of one or another orientation of the director relative to the field varies as a function of the sign of this contribution, which leads, correspondingly, to a variation of the LIFT threshold.

The geometry of the light field is assumed to be the same as in the writing of dynamic arrays in the photoconverted molecule concentration. In this case an interference distribution in the intensity of the two light waves is created in the NLC cell. Hence we will also consider how the magnitude of the spatial period of its intensity affects the threshold value of the light field.

2. FREE ENERGY OF THE NLC

Assume that two plane monochromatic light waves are incident on the xz plane of a plane-parallel NLC cell, bounded by the planes $z=0$ and $z=L$, with the angle 2θ between them. The waves have the same amplitude and are linearly polarized along the y axis of the Cartesian coordinate system, and their wave vectors are oriented symmetrically relative to the yz plane:

$$\mathbf{E}_{1,2}=\mathbf{E}_0\cos(\omega t-\mathbf{q}_{1,2}\mathbf{r}),$$
$$\mathbf{q}_{1,2}=q_0(\pm\sin\theta,0,\cos\theta), \tag{1}$$

where $q_0=\sqrt{\varepsilon_\perp}\,\omega/c_0$ is the magnitude of the wave vector.

As a result of interference in the bulk of the NLC the electric vector of the light field has the form

$$\mathbf{E}=\frac{1}{2}[\mathbf{E}(\mathbf{r})e^{-i\omega t}+\mathbf{E}^*(\mathbf{r})e^{i\omega t}],$$
$$\mathbf{E}(\mathbf{r})=2\mathbf{E}_0\cos\left(\frac{1}{2}x\Delta q\right)\exp(iq_0z\cos\theta), \tag{2}$$
$$\Delta q=2q_0\sin\theta.$$

We will assume absorption of the light field (2) causes the NLC molecules to undergo conformational conversion and form photoconverted states with lifetime τ. If the photoconversion time of a molecule is much smaller than the characteristic time of reorientation of the director, then the photoconverted molecule concentration can be found by solving the kinetic equation

$$\frac{\partial c}{\partial t}=-\frac{c}{\tau}+D\frac{\partial^2c}{\partial x^2}+\delta\kappa_{\alpha\beta}I_{\alpha\beta}(1-c), \tag{3}$$

where D is the diffusion coefficient of the photoconverted molecules, $I_{\alpha\beta}=E_\alpha E_\beta^*/2$ is the intensity tensor of the light field inside the cell; $\kappa_{\alpha\beta}=\kappa_\perp\delta_{\alpha\beta}+\kappa_a n_\alpha n_\beta$ is the absorption coefficient tensor averaged over the orientations of the long axes of the molecules; $\kappa_a=\kappa_\parallel-\kappa_\perp$, $\kappa_\parallel$ and $\kappa_\perp$ are the absorption coefficients for the light polarized parallel to and perpendicular to the director; n_α are the components of the director; and δ is the quantum efficiency of photoconversion of the molecules. In writing down Eq. (3) we have assumed that the absorption is weak and have neglected variation of the intensity of the light field in its direction of propagation.

As noted, the interaction of the photoconverted molecules with the surrounding ground-state NLC molecules differs from the interaction of the latter with each other. Therefore, for $c\ll1$ the photoconverted molecules give a linear contribution to the free energy of the NLC in the concentration, which, taking the inhomogeneity of the spatial distribution of the photoconverted molecules into account, can be written in the form

$$F_{PM}=-\int c(\mathbf{r})\Delta U\,dV, \tag{4}$$

where ΔU is equal to the difference between the interactions of the ground-state NLC molecules and the photoconverted molecules with their environment.

If the orientation time of the director in the electric field of the light wave is much longer than the lifetime τ of the photoconverted molecules and their characteristic diffusion time $[D(\Delta q)^2]^{-1}$, then the stationary value of the concentration of the photoconverted molecules that is reached adiabatically tracks the variation of the director. It can be found by solving the kinetic equation (3) and for $c\ll1$ has the form

$$c(\mathbf{r})=\frac{\tau\delta}{1+\tau D(\Delta q)^2}\left\{\tau D(\Delta q)^2[\kappa_\perp E_0^2+\kappa_a(\mathbf{nE}_0)^2]+\frac{1}{2}[\kappa_\perp|E|^2+\kappa_a(\mathbf{nE})(\mathbf{nE}^*)]\right\}. \tag{5}$$

Taking expression (2) for $\mathbf{E}(\mathbf{r})$ into account, one can see that a diffraction pattern of the photoconverted molecule concentration arises in the NLC cell, with spatial period along the x axis equal to

$$l=\frac{2\pi}{\Delta q}=\frac{\pi}{q\sin\theta}.$$

Thus the free energy of the NLC cell in the field of the light wave initiating formation of the photoconverted molecules can be written in the form

$$F=F_{el}+F_E+E_S+F_{PM}, \tag{6}$$

where F_{PM} is given by Eq. (4),

$$F_{el}=\frac{1}{2}\int dV\{K_1(\operatorname{div}\mathbf{n})^2+K_2(\mathbf{n}\cdot\operatorname{rot}\mathbf{n})^2+K_3[\mathbf{n}\times\operatorname{rot}\mathbf{n}]^2\},$$
$$F_E=-\frac{1}{16\pi}\int\varepsilon_{ik}E_iE_k^*dV, \tag{7}$$
$$F_S=-\frac{W}{2}\int(\mathbf{ne})^2dS,\quad W>0.$$

Here F_{el} is Franck's elastic energy, F_E is the contribution to the free energy of the NLC from the electric field of the light wave (see Ref. 3), F_S is the energy of detachment of the director from the surface of the cell, chosen in the form of the Rapini potential,[10] $\varepsilon_{ik}=\varepsilon_\perp\delta_{ik}+\varepsilon_a n_i n_k$ is the dielectric tensor of the nematic at the frequency of the incident light, $\varepsilon_a=\varepsilon_\parallel-\varepsilon_\perp$ is the anisotropy of the dielectric tensor, and $\mathbf{e}$ is the unit vector of the axis of easy orientation in the planes of the cell. In what follows, we will neglect the weak dependence on the photoconverted molecule concentration of Franck's elastic constants K_i and the dielectric tensor of the NLC.

3. THRESHOLD OF ORIENTATIONAL STABILITY IN A HOMEOTROPIC CELL

We will assume that the initial orientation of the director of the NLC is homeotropic, i.e., the unit vector of the easy orientation axis in the plane of the cell satisfies $\mathbf{e}\|OZ$. Since the field vector $\mathbf{E}\|OY$, we will represent the director in the bulk of the NLC in the form $\mathbf{n}=\mathbf{j}\sin\varphi(x,z)+\mathbf{k}\cos\varphi(x,z)$, where $\mathbf{j}$ and $\mathbf{k}$ are basis vectors of the Cartesian coordinate system. By virtue of the homogeneity of the system in the y direction, the angle φ of the deviation of the director from its initial direction does not depend on y.

We will be interested in the behavior of the director for values of the electric field near the reorientation threshold of the director. Therefore, we will limit ourselves to the linearized (in φ) variational equation. In the single-constant approximation it has the following form:

$$K\left(\frac{\partial^2\varphi}{\partial x^2}+\frac{\partial^2\varphi}{\varphi z^2}\right)+[A+B\cos(x\Delta q)]E_0^2\varphi=\eta\frac{\partial\varphi}{\partial t}, \qquad (8)$$

where

$$A=\frac{\varepsilon_\perp}{4\pi\varepsilon_\parallel}(\varepsilon_a+8\pi\tau\delta\kappa_a\Delta U), \qquad (9)$$

$$B=\frac{\varepsilon_\perp}{4\pi\varepsilon_\parallel}\left[\varepsilon_a+\frac{8\pi\tau\delta\Delta U}{1+\tau D(\Delta q)^2}\kappa_a\right]. \qquad (10)$$

The term on the right-hand side of Eq. (8) is a consequence of taking account of relaxation process (as, for example, in Ref. 2) while neglecting the coupling between the director and the hydrodynamic motions of the NLC, and η is the relaxation constant, $\eta\sim10^{-2}$–1 P.

The boundary conditions on Eq. (8) are

$$\left(\frac{\partial\varphi}{\partial z}-\frac{W}{K}\varphi\right)_{z=0}=0, \qquad \left(\frac{\partial\varphi}{\partial z}+\frac{W}{K}\varphi\right)_{z=L}=0. \qquad (11)$$

The solution of Eq. (8) should be even and periodic in x in accordance with the character of the light field (2) perturbing the director of the NLC. Using the method of separation of variables and requiring that the solution satisfy the boundary conditions (11), we obtain

$$\varphi(x,z,t)=\sum_{n=1}^{\infty}\sum_{r=0}^{\infty}\varphi_{nr}\left[\frac{K\mu_n}{W}\cos(\mu_n z)+\sin(\mu_n z)\right]\mathbf{ce}_r\left(\frac{1}{2}x\Delta q,-\frac{2BE_0^2}{K(\Delta q)^2}\right)\exp(\Gamma_{nr}t), \qquad (12)$$

where the function $\mathbf{ce}_r(\xi,Q)$ is the even solution of the Mathieu equation

$$\frac{d^2f}{d\xi^2}+(a-2Q\cos 2\xi)f=0, \qquad (13)$$

corresponding to the eigenvalue $a_r(Q)$,

$$\Gamma_{nr}=\frac{1}{\eta}\left[AE_0^2-K\mu_n^2-\frac{1}{4}K(\Delta q)^2a_r\left(-\frac{2BE_0^2}{K(\Delta q)^2}\right)\right], \qquad (14)$$

μ_n is the nontrivial solution of the equation

$$\mathrm{tg}(\mu L)=\frac{2WK\mu}{(K\mu)^2-W^2}, \qquad (15)$$

and φ_{nr} are the integration constants.

As can be seen from Eq. (12), the permutation of the director grows with time if any of the growth rates satisfy $\Gamma_{nr}>0$. Since $a_0(Q)<a_1(Q)<a_2(Q)<\ldots$ (Ref. 11), the threshold value of the amplitude E_0 can be found from the condition $\Gamma_{10}=0$ in which μ_1 is the smallest (in absolute value) nontrivial root of Eq. (15) (obviously, it can always be chosen to be positive). This condition has the form

$$AE_0^2=K\mu_1^2+\frac{1}{4}K(\Delta q)^2a_0\left(-\frac{2B}{K(\Delta q)^2}E_0^2\right) \qquad (16)$$

and in general can be solved for E_0 only numerically. Let us first consider the limiting cases for which the solution has an analytic form.

Let Δq be large enough that $|Q|=(2|B|/K(\Delta q)^2)\times E_0^2\ll1$. Then $a_0(Q)\simeq-Q^2/2$ (Ref. 11) and if $A>0$ holds for ($A<0$ the initial, homeotropic orientation of the director is stable), then the expression for the threshold of reorientation of the director follows from Eq. (16):

$$E_{0\mathrm{th}}=\sqrt{\frac{K}{A}}\mu_1\left[1-\left(\frac{\mu_1B}{2\Delta qA}\right)^2\right]. \qquad (17)$$

If Δq is small enough that $|Q|\gg1$, then we have $a_0(Q)\simeq-2(|Q|-\sqrt{|Q|})$ (Ref. 11) and for $A+|B|>0$ we obtain from Eq. (16)

$$E_{0\mathrm{th}}=\sqrt{\frac{K}{A+|B|}}\mu_1\left[1+\sqrt{\frac{2|B|}{A+|B|}}\frac{\Delta q}{4\mu_1}\right]. \qquad (18)$$

In the case of absolutely rigid detachment ($W=\infty$) it follows from Eq (15) that $\mu_1=\pi/L$, and, taking into account that $\Delta q=2\pi/l$, where l is the period of the interference pattern, we find that the conditions $Q\ll1$ and $Q\gg1$ are equivalent, respectively, to the conditions $l\ll L$ and $l\gg L$.

Expressions (16)–(18) determine the magnitude of the threshold value of the electric vector of the light wave as a function of the spatial period of the intensity of the light field, the magnitude of the parameters determining the concentration of the photoconverted molecules and their interaction with the NLC molecules, and also on the energy of detachment of the director from the surface of the cell. In particular, if the boundary conditions on the director are not absolutely rigid ($W\neq\infty$), but the detachment energy is large enough that $WL/K\gg1$, then it is not hard to obtain from Eq. (15) that $\mu_1=\pi/L(1-2L/WK)$ and the magnitude of the threshold field, defined by Eq. (16), decreases as a function of W, as could have been expected. In the opposite case of small detachment energy, so that $WL/K\ll1$ holds, we have $\mu_1=\sqrt{2W/KL}$ and in the limit $\Delta q\rightarrow0$ the threshold field E_0 tends toward the value $\sqrt{2W/(A+|B|)L}$, independent of Franck's elastic constant.

The dependence of the magnitude of the threshold field on Δq for arbitrary value of the detachment energy W, obtained by numerical solution of Eq. (16), is shown in Fig. 1.

For a homogeneous intensity distribution of the incident light ($\Delta q=0$) and absolutely rigid detachment of the director from the surface of the cell ($\mu_1=\pi/L$), the well-known formula for the threshold of the light-induced Fréedericksz transition[2] follows from formula (18) when there are no photoconverted molecules ($\Delta U=0$, $A=B$).

As is clear from expressions (17), (18) and (9), (10), photoconversion changes the reorientation threshold of the detector; the magnitude of the threshold either can decrease or increase, depending on the sign of the product of the anisotropy of the light absorption coefficient κ_a and the difference ΔU in the interaction of the ground-state NLC molecules and the photoconverted molecules with their environment. Decreasing the period of the interference pattern increases the magnitude of the threshold field (see Fig. 1).

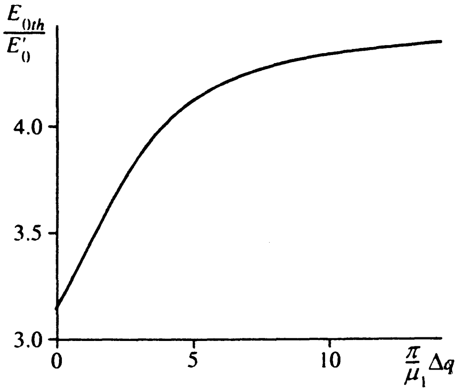

FIG. 1. Dependence of the magnitude of the threshold field on the spatial period of the intensity of the light wave. Here we have written E'_0 $\sqrt{K/2A}(\mu_1/\pi)$, and the diffusion coefficient of the photoconverted molecules D was set equal to zero in the calculation.

If the diffusion coefficient of the photoconverted molecules is small enough that $\tau D(\Delta q)^2 \ll 1$ holds or the intensity of the light field in the NLC cell is homogeneous ($\Delta q = 0$), then we have $A = B$ and, as can be seen from (16) and (17) (or in the general case simply by comparing Eqs. (4) and (5) for F_{PM} with Eq. (7) for F_E) the photoconverted molecules merely renormalize the anisotropy of the dielectric constant: $\varepsilon \to \varepsilon'_a = \varepsilon_a + 8\pi\tau\delta\kappa_a \Delta U$. In this case, if the period of the interference pattern is small enough that $l \ll L$ (i.e., $\Delta q \gg \mu_1$, but $\tau D(\Delta q)^2 \ll 1$), then the magnitude of the threshold field satisfies $E_{0\text{th}} \approx \mu_1 \sqrt{K/A}$ and is $\sqrt{2}$ times larger than when the intensity of the light field is essentially homogeneous ($l \ll L$, i.e., $\Delta q \ll \mu_1$).

If the diffusion of the photoconverted molecules is large enough that $\tau D(\Delta q)^2 > 1$, then $A \neq B$ holds and the dependence of the threshold field on the parameters of the photoconverted molecules becomes more complex in accordance with Eqs. (16)–(18).

It is possible to determine the values of the amplitudes φ_{1r} in Eq. (12) in the steady-state regime in the case in which the threshold value of the field (following from the condition on Γ_{10}) has been exceeded. If the intensity of the light field is essentially homogeneous ($\Delta q \to 0$) and its variation as a consequence of absorption can be neglected, then the calculation proceeds as in Refs. 2 and 3 taking account of terms in Eq. (8) nonlinear in φ and using the geometric optics approximation for Maxwell's equations in an anisotropic inhomogeneous medium. As a result, for infinitely rigid detachment of the director from the surface of the cell we find that

$$\varphi(z) = \varphi_{10} \sin\left(\frac{\pi}{L} z\right) + \ldots,$$

where

$$\varphi_{10}^2 = 2\left(1 - \frac{9\varepsilon_a}{4\varepsilon_\parallel}\right)^{-1}\left[1 + \frac{(8\pi/3)\tau\delta\kappa_a\Delta U}{\varepsilon_a + (16\pi/3)\tau\delta\kappa_a\Delta U}\right]\frac{P - P_{\text{th}}}{P_{\text{th}}}. \tag{19}$$

Here $P = (c_0\sqrt{\varepsilon_\perp}/8\pi)4E_0^2$ is the z component of the Poynting vector and P_{th} is its threshold value.

Thus, as follows from Eq. (19), the formation of photoconverted molecules ($\Delta \neq 0$) leads to a change in the amplitude of the deviation of the director in a light field even when the light field exceeds the threshold power by the same margin as when photoconverted molecules are absent. In this case, if $\kappa_a \Delta U > 0$ holds, then the amplitude of the deviation of the director grows (note that in this case the magnitude of the threshold value of the field vector E_0 decreases).

4. THRESHOLD OF ORIENTATIONAL INSTABILITY IN A PLANAR CELL

Now let the initial orientation of the director in the cell be planar and let the x axis point in the direction of the easy axis. To simplify the expressions, we will take the energy of detachment of the director from the surface of the cell to be infinitely large ($W = \infty$) and the diffusion of the photoconverted molecules to be negligibly small. The perturbed director in the electric field of the light wave (2) will lie in the xy plane, and it follows that it should be sought in the form $\mathbf{n} = \mathbf{i}\cos\varphi(x,z) + \mathbf{j}\sin>\varphi(x,z)$. The linearized (in the angle φ) equation, obtained after a variational procedure as in the previous section, has the form

$$K\left(\frac{\partial^2\varphi}{\partial x^2} + \frac{\partial^2\varphi}{\partial z^2}\right) + \frac{\varepsilon_a}{16\pi}(2\varphi|E_y|^2 + E_y E_x^* + E_y^* E_x) = \eta\frac{\partial\varphi}{\tau}, \tag{20}$$

The electric vector of the light field is found from Maxwell's equation. Discarding the terms in Eq. (20) which are second-order in the small parameter φ, we find that the field component $E_y(x,z)$ is given by Eq. (2) as before, and E_x can be represented following Ref. 3 in the form

$$E_x(x,z) = E_y(x,z)\Psi(x,z), \tag{21}$$

where the function $\Psi(x,z)$, which is smooth in the coordinate z on the scale $(q_0 \cos\theta)^{-1}$, satisfies the equation

$$\frac{\partial\Psi}{\partial z} - i\frac{q_e^2 - q_0^2 + \Delta q^2/4}{q_0\cos\theta}\Psi = i\frac{q_e^2 - q_0^2}{q_0\cos\theta}\varphi,$$
$$q_e = \frac{\omega}{c}\sqrt{\varepsilon_\parallel}, \quad q_0 = \frac{\omega}{c_0}\sqrt{\varepsilon_\perp}. \tag{22}$$

If $\Psi(x,z)$ is also smooth in z on the scale $\xi = q_0\cos\theta/q_e^2 - q_0^2 + (\Delta q)^2/4$, then neglecting the derivative in Eq. (22) we obtain

$$\Psi(x,z) = -\frac{q_e^2 - q_0^2}{q_e^2 - q_0^2 + (\Delta q)^2/4}\varphi(x,z). \tag{23}$$

After expressions (21) and (23) are substituted in Eq. (20), the latter coincides with Eq. (8) with A and B replaced by $(A(\Delta q)^2/4/q_e^2 - q_0^2 + (\Delta q)^2/4)(\varepsilon_\parallel/\varepsilon_\perp)$.

Then the solutions of Eq. (20) and the expressions for the threshold field will be described by Eqs. (12), (14)–(18) with the same replacement of the quantities A and B as before, and also setting $\mu_1 = \pi/L$ in agreement with the solution of Eq. (15) for infinitely rigid detachment of the director from the surface. Since the function $\varphi(x,z)$ in this case should be taken to be smooth in z on the scales $(q_0\cos\theta)^{-1}$ and ξ, from the form of the solution (12) the inequalities $L\gg(q_0\cos\theta)^{-1}$ and $L\gg\xi$ must be satisfied.

If the angle θ is not too small, then, in addition to the latter inequalities, it is possible also to fulfill the condition $L\Delta q=2q_0L\sin\theta\gg1$ (or $2\pi L\gg l$). The threshold value of the field can be found in this case from expression (17) allowing for these changes. We obtain

$$E_{0\text{th}}=\sqrt{\frac{K}{A}\frac{\pi}{L}}\sqrt{\frac{\varepsilon_\perp}{\varepsilon_\parallel}+\frac{\varepsilon_a}{\varepsilon_\parallel\sin^2\theta}}. \tag{24}$$

Comparing expressions (24) and (17), it can be seen that for planar orientation of the director the magnitude of the threshold field is always greater (and for small angles θ much greater) than for homeotropic orientation.

For small angles θ and small thickness of the cell it is possible to fulfill the condition $L\Delta q=2q_0L\sin\theta\ll1$ or $(2\pi L\ll l)$. We obtain an expression for $E_{0\text{th}}$ then by using Eq. (18):

$$E_{0\text{th}}=\sqrt{\frac{K}{2A}\frac{\varepsilon_a}{\varepsilon_\parallel}\frac{\pi}{L\sin\theta}}. \tag{25}$$

It is not hard to verify that the inequality $\sin\theta\ll\sqrt{\varepsilon_a/\varepsilon_\parallel}$ follows from the smoothness condition on $\varphi(x,z)$. Therefore in this case $E_{0\text{th}}$ is always much bigger than $E_{0\text{th}}$ for the homeotropic orientation. In the limit $\Delta q\to0$ the intensity of the light field in the cell becomes homogeneous and it follows from Eq. (25) that $E_{0\text{th}}\to\infty$, i.e., reorientation of the director in the light field does not take place, in accordance with the result obtained earlier in Ref. 3. In this case, as can be seen form relation (23), $\Psi=-\varphi$ holds and the field of an ordinary wave adiabatically tracks the tilts of the director, remaining all the while perpendicular to it (the Mogen regime).

If the function $\Psi(x,z)$ is smooth only on the scale $(q_0\cos\theta)^{-1}$, but not on the scale ξ, then it can be found by solving the differential equation (22). In this case it is sufficient to consider only small angles θ since for large angles $\theta<\pi/2$ the function $\Psi(x,z)$ becomes smooth in z also on the scale ξ and we return to the case already considered.

The solution of Eq. (20) in this case can be obtained in analytic form, following Ref. 3, only for small cell thicknesses, when $(q_e-q_0)L\ll\pi$. As a result, for the threshold field we have the expression

$$E_{0\text{th}}=\sqrt{\frac{K\varepsilon_\perp\pi}{2A\varepsilon_\parallel L}}\left[1+\frac{(q_e-q_0)^2L^2}{2\pi^2}+\frac{L\Delta q}{4}\right], \tag{26}$$

where $L\Delta q\ll1$. Note that since besides the condition of small thicknesses, it is also necessary to fulfill the condition of smoothness of the functions $\Psi(x,z)$ and $\varphi(x,z)$ on the scale $(q_0\cos\theta)^{-1}$, these conditions are compatible only for slightly anisotropic NLC, $\varepsilon_a\ll(2\pi/q_0L)\varepsilon_\perp$.

5. ESTIMATE OF THE CONTRIBUTION TO THE CHANGE IN THE THRESHOLD FIELD FROM PHOTOCONVERTED DIPOLE MOLECULES

We will assume that during photoconversion the NLC molecules acquire a constant electric dipole moment **d** (or its values becomes significantly greater than that of the molecules in the ground state). Since the concentration of the photoconverted molecules satisfies $c\ll1$, we will neglect the interaction between the constant dipoles of the photoconverted molecules, but we will take account of the interaction between the constant dipoles of the photoconverted molecules and the dipole moments they induce in the neighboring molecules of the NLC.

The value of the dipole induced in the mth molecule is obviously equal to

$$\mathbf{P}^m=\hat{\alpha}^m(\Omega_m)\sum_n c_n\mathbf{E}^n(\mathbf{r}_{mn},\Omega_n). \tag{27}$$

where $\hat{\alpha}^m(\Omega_m)$ is the polarizability tensor of the molecule, which depends on its orientation; $\mathbf{E}^n(\mathbf{r}_{mn},\Omega_n)$ is the electric field of the nth constant dipole at the position of the mth NLC molecule; Ω_n are the angular coordinates of the long axis of the molecule; $\mathbf{r}_{mn}=\mathbf{r}_m-\mathbf{r}_n$; $\mathbf{r}_n$ is the radius vector of the center of mass of the molecule; and we have $c_n=1$ if the photoconverted molecules is located at the point $\mathbf{r}_n$ and $c_n=0$ otherwise.

The interaction energy of the induced dipole (27) with the field of constant dipoles induced by it is equal to

$$U^m=-\frac{1}{2}\sum_{i,j}\alpha^m_{ij}(\Omega_m)\sum_n\sum_l c_nc_lE^n_i \\ \times(\mathbf{r}_{mn},\Omega_n)E^n_j(\mathbf{r}_{mn},\Omega_l). \tag{28}$$

It is necessary to average this expression over the distribution of long axes of the molecules relative to their favored orientation (the director) and over all possible spatial distributions of the photoconverted molecules in the vicinity of the mth molecule. Here we assume that the favored orientation of the long axes of the photoconverted molecules and the NLC molecules interacting with them are identical by virtue of the short range of the dipole field.

Neglecting the angular correlation of the long axes of the various molecules when averaging Eq. (28) (which corresponds to the mean field method, frequently used in the theory of nematic liquid crystals) and noting that $\langle\mathbf{E}(\mathbf{r}_{mn},\Omega_n)\rangle=0$, where $\langle...\rangle$ denotes an average over orientations, and $c_n^2=c_n$, after averaging we obtain

$$U^m=-\frac{1}{2}c(m)\sum_{i,j}a_{ij}\langle\alpha^m_{ij}(\Omega_m)\rangle, \tag{29}$$

where

$$a_{ij}=\sum_{n\neq m}\langle E^n_i(\mathbf{r}_{mn},\Omega_n)E^n_j(\mathbf{r}_{mn},\Omega_n)\rangle, \tag{30}$$

and $c(m)$ is the average concentration of the photoconverted molecules in the vicinity of the mth molecule.

Next, summing expression (29) over all the NLC molecules and replacing the sum by an integration over volume, we obtain

$$F_{PM}=\sum_m U^m=-\frac{1}{2}\sum_{i,j}\int c(\mathbf{r})a_{ij}\alpha_{ij}dV, \tag{31}$$

where α_{ij} is the polarizability tensor of the NLC.

The value of the electric dipole field of a photoconverted molecule depends on the direction of the director of the NLC at the position of the photoconverted molecule. Therefore, the macroscopic tensor a_{ij}, like the microscopic tensor α_{ij}, transforms like the product of the components of the director, $n_i n_j$. Since we neglected the variation of the director along the interaction distance of the dipole when we averaged over the orientations of the long axes (accounting for gradients of the director gives the dependence of Franck's elastic constants on the concentration of the photoconverted molecules), the product $n_i n_j$ refers to the same macroscopic point in space for both tensors. By virtue of this, the convolution $\Sigma_{i,j} a_{ij}\alpha_{ij}$ does not depend on the director and the expression for ΔU in Eq. (4) [compare Eqs. (31) and (4)] can be written in the form

$$\Delta U=\frac{1}{2}\sum_i a_{ii}\alpha_{ii}, \tag{32}$$

where the subscript i denotes the axes of the coordinate system associated with the director and, $\alpha_{ii}=1/4\pi[(\varepsilon_\perp^0-1)+\varepsilon_a^0\delta_{iz}]$; here we have written $\varepsilon_a^0=\varepsilon_\parallel^0-\varepsilon_\perp^0$, and $\varepsilon_\parallel^0$ and $\varepsilon_\perp^0$ are the principal values of the static dielectric tensor.

Substituting the value of the dipole field in formula (30) and performing the average, we obtain

$$\Delta U=\frac{1}{8\pi}\left\{(\varepsilon_\perp^0-1)(3\gamma+\gamma')\right.$$
$$\left.+\varepsilon_a^0\left[\gamma+\frac{2}{3}\gamma'(s+1/2)\cos^2\beta\right]\right\}, \tag{33}$$

where s is the orientational order parameter, β is the angle that the constant dipole makes with the long axis of the photoconverted molecule, and the positive quantities γ and γ' are of d^2/R^6, where R is the mean distance between the molecules.

Neglecting diffusion, the contribution of the photoconverted molecule to the threshold value of the field is given in Eqs. (9) and (10) by the term $\Delta\varepsilon=8\pi\tau\delta\kappa_a\Delta\mathrm{U}$, which renormalizes the anisotropy ε_a of the dielectric tensor (to estimate it, we will consider a field with a homogeneous intensity distribution, $\Delta q=0$). Note that the absorption coefficient is $\kappa(\mathrm{cm}^3/\mathrm{erg}\cdot\mathrm{s})=(c_0/4\pi)(\alpha/N\hbar\omega')$, where c_0 is the speed of light, N (cm^{-3}) is the density of the NLC molecules, α is the absorption coefficient in cm^{-1}, and ω is the frequency of absorption of the light. In the NLC MBBA, for example, photoconversion takes place when light with wavelength $\lambda=0.44$ μm is absorbed, with $\alpha=10$ cm^{-1} (Ref. 12) and $N=3\cdot 10^{21}$ cm^{-3}. Setting $\Delta U\sim 10^{-1}\gamma\varepsilon_\perp^0$, $\kappa_a\sim\kappa$, $R\simeq 10^{-7}$ cm, and $\delta=10^{-1}$ in accordance with Eq. (33), we obtain $|\Delta\varepsilon|\sim 10^{40}\varepsilon_\perp^0 d^2$. Thus, already at typical values of the dipole moment of the photoconverted molecules $d\sim 10^{-18}$ cgs units, the renormalizing terms are of order $10^4\varepsilon_\perp^0$ and dominate (since usually we have $\varepsilon_\perp^0>\varepsilon_a$). If $\kappa_a>0$ holds, then LIFT will take place and the threshold field will decrease by roughly two orders of magnitude from its value in the absence of photoconversion. The decrease of the LIFT threshold by the same order of magnitude when incident light is absorbed by dye molecules introduced into the NLC, observed experimentally in Refs. 4 and 5, may be explained by the formation of photoconverted dipole molecules.

In conclusion let us turn our attention to the fact that if the terms renormalizing ε_a dominate, then LIFT is possible in the NLC cell independent of the sign of the dielectric anisotropy ε_a if the anisotropy of the absorption coefficient for the light inducing the formation of photoconverted dipole molecules is positive ($\kappa_a>0$). Therefore it would be of interest as a check of the proposed model to carry out an experimental study of how the anthroquinone dyes mentioned above affect threshold LIFT in a NLC with $\varepsilon_a>0$, for which LIFT is impossible in the absence of photoconverted molecules.

This work was carried out with the financial support of the International Scientific Fund (Grant No. U58000) and the State Fund for Fundamental Research of the State Committee for Science and Technology of the Ukraine.

[1] A. S. Zolotko, V. F. Kitaeva, N. Kroo *et al.*, Pis'ma Zh. Éksp. Teor. Fiz. **32**, 170 (1980) [JETP Lett. **32**, 158 (1980)].

[2] B. Ya. Zel'dovich, N. V. Tabiryan, and Yu. S. Chilingaryan, Zh Éksp. Teor. Fiz. **81**, 72 (1981) [Sov. Phys. JETP **54**, 32 (1981)].

[3] B. Ya. Zel'dovich and N. V. Tabiryan, Zh. Éksp. Teor. Fiz. **82**, 1126 (1982) [Sov. Phys. JETP **55**, 656 (1982)].

[4] I. Janossy, A. D. Lloyd, B. S. Wherrett, Mol. Cryst. Liq. Cryst. **179**, 1 (1990).

[5] I. Janossy and A. D. Lloyd, Mol. Cryst. Liq. Cryst. **203**, 77 (1991).

[6] G. Abbate, P. Maddalena, L. Marrucci *et al.*, Mol. Cryst. Liq. Cryst. **223**, 11 (1992).

[7] W. E. Haas, K. F. Nelson, I. E. Adams *et al.*, J. Electrochem. Soc. **121**, 1167 (1974).

[8] S. G. Odulov, Yu. A. Reznikov. V. Yu. Reshetnyak, Mol. Cryst. Liq. Cryst. **207**, 43 (1991).

[9] I. P. Pinkevich, Yu. A. Reznikov, and V. Yu. Reshetnyak, Mol. Cryst. Liq. Cryst. **207**, 43 (1991).

[10] A. Rapini and M. Papolar, J. Phys. Colloids **30**, 54 (1969).

[11] M. Abramowitz, and I. A. Stegun, *Handbook of Mathematical Functions*, U.S. Government Printing Office, Washington, D.C. (1972).

[12] I. P. Pinkevich, Yu. A. Reznikov, and V. Yu. Reshetnyak, Ukr. Fiz. Zh. **32**, 1216 (1987).

Translated by Paul F. Schippnick