Light-induced second-order phase transition in a spatially bounded region of a nematic liquid crystal

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The dynamics of reorientation and relaxation of the director field of a homeotropic OCBP crystal in a narrow light beam is studied experimentally and theoretically. Specific properties are observed.

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1. The state of a nematic liquid crystal (NLC), characterized by the director field, can vary under the action of external fields. Under certain conditions, reorientation of the direction can occur following the laws governing a seond-order phase transition[1] (Freedericksz transitions[2]). Freedericksz transitions were first observed in light fields in Ref. 3. However, strictly speaking, the appearance of a phase transition was not studied in Ref. 3.

In this paper, we present the results of the first study of the dynamics of reorientation and relaxation of the director field in a homeotropic OCBP crystal in a narrow

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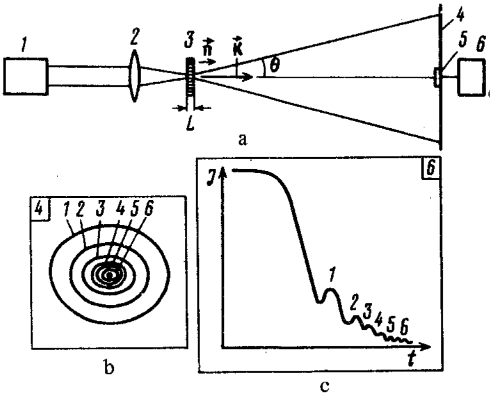

FIG. 1. (a) Diagram of the experimental setup: 1) laser; 2) focusing lens; 3) NLC; 4) screen; 5) photodiode; 6) automatic plotter. (b) Aberrational pattern on the screen. (c) Characteristic temporal dependence of the intensity at the center of the aberrational pattern with reorientation of the director.

light beam (the transverse size of the beam, $w_0 \lesssim L$, is the thickness of the crystal). The case examined by us differs from that studied in Ref. 4, wherein the investigations were carried out in wide light beams ($w_0 \gg L$) and where the theory of the Freedericksz transition in spatially homogeneous fields is valid.[5]

2. The experimental arrangement is shown in Fig. 1a. An argon laser beam 1, passing through the focusing lens 2 (it serves to change the cross section of the beam w_0), was incident normally on the cell 3 containing the OCBP crystal with thickness $L = 150\ \mu$m. A screen 4, on which the aberrational structure was observed (Fig. 1b), was placed beyond the crystal in the transmitted beam at a distance $d \sim 2$ m. A photodiode 5, whose signal was recorded on an automatic plotter 6, was placed near the screen. A typical trace is shown in Fig. 1c.

The intensity of the laser beam with the development of self-focusing in NLC, as can be seen from Fig. 1c, oscillates. The appearance of each ring in the transverse structure of the beam is accompanied by an oscillation in the intensity along its axis. The number of oscillations N (or the number of rings) is related to the maximum angle of rotation of the director ψ_m by the relation[6]

$$N(t) = \frac{L}{2\lambda}(n_{\parallel} - n_{\perp})\,\psi_m^2(t), \tag{1}$$

where λ is the wavelength, $n_{\parallel} = \sqrt{\epsilon_{\perp}}$, $n_{\perp} = \sqrt{\epsilon_{\parallel}}$, $\epsilon_{\parallel}$, and $\epsilon_{\perp}$ are the components of the dielectric constant tensor.

3. The dynamics of the reorientation of the director $\psi(t)$ was studied from the development of oscillations in the intensity $N(t)$ after a sharp increase in beam power from some initial value $P_i < P_{\text{th}}$ to a magnitude $P_{\text{st}} > P_{\text{th}}$ (the threshold power for the phase transition P_{th} was 85 mW with a beam radius of $w_0 = 77\ \mu$m and 47 mW with $w_0 = 42\ \mu$m). Analysis of traces on the automatic plotter showed that the angle of rotation of the director increases exponentially with time. In the initial region of reorientation: $\psi_m^2(t) \sim N \sim \exp(\chi_{OR}t)$ (χ_{OR} is the rate of reorientation). The experiments showed that the quantity χ_{OR} does not depend on the initial weak field $E_i(P_i)$. The dependence of the rate of reorientation on the beam radius and electric field intensity on the beam axis E is shown in Fig. 2 [the positive branch of the curve $\chi(E)$]. We see

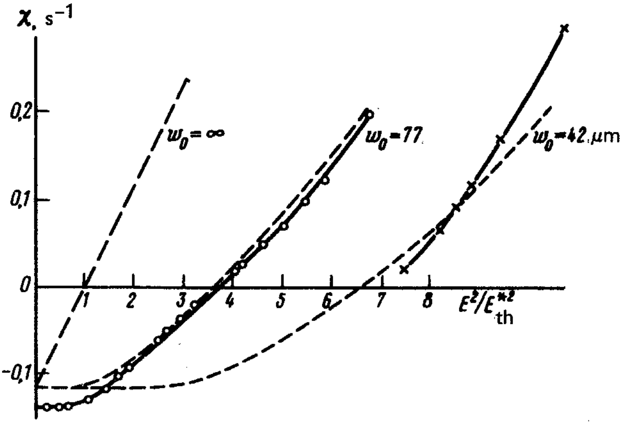

FIG. 2. Experimental points (0,χ) and theoretical (dashed curves) dependences of the reorientation and relaxation rates as a function of beam width and electric field intensity E.

that the threshold value of the electric field increases with decreasing constriction, while the reorientation process slows down.

Relaxation processes were observed after a sharp decrease in beam power from $P_{st} > P_{th}$ to $P_f < P_{th}$. In addition, the magnitude of the final power P_f varied from 5 mW to the threshold magnitude P_{th} (the beam with power P_f also played the role of a probing beam). After removing the strong field, the angle of rotation of the direction decreased exponentially with time along the linear section: $\psi_m^2(t) \sim N \sim \exp(-\chi_R t)$. Figure 2 shows the experimental dependence of the relaxation rate (orientational branch of the curve $\chi(E)$) from the final intensity of the light field $0 < E_f < E_{th}$. Here, we discovered a new important property of the phase transition in NLC: In the low-field region $0 < E_f < E_{cr}$, the rate of relaxation χ_R is constant (see Fig. 2). This effect is related to the appearance of the transverse diffusion of the director field and occurs only in narrow light beams. When the final field approached the threshold field $E_f \to E_{th}$, the relaxation process slowed down sharply, $\chi_R \to 0$ (this, in general, is characteristic of all phase transitions).

4. The characteristics of reorientation and relaxation of the director observed in the experiment can be explained within the scope of the theory of the Freedricksz transition, taking into account the spatially finite light beam. The nonstationary equation that describes the behavior in the presence of the light field of a Gaussian beam has the form

$$\gamma_1 \frac{\partial \psi}{\partial t} = \Delta \psi + \frac{1}{\xi^2} e^{-2\rho^2/w_0^2} \sin\psi \cos\psi , \tag{2}$$

where $\gamma_1 = -\lambda_1/K$, λ_1 is the viscosity, K is Frank's constant, ρ is the distance to the beam axis, and $1/\xi^2 = 2\Delta\epsilon\epsilon_\perp^{1/2} P/\pi c K \epsilon_\parallel W_0^2$.

For small angles of rotation, $\psi^2 \ll 1$, and on the linear section, $\sin\psi\cos\psi \simeq \psi$.

The particular solution of Eq. (2), taking into account the fact that on the walls (at $Z = 0$ and $Z = L$) $\psi = 0$, can be written in the form

$$\psi \sim e^{\chi t/2} \psi'(\rho) \sin \frac{\pi Z}{L}.$$

In wide laser beams ($w_0 \gg L$, $\partial^2\psi/\partial x^2 + \partial^2\psi/\partial y^2 = 0$), the rate of reorientation and relaxation is expressed by the following well-known equation[7]:

$$\chi_{\text{homogen.}} = \frac{2\pi^2}{\gamma_1 L^2} \left(\frac{E^2}{E_{th}^{*2}} - 1 \right), \tag{3}$$

where $E_{\text{th}}^* = (\pi/L)\sqrt{8\pi K \epsilon_{\parallel}}/\epsilon_{\perp} \Delta\epsilon$ is the threshold field for the Freedericksz transition in a homogeneous light field. Because of transverse diffusion of the director field, a spectrum of characteristic values of χ, containing a discrete set and a continuum, appears in narrow beams $w_0 \ll L$. The transverse mode of lowest order, which can be approximately described by a Gaussian distribution $\psi'(\rho) + e^{-\rho^2/a^2}$, is the largest mode (it has the highest increment and smallest decrement). Applying Ritz' variational method, we find

$$\chi = \frac{2}{\gamma_1} \frac{\pi^2}{L^2} \left\{ (1+g)^2 \left(\frac{E}{E_{\text{th}}} - \frac{g}{1+g} \right)^2 - 1 \right\} (E > E_c) \tag{4}$$

$$a^2 = \frac{w_0^2}{E/E_c - 1},$$

where $E_{\text{th}} = E_{\text{th}}^*(1+g)$ is the threshold power of the transition in narrow beams, $g = \sqrt{2}L/\pi w_0$, and $E_c = E_{\text{th}} g/(1+g)$.

It follows from Eqs. (4) that for $E > E_{\text{th}}$, $\chi > 0$, which corresponds to reorientation, while for $E < E_{\text{th}}$, $\chi < 0$, which corresponds to relaxation. The finiteness of the beam gives rise to the following two effects. First, it increases the transition threshold, decreases the rate of reorientation, and increases the relaxation rate. Second, when the final field intensity E_f decreases to the critical magnitude E_{cr}, the transverse dimension of the mode increases indefinitely ($a \to \infty$) and for $E < E_{\text{cr}}$, the discrete spectrum of χ disappears altogether. In the low power region (weak fields), $0 < E < E_{\text{cr}}$, the relaxation rate does not depend on the presence of the final field $\chi_R(0) = (2/\gamma_1)(\pi^2/L^2)$.

The theoretical dependences (4) of the experimental parameters are represented in Fig. 2 by the dashed curves. We see that they describe well the results of the experiments performed.

5. Thus, in this paper we have presented a theory of the orientational second-order phase transition (Freedericksz transition) in narrow light beams. This theory explains the experimental data. It is important to measure the reorientation and relaxation times, not only from the purely physical point of view but also in order to obtain information on the properties of NLC (in particular, on the viscosity).

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