Light-induced Fredericks transition in a liquid crystal

S. R. Galstyan, O. V. Garibyan, N. V. Tabiryan, and Yu. S. Chilingaryan
Erevan State University

(Submitted 4 February 1981)

Pis'ma Zh. Eksp. Teor. Fiz. **33**, No. 9, 454–458 (5 May 1981)

The Fredericks transition in a homeotropically oriented layer of a nematic liquid crystal (NLC) due to the action of continuous laser radiation has been investigated. The measured value of the threshold power is in good agreement with the given theoretical estimate. The dynamics of the observed effect and the role of thermal effects have been investigated.

PACS numbers: 64.70.Ew, 79.20.Ds

1. A giant, cubic, optical nonlinearity of liquid crystals, which is caused by a reorientation of the director in a light field, was recently predicted[1-7] and observed experimentally.[5,7] Subsequent experimental work[8-10] confirmed the presence of giant optical nonlinearities for a whole series of specific NLC and experimental geometries. In the present paper we present the results of an experimental investigation of the Fredericks transition in a homeotropically oriented MBBA layer in the continuous-radiation field of an argon laser. Formulas are given for calculating the threshold power and for describing the evolution of the process with time.

2. Reorientation of the director of a homeotropically oriented NLC due to the

action of linearly polarized laser radiation incident normally on the cell plates is possible only when a certain wave-power threshold is reached.

To estimate the threshold power, we write the free energy of a unit volume of the NLC in the one-constant approximation[11]

$$F = \frac{1}{2} K \left(\frac{\partial n_\alpha}{\partial x_\beta} \right)^2 - \frac{\epsilon_a}{16\pi} (\mathbf{n}\mathbf{E})(\mathbf{n}\mathbf{E}^*). \tag{1}$$

The following symbols were introduced in Eq. (1): K (erg/cm) is the Franck constant, **n** is the unit vector of the director the unperturbed direction of the director coincides with the z axis, **E** is the complex amplitude of the monochromatic light field, and $\epsilon_a = \epsilon_\| - \epsilon_\perp$ is the anisotropy of the dielectric constant of the NLC at the light frequency. After setting $\mathbf{n} = \{n_x, n_z\} = \{\sin\theta(\mathbf{r}), \cos\theta(\mathbf{r})\}$ and assuming that $\theta(\mathbf{r}) = \theta_m(x,y)\sin(\pi z/L)$ and $|\theta_m| \ll 1$, which is valid near the Fredericks threshold, we obtain from Eq. (1)

$$\int F d^3 \mathbf{r} \backsim a_\perp^2 L \theta_m^2 \left\{ K \left[\frac{\pi^2}{L^2} + \frac{2}{a_\perp^2} \right] - \frac{\epsilon_a |E|^2}{8\pi} \right\}. \tag{2}$$

Here $a_\perp$ is the transverse dimension of the beam and L is the cell thickness. The unperturbed state $\theta_m \equiv 0$ is stable when

$$|E|^2 < |E|^2_{\mathrm{Fr}} = \frac{8\pi K}{\epsilon_a} \left(\frac{\pi^2}{L^2} + \frac{2}{a_\perp^2} \right). \tag{3}$$

Thus, the threshold power density is equal to $S_{\mathrm{Fr}} = c\epsilon_\perp^{1/2} |E|^2_{\mathrm{Fr}}/8\pi$. A detailed theoretical investigation of the light-induced Fredericks transition is given in Ref. 12, where more precise expressions are given for determining the threshold power for different intensity profiles of the incident beam.

3. The following experiment was performed. The radiation of an argon laser ($\lambda = 0.488$ μm) was focused by a lens with a focal length $f = 5$ cm in a homeotropically oriented 50-μm-thick MBBA layer at room temperature. The wave vector of the radiation was directed along the NLC director. The Fredericks transition was detected in the far zone from the change in divergence of the radiation that passed through the NLC layer.

At radiation power $P < 100$ mW incident on the NLC no change in divergence was detected. The observation time amounted to ten minutes. A sharp increase in divergence was observed with a further increase in the power. The dependence of the divergence ϕ on the radiation power P incident on the NLC is shown in Fig. 1a. As seen in this figure, the threshold of the Fredericks transition is reached at a radiation-power density $S_{\mathrm{Fr}} = 2.6$ kW/cm^2, consistent with the integrated radiation power $P = 100$ mW. For the LC parameters[13] $\epsilon_\| = 3.06$, $\epsilon_\perp = 2.75$, $K = 6.8 \times 10^{-7}$ dyne, and $L = 50$ μm the threshold value of the power density, calculated from Eq. (3), is $S_{\mathrm{Fr}} = 2.4$ kW/cm^2.

To determine the role of thermal effects, we repeated the measurements for cir-

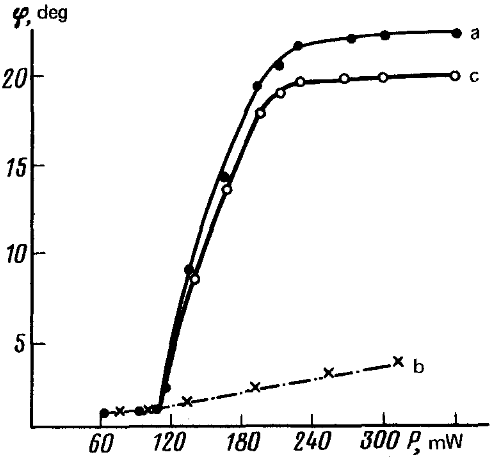

FIG. 1. Dependences of the divergence angle ϕ on the incident radiation power. (a) Curve for linearly polarized light, (b) curve for cirularly polarized light, (c) curve determined only by the orientational of nonlinearity (the difference between plots a and b).

cular polarization of the laser radiation. Since the direction of the director cannot follow the variation of the direction of wave polarization at the light frequency, the change in radiation divergence in this case is caused by thermal self-focusing (since $\partial\epsilon_{\perp}/\partial T > 0$, self-focusing must occur[14]).

The dependence of radiation divergence on the power of the incident wave, which is governed solely by the orientation mechanism of interaction, is shown in Fig. 1c.

Periodic variations of the radiation divergence with time, which are similar to those described in Ref. 9, were detected in the experiment. Such pulsations were also observed when circularly polarized light was incident on the cell. Our investigation showed that they are irregular – their period and number depend on the specific area of the LC on which the radiation is incident. We were able to simulate analogous pulsations by means of an absorbing-liquid cell with a thickness of ~0.5 cm, to which carbon powder was added (particle size $d \lesssim 50$ μm). This shows that the pulsations may be caused by heating and subsequent convection of comparatively large foreign particles in the NLC cell.

We have also investigated the dynamics of variation of the divergence of the radiation that passed through the NLC layer. Figures 2 and 3 show, respectively, the dependences of the LC response time and the transient time of the process on the power of the incident, linearly polarized light.

Since the Fredericks transition is a second-order phase transition, the transient time at slightly higher values than the threshold is described by the formula[15] $\tau_t = C_t/(P - P_{\mathrm{Fr}})$, where C_t is a constant. As seeen in Fig. 2, this function is a good approximation of the experimental curve when $C_t = 1.8$ J.

The dependence of the response time τ_r on the wave power is approximated well by the formula

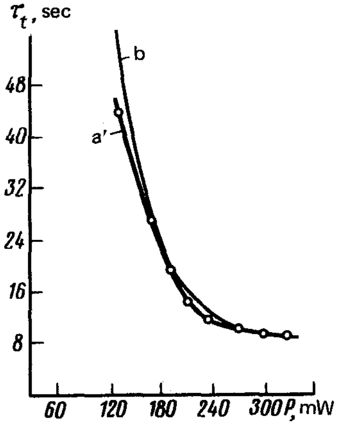

FIG. 2. Dependence of the process transient time τ_t on the incident radiation power. (a) Experimental curve, (b) theoretical curve.

$$\tau_r = \tau_{nr} \exp\left\{ \frac{c_r}{P - P_{\mathrm{Fr}}} \right\} \tag{4}$$

when the constants have the values $\tau_{nr} = 1.5$ sec and $c_r = 184$ mW. Such a dependence can be interpreted qualitatively as follows. If the light-wave power $P > P_{\mathrm{Fr}}$, then the state of the system with $\theta \equiv 0$ is an unstable-equilibrium state, and thermal fluctuations remove it from this state. At the same time, the higher the power of

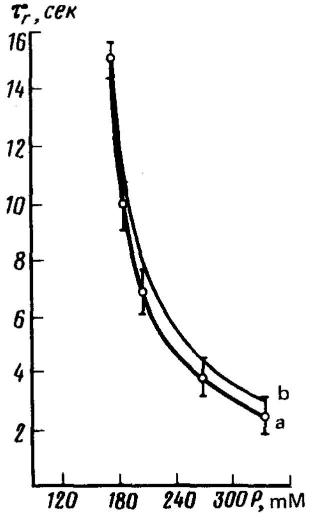

FIG. 3. Dependence of process response time on the incident radiation power. (a) Experimental curve, (b) curve determined from Eq. (4).

the light field, the smaller are the fluctuational deviations of the director from the equilibrium value at which the field is capable of carrying the director. We assume that the relationship between the amplitude $\delta\theta$ of the director fluctuations and the power P of the light field that is necessary to carry it along can be represented in the form $P - P_{\mathrm{Fr}} = \mathrm{const}/(\delta\theta)^2$. Thus, after substituting this expression in the probability distribution function $f(\delta\theta) \propto \exp\{-(\delta\theta)^2/2<(\delta\theta)^2>\}$ and taking into account that $\tau_r \sim f^{-1}$, we obtain Eq. (4).

We note that if the power is high $P - P_{\mathrm{Fr}} \gg C_r$, the P dependence of the quantity τ_{nr} cannot be ignored in Eq. (4).

Thus, we have identified and analyzed certain characteristic aspects of the light-induced Fredericks transition in an NLC.

The authors thank R. S. Akopyan, N. N. Badalyan, and B. Ya. Zel'dovich for a discussion of the results.

1. B. Ya. Zel'dovich and N. V. Tabiryan, Pis'ma Zh. Eksp. Teor. Fiz. **30**, 510 (1979) [JETP Lett. **30**, 478 (1979)].
2. B. Ya. Zel'dovich and N. V. Tabiryan, Preprint FIAN No. 61, 1980.
3. B. Ya. Zel'dovich and N. V. Tabiryan, Preprint FIAN No. 62, 1980.
4. B. Ya. Zel'dovich and N. V. Tabiryan, Preprint FIAN No. 63, 1980.
5. B. Ya. Zel'dovich, N. F. Pilipetskiĭ, A. V. Sukhov, and N. V. Tabiryan, Pis'ma Zh. Eksp. Teor. Fiz. **31**, 287 (1980) [JETP Lett. **31**, 263 (1980)].
6. B. Ya. Zel'dovich and N. V. Tabiryan, Kvantovaya Elektron. **7**, 770 (1980) [Sov. J. Quantum Electron. **10**, 440 (1980)].
7. N. F. Pilipetskiĭ, A. V. Sukhov, N. V. Tabiryan, and B. Ya. Zel'dovich, Opt. Commun. (to be published).
8. V. B. Pakhalov, A. S. Tumasyan, and Yu. S. Chilingaryan, Tezisy dokladov X Vsesoyuznoĭ konferentsii po Kogerentnoĭ i Nelineĭnoĭ optike (Abstracts of Papers of the 10th All-Union Conference on Coherent and Nonlinear Optics), Kiev, October 14–17, 1980, Part 1, p. 18.
9. A. S. Zolot'ko, V. F. Kitaeva, E. Kroo, N. N. Sobolev, and L. Chillag, Pis'ma Zh. Eksp. Teor. Fiz. **32**, 170 (1980) [JETP Lett. **32**, 158 (1980)].
10. S. M. Arakelyan, S. R. Galstyan, O. V. Garibyan, A. S. Karayan, and Yu. S. Chilingaryan, Pis'ma Zh. Eksp. Teor. Fiz. **32**, 561 (1980) [JETP Lett. **32**, 543 (1980)].
11. P. de Gennes, Physics of Liquid Crystals (Russ. Transl., Mir. Moscow, 1977).
12. B. Ya. Zel'dovich, N. V. Tabiryan, and Yu. S. Chilingaryan, submitted to Zh. Eksp. Teor. Fiz. **81**, No. 7 (1981) [Sov. Phys. JETP **54**, (to be published)].
13. L. M. Blinov, Elektro- i magnitooptika zhidkikh kristallov (Electro- and Magneto-optics of Liquid Crystals), Nauka, Moscow, 1978.
14. V. Volterra and E. Wiener-Avnear, Opt. Commun. **12**, 194 (1974).
15. E. M. Lifshitz and L. P. Pitaevskiĭ, Fizicheskaya kinetika (Physical Kinetics), Nauka, Moscow, 1979.

Translated by Eugene R. Heath
Edited by S. J. Amoretty