Photoinduced Fréedericksz transition in an oblique ordinary wave

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A linearized theory is derived for the photoinduced Fréedericksz transition in the field of an oblique ordinary wave for a broad beam. The threshold for the transition is found as a function of the angle of incidence from this theory. Experiments carried out on the transition in a homeotropic cell filled with the nematic liquid crystal 5SV, in a layer 135 μm thick, with light at $\lambda = 0.4880\,\mu$m confirm the theoretical predictions. The threshold rises by a factor of three at an angle of incidence of 0.15 rad.

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When an extraordinary light wave is incident obliquely on the director of a nematic liquid crystal, a threshold-free Fréedericksz effect is observed: a change in the orientation of the director proportional to the light intensity.[1–3] If the light is incident strictly along the direction of the director, there is a threshold in the light intensity for this reorientation; this effect is called the "photoinduced Fréedericksz transition" in the narrow sense of the term.[4–6] This letter reports a theoretical and experimental study of the threshold photoinduced Fréedericksz effect for the case in which an ordinary wave is incident obliquely on a homeotropic cell with a nematic liquid crystal.

1. Theory. We assume that the wave vector **k** of the incident *o*-type wave $\mathbf{E} = \mathbf{e}_y E_0$ makes with the z axis (i.e., with the unperturbed direction of the director) an angle $\alpha = \alpha_{\rm inc}/n_\perp \ll 1$; i.e., $\mathbf{k}_0 = k_0[(1-\alpha^2/2)\mathbf{e}_z] + \alpha \mathbf{e}_x$, $k_0 = (\omega n_\perp/c)$, (Fig. 1). As a result of the interaction with the perturbations of the director, the wave can acquire a weak extraordinary component, so that the field in the medium can be written

$$\mathbf{E}e^{-i\omega t} = \exp[-i\omega t + ik_0\alpha x + ik_0 z(1 - \alpha^2/2)\{\mathbf{e}_y E_y + (\mathbf{e}_x - \alpha \frac{\epsilon_\perp}{\epsilon_\parallel}\mathbf{e}_z)E_x\}. \quad (1)$$

If the perturbation of the director $(\mathbf{n} - \mathbf{n}_0)$ is uniform in the (x,y) plane (if the light beam is broad), then we have $\mathbf{n} \approx \mathbf{e}_z + \mathbf{e}_x\theta_x(z,t) + e_y\theta_y(z,t)$. Equations for the slow field amplitudes E_x and E_y can be derived from Maxwell's equations; in the case $|\vec{\theta}| \ll \alpha$ these equations are

$$\frac{dE_y}{dz} = -i\theta_y \alpha^{-1}\mu E_x; \quad \frac{dE_x}{dz} = i\mu E_x - i\theta_y \alpha^{-1}\mu E_y. \quad (2)$$

Here $\mu = \mu(\alpha) = 0.5k_0\alpha^2(1 - n_\perp^2/n_\parallel{}^2)$ is the difference between the z components of the wave vector of the o and e waves at a fixed value of α. If the $\vec{\theta}(z)$ change occurs over a scale $\Delta z \gg \mu^{-1}$, then the o wave (E_y) acquires an additional component E_x,

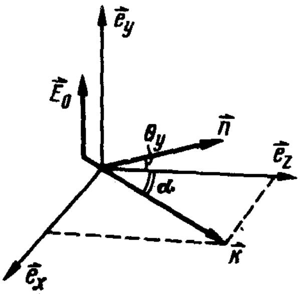

FIG. 1.

which is determined by the second of Eqs. (2) with dE_x/dz ignored; i.e., $E_x(z)\approx\theta_y(z)\alpha^{-}E_y$. This situation corresponds to conservation of the *o* nature of the wave during an adiabatic tracking of the local orientation of the director.

Equations for the perturbation of the director are found by a variational principle; in the approximation $\epsilon_a/\epsilon_\perp \ll 1, |\vec{\theta}| \ll \alpha \ll 1, E_x \sim |\vec{\theta}|E_y$ they are

$$\gamma\frac{\partial\theta_y}{\partial t} - K_{33}\frac{\partial^2\theta_y}{\partial z^2} = \frac{\epsilon_\perp}{\epsilon_\parallel}\frac{\epsilon_a}{8\pi}[\,|E_y|^2\theta_y - 0,5\,\alpha\,(E_xE_y^* + E_x^*E_y)\,]. \qquad (3)$$

Here $\epsilon_a = n_\parallel^2 - n_\perp^2$, K_{33} is the Frank constant, and γ (in poises) is the orientational viscosity constant. The equation for θ_x in the same approximation is $\gamma\partial\theta_x/\partial t = K_{33}\partial^2\theta_x/\partial z^2$; i.e., the field is not filled. A hard homeotropic orientation at the walls corresponds to the conditions $\vec{\theta}(z=0,t) = \vec{\theta}(z=L,t) = 0$.

In the problem of the threshold for the photoinduced Fréedericksz transition, we can write $E_y = E_0 = \text{const}$ to an accuracy sufficient for our purposes. If we describe the component $E_z(z)$ by an expression corresponding to the adiabatic approximation, $E_x = \theta_y(z)E_y/\alpha$, we find that the right side of Eq. (3) vanishes. In other words, the *o* wave does not cause orientational effects in the adiabatic approximation. The second of Eqs. (2) can also be integrated exactly for an arbitrary $\theta_y(z)$. Substitution of the result into (3) under the condition $E_x(z=0)=0$ yields

$$\frac{\partial\theta_y}{\partial t} = \Gamma_0\left(\frac{L}{\pi}\right)^2\left\{\frac{\partial^2\theta_y}{\partial z^2} + \left(\frac{\pi}{L}\right)^2\rho\left[\theta_y(z,t) - \mu\int_0^z dz'\,\theta_y(z',t)\sin\mu(z-z')\right]\right\}. \qquad (4)$$

Here $\Gamma_0 = K_{33}(\pi/L)^2\gamma^{-1}$ is the damping constant for the lower-order mode, $\delta\theta \sim \sin\pi z/L$, $[\Gamma_0] = \mathrm{s}^{-1}$, and $\rho = \epsilon_\perp/\epsilon_\parallel|E_0|^2L^2/8\pi^3K_{33}$ is the extent to which the intensity of the incident wave exceeds the value which would be the threshold value in the case of normal incidence.

An equation like (4) was derived and analyzed in Ref. 6 in connection with the related problem of the photoinduced Fréedericksz transition in the field of an *o* wave for a planar cell, with $\mu = \omega(n_\parallel - n_\perp)/c$. It is not difficult to see that if $\mu L \lesssim \pi$ the threshold for the photoinduced Fréedericksz transition determined from the appearance of a solution of Eq. (4), which grows exponentially over time, can be found by

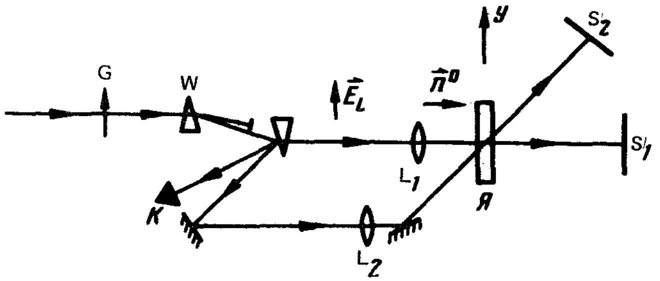

FIG. 2. Experimental arrangement (explained in the text proper). The rotation of the cell through the angle α_{exc} occurs around the y axis, which lies in the plane of the figure and which coincides with the direction of the polarization of the main laser beam.

perturbation theory. This threshold is[6]

$$\rho_{th} \approx 1 + (\mu L/\pi)^2 + O((\mu L/\pi)^4). \qquad (5)$$

Arakelyan *et al.*[7] have calculated the threshold from Eq. (4) without resorting to perturbation theory, but for our comparison with experiment our expression (5) is sufficient.

2. Experimental procedure and results. The arrangement in Fig. 2 was used for an experimental study of the photoinduced Fréedericksz transition for the case of an obliquely incident *o* wave. The beam from a single-mode argon laser with a power $\sim$600 mW passed through a system of polarizers consisting of a Glan prism G and a birefringent wedge LW (for continuous adjustment of the power) and was focused by lens $L_1(f = 16$ cm) into a homeotropic cell holding the nematic liquid crystal 5SV, 135 μm thick (this thickness was measured with the cell empty, through observation of the Fabry-Perot rings which appear when the cell is illuminated by a diverging beam). After transmission through the cell, the beam is monitored visually on screen S_1. At the same time, the focal region of this beam is probed by another beam, focused by lens L_2; this probing beam is incident obliquely with respect to the director. It is thus possible to determine the threshold for the photoinduced Fréedericksz transition very accurately, since the phase shift $\delta\phi_3$ of the oblique-polarization beam is far more sensitive to slight deviations of the director δn_y than the phase shift of the main beam, $\delta\phi$ $[\delta\phi_3 \sim \delta n_y$, while $\delta\phi \sim (\delta n_y)^2]$. Correspondingly, the probe beam acquires an annular structure at power levels at which the angular divergence of the main beam is still not changing, within the experimental errors. In the experiments we studied the threshold for the photoinduced Fréedericksz transition (working from the appearance of 0.5 of a ring in the probing beam) and the behavior above the threshold for various angles of incidence of the main beam on the sample, α_{inc} (α_{inc} is the angle through which the cell is rotated around the y axis, so that the main beam always corresponds to a polarization of the *o* type). It should be noted that the diameter of the focal region of the main beam is $a = FWe^{-2}M = 180\ \mu$m, or greater than the thickness of the cell (here M corresponds to the intensity). Figure 3a shows the dependence of the threshold for the photoinduced Fréedericksz transition on the angle of incidence. It can be seen from Fig. 3b that this dependence corresponds very accurately to $P_0(1 + \xi^2\alpha^4)$, where P_0 is the threshold for $\alpha = 0$. Shown in the same figure is a plot of $P_0[1 + (\mu L/\pi)^2_{expt}]$,

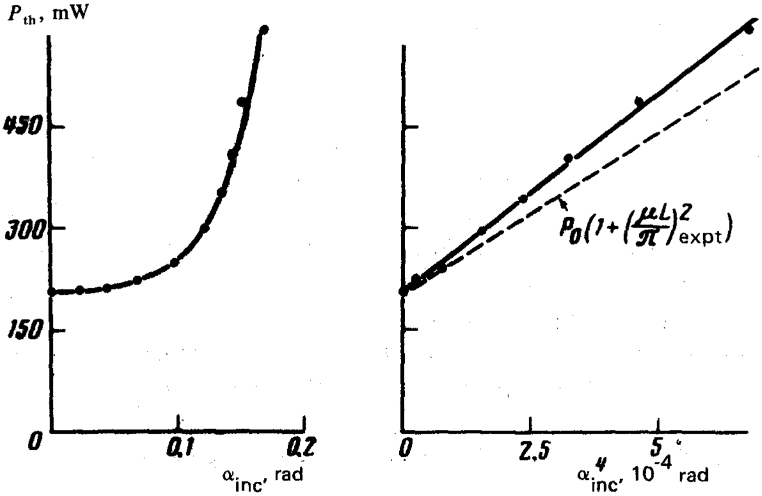

FIG. 3.

where $(\mu L/\pi)$ is measured directly, from the number of fringes of the conoscopic pattern corresponding to the given value of α. Figure 4 shows the time evolution of the number of self-focused rings in the main beam at a fixed power $P \sim 300$ mW and various angles of incidence α. We see that as the angle is increased, the number of rings goes through an oscillation over time ("beat"); the scale time of this oscillation decreases rapidly with increasing α. This oscillation over time during the excitation of a photoinduced Fréedericksz transition by an o wave in a homeotropic cell was noted previously.[4] The reasons for this oscillaiton have not been studied.

Significantly, by using an obliquely incident probing beam we can determine the *sign* of the θ_y which arises in the Fréedericksz transition. Specifically, if θ_y is positive

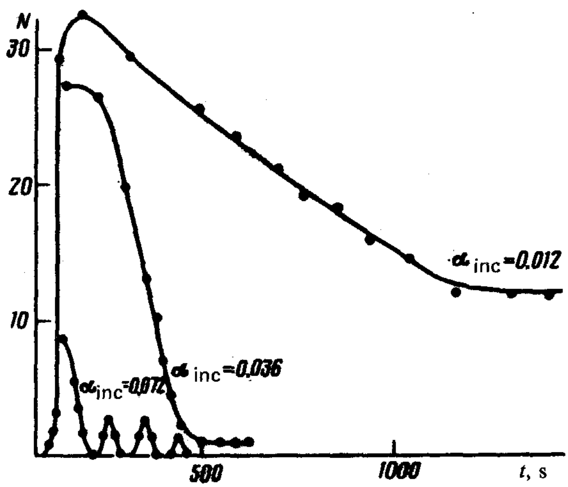

FIG. 4.

(in the arrangement of Fig. 2), the probing beam initially acquires a system of defocused rings, whose number rises to a maximum $N = 7$ with increasing power and then remains constant. Later, against the background of this first system of rings, a second system of *focused* rings appears. If θ_y is *negative*, only the system of *focused* rings appears. The sign found for θ_y by this method changes in an irregular fashion in different instances of the Fréedericksz transition as we switch from one part of the cell to another and also if we stay in the same part of the cell. These changes in the sign correspond to the way in which the threshold photoinduced Fréedericksz transition evolves from the level of the initial fluctuations.

In summary, we have derived a linearized theory for the photoinduced Fréedericksz transition in the field of an ordinary wave. The present experiments verify the theory and point to some extremely interesting qualitative aspects of the nonlinear stage.

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