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TWO-BEAM MIRRORLESS OPTICAL BISTABILITY IN NEMATIC LIQUID CRYSTAL FILM

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Abstract We report here the first experimental observation of first-order optical Freedericksz transitions, which arise when two laser beams interact incoherently in a homeotropically aligned nematic film. Experimental evidence for hysteresis cycles (bistability) is given. The comparison with the theory shows that the two transitions in a cycle have an essentially different character.

INTRODUCTION

In the last years a renewed and growing attention has been devoted to nonlinear optical effects in liquid crystals (LC). This is due to the recent theoretical[1] and experimental[2] demonstration of the extraordinary large optical nonlinearity shown by LC in the nematic phase, owing to the laser induced reorientation of the molecular director. Concomitant nonlinear optical effects, such as self-focusing, degenerate four-wave mixing, self-phase modulation are easily observed even with cw lasers. For some experimental geometries, it is found that the molecular reorientation can be effectively induced only if the light intensity exceeds

a characteristic threshold[3] (the optical Freedericksz threshold). This effect has no analogue in traditional nonlinear optics.

In this paper we report the experimental observation of a new nonlinear effect: mirrorless, hence intrinsic, optical bistability arising when two p-polarized light beams interact incoherently in a homeotropically aligned nematic LC film. This effect is observed when both beams impinge upon the sample at oblique incidence from opposite sides with respect to the normal to the film faces. In this geometry the two beams produce opposite torques on the LC molecules. The torques exactly compensate when the intensity I_1 and I_2 of the beams are related by

$$I_1 \sin(i_1) = I_2 \sin(i_2) \qquad (1)$$

i_1 and i_2 being the incidence angles of the beams in air. When the equilibrium condition (1) is satisfied, the sample remains undistorted and the tilt angle Θ between the molecular director and the sample normal is zero, provided the homeotropic configuration is stable. It can be shown, however, (see theory below) that the undistorted configuration becomes unstable, even though Eq. (1) is satisfied, when the beam intensity I_1 or I_2 exceeds a threshold value I_{th}. Consider now the case $I_1 > I_{th}$ and $I_2 = 0$. Then, the molecular director will be oriented towards the beam I_1 and the molecular tilt angle is, say, positive. If I_2 is now increased to a value much greater than I_1, the director will turn, finally, towards I_2, passing from a positive

to a negative tilt angle. This transition, however, must be necessarily discontinuous (first-order), since the state $\Theta=0$ is already unstable (because $I_1>I_{th}$). Once the system has switched to negative values of Θ, the intensity I_2 must be greatly reduced before I_1 dominates again and Θ passes, again with a first-order transition, to positive values. This results in a characteristic hysteresis loop and in a bistable behaviour. It should be stressed that, since the first observation made by Freedericksz in 1927, all observed Freedericksz transitions in nematic LC were second-order. This is, therefore, to our knowledge, the first evidence of first-order optical transition in the nematic mesophase.

THEORY

The local molecular orientation of the LC is described by the director $\underline{n}=(\sin\Theta,0,\cos\Theta)$, where Θ is the tilt angle between the director and the z axis along the sample normal. Variation of the total free-energy (consisting of the elastic deformation energy and of the optical field energy for oblique incidence and p-polarization) leads to the Euler equation ($\Theta'=d\Theta/dz$)

$$\left\{1 - k\sin^2(\Theta)\right\}\Theta'' - k\sin(\Theta)\cos(\Theta)\Theta'^2 + M_{Opt}(\Theta)/k_{33}=0 \qquad (2)$$

for the steady-state tilt angle $\Theta(z)$ in the sample. In Eq.2 $k=1-(k_{11}/k_{33})$, k_{11} and k_{33} being the elastic constants for splay and bend, respectively, and M_{Opt} is the total optical

torque produced by the two beams. We assume that the beams act incoherently on the sample, i.e., that the total optical torque is given by

$$M_{Opt}(\Theta) = M(\Theta;I_1,i_1) + M(\Theta;I_2,i_2), \tag{3}$$

any interference term being neglected. This can be either achieved by using different laser sources, or by making the difference in the optical path larger than a coherence length. The torque $M(\Theta;I,i)$ produced by a p-polarized plane wave of intensity I, impinging upon the sample at incidence angle i, can be obtained from Maxwell's equations in the slowly-varying envelope approximation as

$$M(\Theta;I,i) = \left\{(n_e^2 - n_o^2)I/(cE_{33}^2)\right\}\left\{n_o n_e (E_{33} - 2s^2)(E_{33} - s^2)^{-1/2}\sin(\Theta)\cos(\Theta) - s(n_e^2\cos^2(\Theta) - n_o\sin^2(\Theta))\right\}, \tag{4}$$

where c is the speed of light, $s=\sin(i)$, $E_{33}=n_e^2\cos^2(\Theta) + n_o^2\sin^2(\Theta)$, and n_o and n_e are the ordinary and extraordinary refractive indeces of the liquid crystal, respectively.

From Eq.s (3) and (4) we see that the two beams exert opposite torques on the liquid crystal molecules if i_1 and i_2 have opposite sign.

An analysis of the solutions of Eq.(2) as a function of I_1 and I_2 shows that, in the particular case when the equilibrium condition (1) is fulfilled, $\Theta=0$ is always a solution; however, when one of the two intensities (e.g. I_1) reaches a threshold value I_{th}, Eq.(2) has a branching point; the solution $\Theta=0$ becomes unstable and two additional

opposite solutions appear, giving rise to a second-order Freedericksz transition. As in usual Freedericksz transition, the threshold I_{th} scales as $1/L^2$, L being the sample thickness. Typical I_{th} values are in the range of 100-1000 W/cm^2 for a film thickness of about 100 μm. Such intensities can be readly achieved with cw laser beams. In the general case, when Eq.(1) does not hold, the first-order transition, described in the introduction, occurs, provided I_1 or I_2 is greater than I_{th}.

RESULTS

The experimental set-up is shown in Fig.1. Two p-polarized light beams, coming from the argon laser, were sent to a 90 μm thick film of MBBA nematic LC at oblique incidence (i_1=-10°, i_2=20°). Both beams were focused by means of equally convergent lenses (25 cm focal length); spot size was

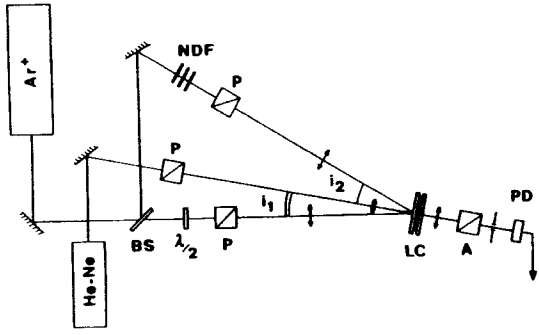

Figure 1. Experimental layout. P's, polarizers. BS, beam splitter; NDF, neutral density filters; LC, liquid crystal cell; PD, photodiode.

of the order of $5 \cdot 10^{-5}$ cm^2. The optical path length of the two beams was made different by an amount greater than the coherence length of the laser so that the interaction in the sample was completely incoherent and no fringes were formed therein. The power P_1 of the first beam could be varied continuosly by a variable attenuator while the power P_2 was changed step-wise by means of appropriate neutral density filters. The sample birefringence, (related to the tilt angle distribution along the sample) was monitored by a probe He-Ne laser beam tightly focused at normal incidence onto the sample.

The experimental results are shown in Fig.2, where the measured sample birefringence is plotted versus the power P_1 for two different values of P_2.

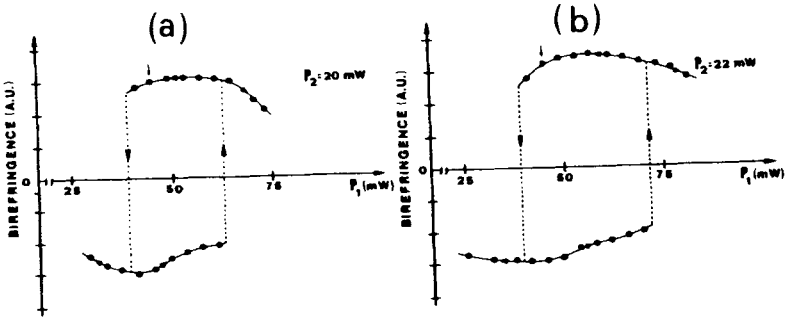

Figure 2. Measured sample birefringence as a function of P_1 for P_2=20mW (a) and P_2=22mW (b).

The discontinuous change of sign of the birefringence and the hysteresis loops are clearly seen in both cases. We can remark from the two drawings: a) in the points marked by the arrows, where Eq. (1) holds (exactly compensating beams), there are two symmetric values of the measured birefringence as we expect from Eq. (2); b) the transitions at lower power (shown as down-transitions in Fig. 2) occur at points for which the normalized power, i. e. the ratio P_1/P_{th}, is independent on P_2; c) the normalized power of the up-transition points grows with increasing P_2.

A comparison between the experimental data and the theory given above was made solving numerically Eq. (2).

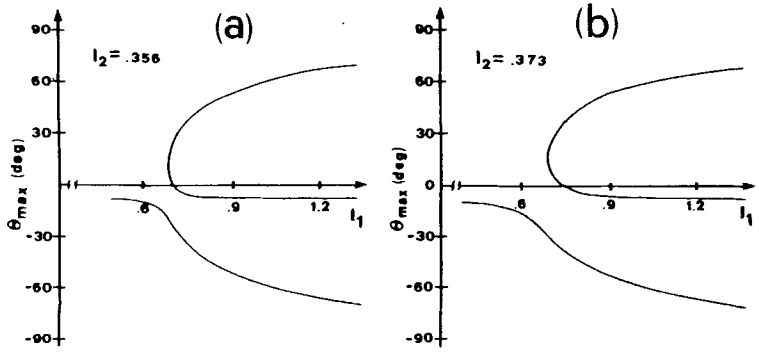

Figure 3. Theretical maximum tilt angle θ_{max} vs. the normalized intensity I_1/I_{th} for I_2= 20 mW/S_0 (a) and I_2= 22 mW/S_0 (b); I_{th}- 1.2 kW/cm^2 and S_0- .5·10^{-5}cm^2 are the threshold intensity and the spot size in the sample, respectively.

The computed solutions, for the physical situations corresponding to our measurements, are plotted in Fig. 3. We immediately note from these curves that no hysteresis cycle can be envisaged, since only one discontinuous jump, leading to the down-transition, is possible.For this transition point the normalized intensity is confirmed to be constant, i.e. independent on the value of I_2. This is a good test for our model since it is independent on the laser spot size measurement on the sample. On the other hand, no up-transition point comes up from the theory (except for some particular values of the parameters, not corresponding to the experimental conditions and hence not reported here), in contrast with the experimental evidence. This monostable rather than bistable behaviour is not so surprising, however, from the point of view of the theory given above. In fact, we must consider that the beam I_2 acts on the sample predistorted by the beam I_1 . Then, in the regions of the LC, where the angle between the director and the beam I_2 is less than 90°, the torques exerted by the two beams on the LC molecules are competitive. On the contrary, in the regions where this angle is greater than 90° (that is never the case for the undistorted sample) the effect of the two torques is cohoperative and no more change in the sign of the tilt angle θ occurs.

We are still attempting to find a quantitative explanation to such a discrepancy between theory and experiment. It is wothnoting, however, some qualitative features of the

observed transitions. Firstly we noted that the down-transition occurred in a time of a few seconds (<5 sec) and this is in agreement, at least as order of magnitude, with the estimate given by a theory that extends Eq.(2) to the time-dependent case[4]. During the switching time in the far-field pattern the number of diffraction rings, due to the sample birefringence[6,7], regularly decreased to zero and increased again to the new stable value. A quite different situation was ever encountered in the up-transition. The switching time was of the order of a minute and in the far-field pattern the rings became more and more trembling and confuse until only diffuse light could be seen onto the screen; hence, once the birefringence, monitored by the He-Ne probe, had reversed its sign, the pattern slowly came back to a neat picture with the appropriate number of rings.

We can conclude that the down-transition is surely due to the disappearing of the stationary solution of the upper branch, so that the system is forced to evolve towards to lower branch solution. Instead, this is certainly not the case for the up-transition.

Furthermore, since the incident power was kept constant during the transition, we can exclude that sample heating can play any role in this effect. On the basis of all these consideration, we have the feeling that thermal fluctuations could have a decisive role in the up-transition.

Further theoretical investigations, as rigorous stability analysis of the solutions, and experimental measure-

ments are in progress to better understand this point.

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