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# FIRST-ORDER OPTICAL FRÉEDEERICKSZ TRANSITION IN NEMATICS DOPED WITH CHIRAL AGENTS

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Abstract The Optical Fréedericksz Transition is studied in homeotropically aligned nematic E7 doped with cholesteric C15. The transition is found to be first-order and with large hysterisis. A simple model is also proposed.

## INTRODUCTION

Mixtures of nematic liquid crystals (NLC) and cholesteric liquid crystals (CLC) have been widely studied in recent years mainly because of their practical interest in the realization of liquid crystal displays, e.g. twisted or supertwisted cells. Since the 70th' it was well established that very small amount of chiral agents, like CLC, can induce an helical structure on the equilibrium configuration of an NLC cell, modifying its response to external fields[1].

Twisted nematic cells are usually realized in a planar geometry, thus the great majority of the studies on these mixtures has been performed in this configuration. Far fewer papers appeared on chiral-nematic mixtures with 'homeotropic' boundary conditions. For example, it was shown that, above some critical concentration of chiral dopant, bubbles of cholesteric phase appear in the nematic matrix, that can be quenched by suitable electric field[2].

On the other hand, in recent years, it was clearly demonstrated that also optical fields can reorient pure nematics[3]. To our knowledge, however,no studies have been done on the optical reorientation of chirally doped NLC. In this paper, we show, both theoretically and from experiment, that not only chiral-nematic mixtures can be reoriented by light, but also that the reorientation process take place in a quite different way. Unlike in pure nematics, the optical Fréedericksz transition is generally first-order and

usually is characterized by large hysteresis. Moreover, since the molecular director tends to achieve an helix structure, the angular momentum carried by the light beam plays an important role in the reorientation, and may lead e.g. to the phenomenon of optical phase locking[4].

## THE CHIRAL TORQUE

Consider a nematic film of thickness L enclosed between two glass walls coated for homeotropic alignment. When the nematic is doped with small amounts of suitable chiral agent, it tends to assume a cholesteric helix structure, having a pitch $p_0$ depending on the dopant concentration. This produces a new non-centrosymmetric term in the free-energy density having the general form $k_{22}q_0\mathbf{n}\cdot\mathrm{rot}\mathbf{n}$[5], where $\mathbf{n}$ is the molecular director, $k_{22}$ is Frank's elastic constant for twist and $q_0$ is a material constant related to the induced pitch by $p_0 = \frac{2\pi}{q_0}$. Depending on the dopant material, $q_0$ may be positive as well as negative, corresponding to right- or left-handed induced helix. In the absence of external fields, the chiral term in the free-energy leads to the following Euler-Lagrange equations for steady-state director distribution

$$\frac{d}{dz}\left[(k_{22}\sin^2\vartheta+k_{33}\cos^2\vartheta)\sin^2\vartheta\,\frac{d\phi}{dz}\right] - 2q_0k_{22}\sin\vartheta\cos\vartheta\,\frac{d\vartheta}{dz} = 0$$

$$(k_{11}\sin^2\vartheta + k_{33}\cos^2\vartheta)\,\frac{d^2\vartheta}{dz^2} - (k_{33}-k_{11})\sin\vartheta\cos\vartheta\left(\frac{d\vartheta}{dz}\right)^2 -$$

$$- [k_{33}-2(k_{22}-k_{11})\sin^2\vartheta]\sin\vartheta\cos\vartheta\left(\frac{d\phi}{dz}\right)^2 + 2q_0k_{22}\sin\vartheta\cos\vartheta\,\frac{d\phi}{dz} = 0, \qquad (1)$$

where the k's denote Frank's elastic constants of the nematic and $(\vartheta,\phi)$ are the polar and azimuthal angles of the molecular director $\mathbf{n} = (\sin\vartheta\cos\phi,\ \sin\vartheta\sin\phi,\ \cos\vartheta)$. The polar z-axis is normal to the sample walls and variations of $\mathbf{n}$ in the (x,y)-plane have been neglected. The proper boundary conditions at planes $z = 0$ and $z = L$ for homeotropic strong anchoring are

$$\frac{d\phi}{dz} = q_0 \frac{k_{22}}{k_{33}} \qquad (z = 0;\ z = L) \qquad (2)$$

$$\vartheta = 0.$$

The first of Eqs.(1) can be integrated to

$$(k_{22}\sin^2\vartheta + k_{33}\cos^2\vartheta)\sin^2\vartheta \frac{d\phi}{dz} - q_0 k_{22}\sin^2\vartheta = M_z = \text{const.} \qquad (3)$$

Boundary conditions (2) yield $M_z = 0$ for homeotropic anchoring. This is due to the fact that no torque along z can be exerted on the wall so that the flux of elastic angular momentum along z is zero. Using the first integral $M_z = 0$, Eqs.(1) can be separated yielding

$$\phi(z) = \int_0^L \frac{q_0 k_{22}}{k_{33} + (k_{22} - k_{33})\sin^2\vartheta}\, dz$$

$$(k_{11}\sin^2\vartheta + k_{33}\cos^2\vartheta)\frac{d^2\vartheta}{dz^2} - (k_{33} - k_{11})\sin\vartheta\cos\vartheta\left(\frac{d\vartheta}{dz}\right)^2 + \qquad (4)$$

$$+ \frac{(q_0 k_{22})^2 k_{33}\sin\vartheta\cos\vartheta}{(k_{22}\sin^2\vartheta + k_{33}\cos^2\vartheta)^2} = 0$$

From the second of Eqs.(4) we see that the overall effect of the impurity induced chirality is of superimposing a "chiral" torque on the nematic. This torque is proportional to $q_0^2$, and hence it does not depend on the sign of the induced chirality. Moreover, a simple stability analysis shows that the chiral torque tends to destabilize the initial homeotropic state $\vartheta \equiv 0$ of the nematic. This is due of course to the fact that the anchoring forces at the walls are incompatible with the helix structure induced by the chiral dopant. The homeotropic alignment, indeed, becomes unstable if the induced value of $q_0$ exceeds the characteristic threshold value $q_{th}$, given by

$$q_{th} = \frac{\pi k_{33}}{k_{22} L}. \qquad (5)$$

As expected, the threshold value decreases with increasing thickness L because the anchoring forces become less effective in maintain the homeotropic alignment. It can also been shown that the transition from homeotropic to chiral is first-order provided the elastic constants of the pure material obey the inequality

$$k_{11} + 3k_{22} < 3k_{33}. \tag{6}$$

In the one elastic constant approximation this inequality is not satisfied, so that the transition is second-order. But in most very common nematics (as 5CB and E7, for example) we have $k_{11} \cong k_{33} \cong 2k_{22}$, and inequality (6) is fulfilled, leading to a first-order transition.

In the presence of a linearly polarized laser beam, impinging at normal incidence onto the sample, an optical torque $\tau_0$ must be added on the right of the second of Eqs.(4), given by[6]

$$\tau_o = \frac{\pi^2 \tilde{I} n_e^3 \sin\vartheta\cos\vartheta}{\sqrt{(n_e^2 \sin^2\vartheta + n_o^2\cos^2\vartheta)^3}}, \tag{7}$$

where $\tilde{I} = \frac{I}{I_{th}}$ is the light intensity normalized to the threshold for the Optical Fréedericksz Transition (OFT) on the pure nematic, given by

$$I_{th} = \frac{\pi^2 c k_{33} n_e^2}{n_o(n_e^2 - n_o^2)L^2}, \tag{8}$$

where $n_o$ and $n_e$ are the ordinary and extraordinary refractive indices of the medium.

The equations (4) in the presence of the optical field have been integrated numerically for the nematic material E7 and the results are shown in Fig.1, where the distortion polar angle $\vartheta_{max}$ at the center of the film is reported as a function of the normalized light intensity $\tilde{I}$.

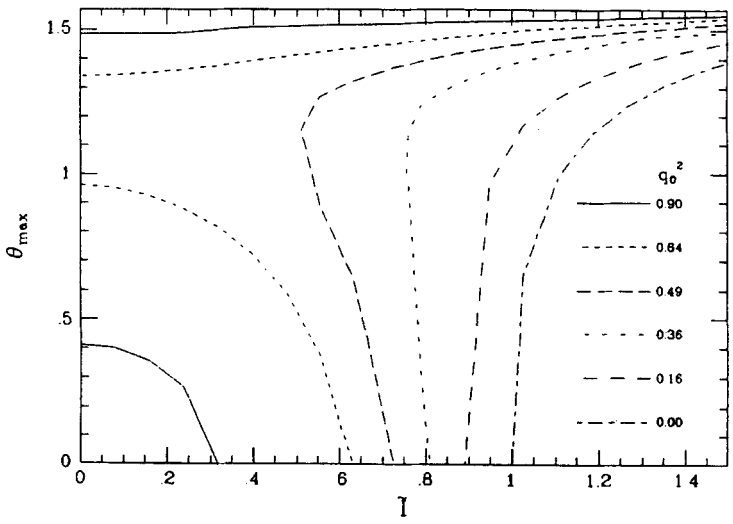



FIGURE 1 Computed maximum distortion angle $\vartheta_{max}$ vs. normalized intensity $\tilde{I}$. The parameter is $q_0^2$.

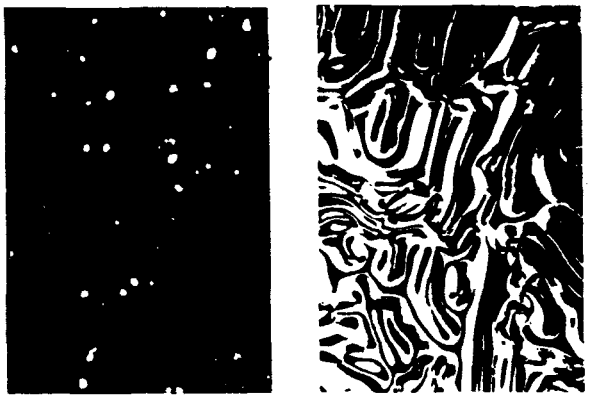
The parameter on the curves is $q_0^2$. The curve with $q_0^2 = 0$ refers to the pure nematic. The possibility of intrinsic first-order OFT with a large hysteresis is evident. For high enough $q_0^2$, we have a true optical bistable behaviour, in the sense that once the system is switched to the distorted state, this state remains permanent even when the light beam is removed.

In the simple model proposed here, the nonlinear feedback of the liquid crystal on the light polarization has been neglected. Nevertheless, we expect that, at least qualitatively, this model may be sufficient to understand the main features of the observed phenomena.

## THE EXPERIMENT

Our measurements were performed on thin films made by mixing small amounts of cholesteric C15 with nematic E7. Both liquid crystals were provided by BDH and were used without further purification. The samples we prepared had the same thickness (50μm) and different concentrations of C15 dopant [0.8%, 1%, 1.56%, 1.7%, 1.8%, 2%, in

w/w]. The sample walls were coated with HTAB for homeotropic alignment. The films with C15 concentration ≤1.7% showed a good homeotropic alignment when seen under a polarizing microscope. The samples with higher dopant concentration showed a characteristic distorted texture similar to the bubble domains observed in Ref.2 (see Fig.2). This confirms the existence of a critical impurity induced pitch $p_0$ at which the homeotropic alignment becomes unstable.



A) B)

FIGURE 2 Sample texture as seen by polarizing microscope. A) 0.8% w/w C15 in E7. B) 2% w/w C15 in E7. In sample B bubble domains are formed.

Measurements of OFT were performed on the samples having doping concentrations 0.8%, 1%, 1.56% and 1.7% by using a slightly focused linearly polarized argon laser beam at normal incidence. In this preliminary stage of the experiment, we simply look to the laser far-field pattern, counting the number of diffraction rings and roughly measuring the outer ring diameter. In the first two samples we found a behavior very similar to the one of pure nematic: the transition was second-order and also the threshold was almost the same. The samples at higher doping concentration, showed, instead, an evident first-order transition with large hysteresis, as shown in Figs.3 and 4.

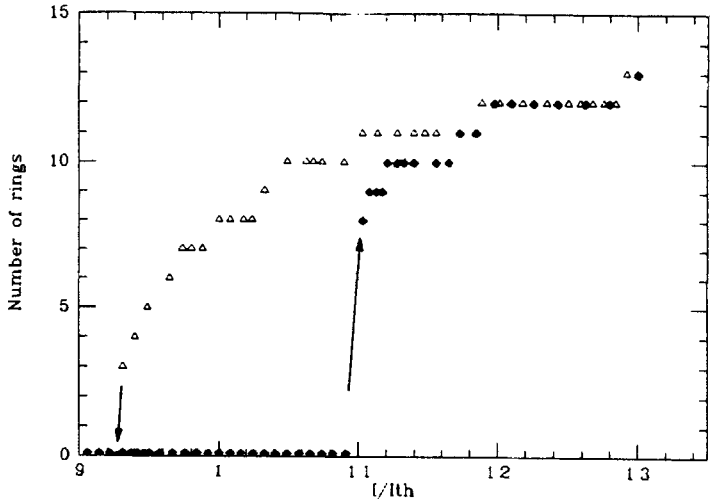



FIGURE 3 Number of rings in the laser far-field vs. power. Filled-in symbols refer to increasing power. 1.56% w/w of C15.

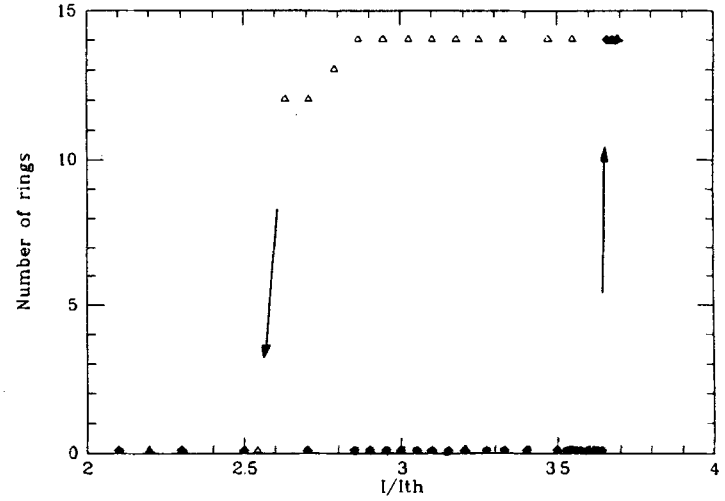



FIGURE 4 Number of rings in the laser far-field vs. power. Filled-in symbols refer to increasing power. 1.7% w/w of C15.

For the highest concentration, the far-field ring pattern remained visible for a time larger than 60 s (after which the rings quickly shrank off in about one second), even when the laser was switched off[7], confirming the presence of memory in that all-optical system.

Although these observations seem to confirm the main previsions of our model, some discrepancies also occurred. For example, the thre-

shold for the up-transition was seen to unexpectately increase with chiral concentration. We stress however, that these results are only based on preliminary observations and require further investigations.

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6. See, for example, H.L.Ong, Phys.Rev., A28, 2393 (1983).
7. To see the ring pattern, the power was actually held to a few milliwatts.