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Optical reorientation in cholesteric nematic mixtures

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The optical Fréedericksz transition for linearly polarized light at normal incidence is studied in mixtures of nematic E7 and cholesteric C15 in cells coated for homeotropic alignment. The reorientation process is found to be dramatically different from the case of pure nematic samples showing the phenomenon of optical phase locking and large hysteresis. These effects are ascribed to the occurrence of self-induced stimulated light scattering, which does not occur in pure nematics.

1. Introduction

It is well known that very small amounts of chiral agents, like cholesteric liquid crystals (CLC's) can induce a helical structure of the equilibrium configuration of a nematic liquid crystal (NLC) cell [1]. Such chiral nematic mixtures have been widely used in the realization of twisted or supertwisted cells for liquid crystal displays.

Twisted nematic cells are usually made in the planar geometry, so that the behaviour of chiral nematic mixtures with homeotropic boundary conditions has received much less attention. It is known, however, that, above a critical concentration of chiral dopant, bubbles of the cholesteric phase appear in the nematic matrix, that can be quenched by applying a suitable external electric field [2]. The existence of a critical concentration (for fixed cell thickness and temperature) is due to the incompatibility between the homeotropic alignment imposed by the anchoring at the walls and the helical structure induced by the chiral agent. Spontaneous formation of bubbles in the sample lowers its optical quality, thus preventing any practical use of homeotropically aligned chiral nematic mixtures in devices driven by static fields.

In a recent experiment, however, it was shown that the optical reorientation in a chiral nematic cell occurs without bubble formation and that, unlike in pure nematics, the reorientation is accompanied by very large hysteresis [3]. We stress here that the reorientation process in this case is quite different from the well-known optical Fréedericsz transition (OFT), in pure nematics, because it is essentially affected by a direct transfer of angular momentum from the light field to the medium, a process known as self-induced stimulated light scattering (SISLS) [4], even if the incident beam is linearly polarized.

In this paper we present a detailed experimental study of the optical reorientation in cholesteric nematic mixtures with linearly polarized light at normal incidence. The main result is that after a first threshold (which is lower than the threshold for the OFT in pure nematics) the sample birefringence [5] remains frozen to a value $\simeq \pi$, irrespective of the light beam intensity. This optical phase locking (OPL) phenomenon is indeed characteristic of many reorientational processes in which SISLS takes place [6]. We

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think that the present is the first experiment where this peculiar phenomenon was clearly observed. For higher light intensity, a second critical threshold was found where the sample birefringence jumps (via a forst order transition) to a very large value. This second transition is accompanied by large hysteresis as reported in [3].

In the next section we present a brief linearized theory that allows us to obtain a formula for the threshold of the OFT in chiral nematic mixtures. The calculated threshold values have been compared with the experimental observation, obtaining a good agreement as shown in figure 1.

2. Theory

When a pure nematic is doped with some amount of chiral agent (for example, a CLC), a new term is added to the elastic free energy density having the form $q_0 \mathbf{n} \operatorname{rot} \mathbf{n}$, where **n** is the molecular director and q_0 is related to the pitch $p_0 = 2\pi/q_0$ induced by the chiral dopant. For small dopant concentrations, q_0 is proportional to the concentration; moreover, q_0 depends strongly on temperature. In the presence of the optical field a further term must be added to the free energy density, having the form $-(\varepsilon_a/16\pi)|\mathbf{n}\cdot\mathbf{E}|^2$, where ε_a is the material dielectric anisotropy at the optical frequency and **E** is the complex electric field of the light beam. Unlike in the case of ordinary OFT in pure nematics, the molecular distortion in chiral nematic mixtures is conical, in general, so that the light in the interior of the sample is elliptically polarized. It is precisely the interaction between the polarization state of the light and the molecular distortion in the sample which is at the basis of SISLS. The whole problem consists in solving the coupled set of equations governing both the molecular distortion and the change of the beam polarization in traversing the sample [7]. In this paper, however, we consider only the linearized theory for small distortions about the initial homeotropic alignment. In this approximation, the light polarization remains unaffected by the sample reorientation and we are left with a simple equation for the director **n** alone. A

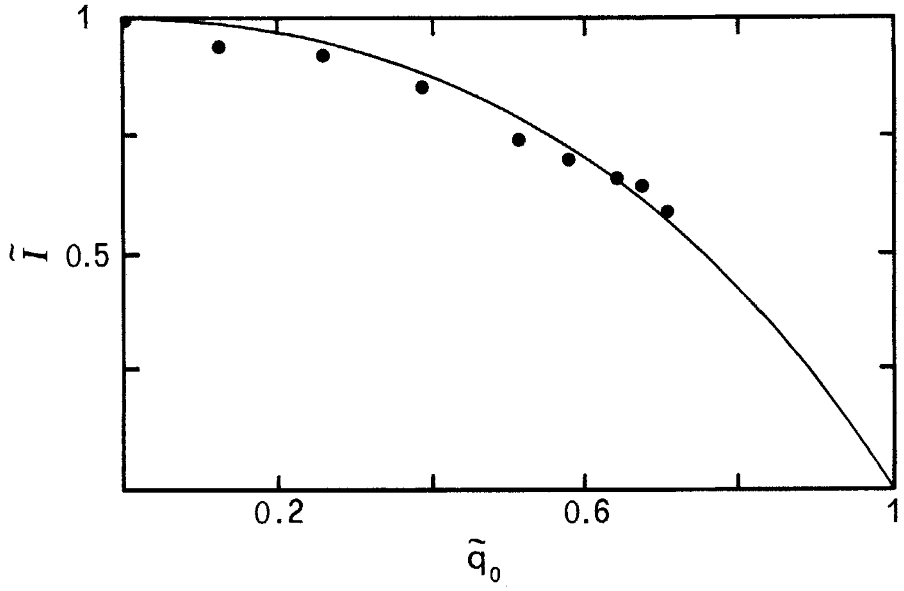


Figure 1. Reduced threshold intensity $\tilde{I}$ versus chiral parameter $\tilde{q}_0$. The dots are the experimental points and the solid line is from theory.

standard calculation, starting from the total free energy in the plane wave approximation and for normal incidence, shows that the steady state equation governing the small distortions about the homeotropic alignment can be written in the complex form

$$\tilde{n}'' - 2\pi \mathrm{i} \tilde{q}_0 + \tfrac{1}{2}\pi^2 \tilde{I}(\tilde{n} + \tilde{s}\tilde{n}^*) = 0, \tag{1}$$

where $\tilde{q}_0 = (k_{22}L/\pi k_{33})q_0$, L being the sample thickness and the k's the material elastic constants, $\tilde{I} = I/I_{\mathrm{th}}$, I_{th} being the threshold intensity for the OFT in the pure sample, $\tilde{n} = n_x + \mathrm{i}n_y$ is a complex variable built up from the components n_x and n_y of $\mathbf{n}$ parallel to the sample walls, and $\tilde{s} = s_1 + \mathrm{i}s_2$, s_1 and s_2 being the reduced Stokes parameters of the light polarization. In equation (1) the double prime denotes derivation with respect to the coordinate z normal to the boundaries. Boundary conditions for homeotropic alignment are $\tilde{n}(0) = \tilde{n}(L) = 0$.

For a linearly polarized light beam $\tilde{s} = 1$, and equation (1) can be solved yielding

$$\left.\begin{aligned}
n_x &= \frac{4\tilde{q}_0^2}{\Delta^2} A\left[\left(\cos\left(\frac{\pi\Delta z}{L}\right) - 1\right) + \frac{1-\cos\pi\Delta}{\sin\pi\Delta}\sin\left(\frac{\pi\Delta z}{L}\right)\right],\\
n_y &= 2\pi\tilde{q}_0 A\left\{\left(1 - \frac{4\tilde{q}_0^2}{\Delta^2}\right)\frac{z}{L}\right.\\
&\quad \left. + \left(\frac{4\tilde{q}_0^2}{\pi\Delta^3}\right)\left[\left(\frac{1-\cos\pi\Delta}{\sin\pi\Delta}\right)\left(1 - \cos\left(\frac{\pi\Delta z}{L}\right)\right) + \sin\left(\frac{\pi\Delta z}{L}\right)\right]\right\}
\end{aligned}\right\} \tag{2}$$

where A is an arbitrary real constant and $\Delta = \sqrt{(I + 4\tilde{q}_0^2)}$. The threshold intensity $\tilde{I}$ in equations (2) and (3) is given by the transcendent equation

$$\tan\left(\frac{\pi\Delta}{2}\right) = -\left(\frac{\pi\Delta}{2}\right)\left(\frac{\tilde{I}}{4\tilde{q}_0^2}\right). \tag{3}$$

We notice that $\tilde{I}$ depends on the square of q_0, so that it is unaffected by the sign of the cholesteric induced helix. A plot of the threshold reduced intensity $\tilde{I}$ as a function of $\tilde{q}_0$ is shown in figure 1. For no incident beam ($\tilde{I} = 0$) the homeotropic alignment becomes unstable when $\tilde{q}_0$ is larger than one, because of spontaneous domain formation. On the other hand, for a pure sample ($\tilde{q}_0 = 0$) the homeotropic alignment becomes unstable when $\tilde{I}$ is larger than one, because of the OFT. We notice, however, that for any dopant concentration so that $\tilde{q}_0 < 1$ we have $\tilde{I} < 1$, i.e. the light intensity needed to reorient the sample is always lower than the intensity for the OFT in a pure material.

3. Experiment

The main parameter affecting the qualitative behaviour of the OFT in chiral nematic mixtures is the helix parameter $\tilde{q}_0$. This depends essentially on the dopant concentration, temperature and sample thickness. We made several measurements of OFT in various samples by varying each of these parameters, and we found always consistent results. All data reported in this work, however, refer to a sample made of a mixture of nematic E7 with cholesteric C15 (both provided by BDH) having a thickness of 110 μm. The sample was made by two glass plates spaced by teflon strips. The walls were coated with HTAB (cetyltrimethyl-ammonium bromide) for homeotropic alignment.

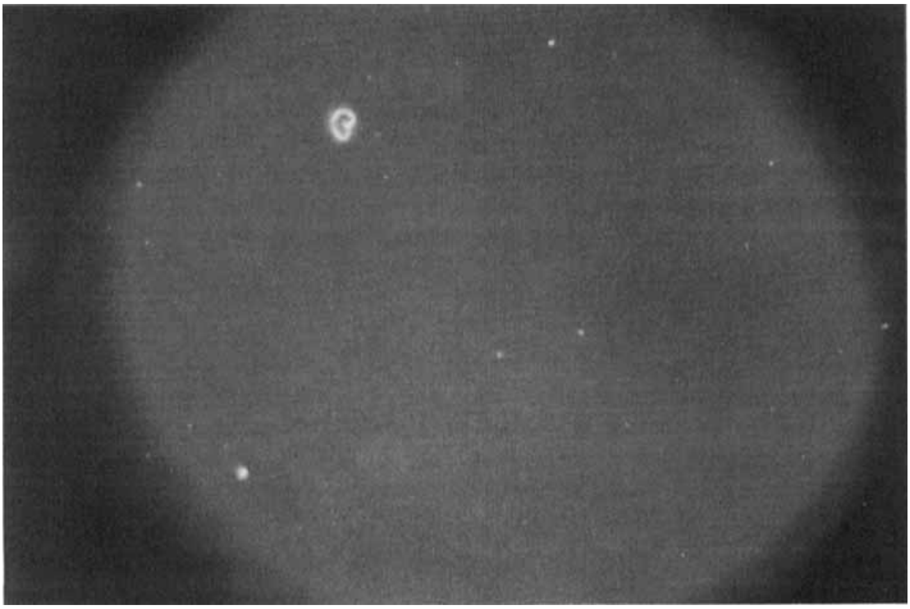
Before starting our study on the OFT, we made preliminary measurements to find the critical temperature T_c at which spontaneous domain formation occurred. The samples were placed into a temperature stabilized oven and observed by crossed polarizers under a microscope. The temperature was increased until domains appeared. Our results are summarized in the table. Since the chirality (inverse of pitch) usually increases with both temperature [1] and dopant concentration, the critical temperature T_c is expected to decrease as the concentration is increased as actually found.

We observed also that slightly above the critical temperature for the given concentration a characteristic bubble domain pattern appeared, the typical dimension of any bubble being about 1 μm. Further increasing the temperature produces the aggregation of many such bubbles until a typical filament pattern was stably formed. The bubble and filament patterns are shown in figure 2.

Our results regarding the OFT are summarized in figures 1, 3 and 4. The experiments were carried out with a commercial continuous wave argon laser Coherent INNOVA-90-6 (wavelength 514·5 nm) working in TEM_{00} mode. The beam profile was measured to be gaussian with $1/e^2$ waist of 81 ± 2 μm. All measurements were repeated

Critical temperature versus concentration.

T_c°C	Conc. per cent w/w
$26{\cdot}9 \pm 0{\cdot}1$	$0{\cdot}721 \pm 0{\cdot}004$
$27{\cdot}3 \pm 0{\cdot}1$	$0{\cdot}502 \pm 0{\cdot}004$
$27{\cdot}5 \pm 0{\cdot}1$	$0{\cdot}421 \pm 0{\cdot}004$
$28{\cdot}1 \pm 0{\cdot}1$	$0{\cdot}315 \pm 0{\cdot}004$
$28{\cdot}5 \pm 0{\cdot}1$	$0{\cdot}224 \pm 0{\cdot}006$
$29{\cdot}1 \pm 0{\cdot}1$	$0{\cdot}132 \pm 0{\cdot}006$



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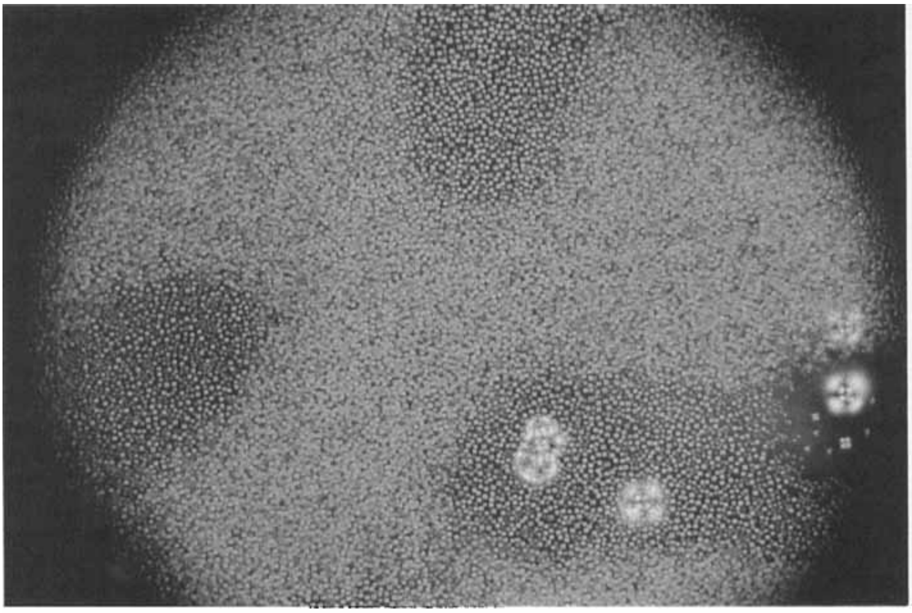

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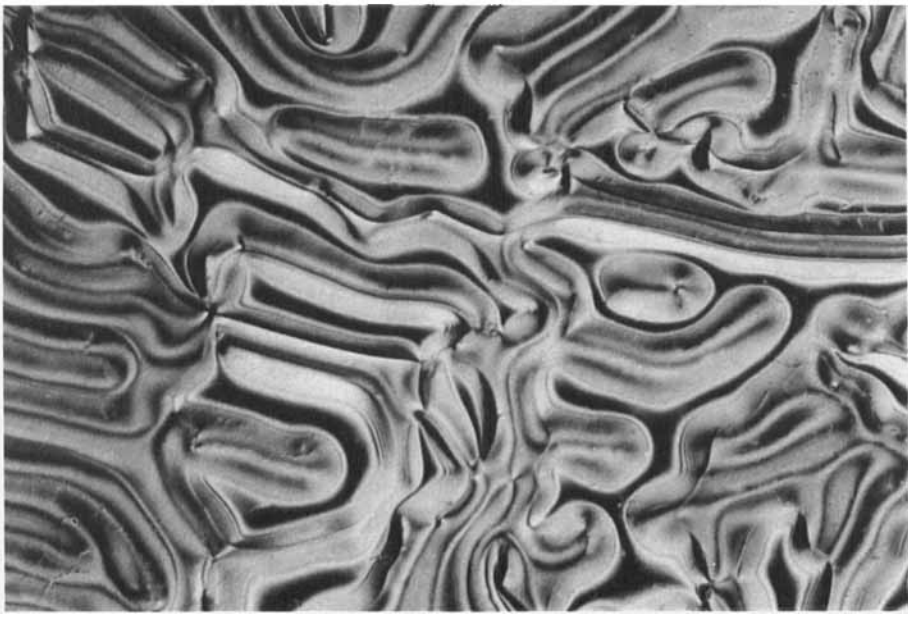

(*c*)

Figure 2. Microphotographs under a polarizing microscope. Sample thickness 110 μm; C15 0·242 per cent w/w. (*a*) Homeotropic alignment ($T < 28{\cdot}5°C$)); (*b*) bubble domains ($(28{\cdot}5°C) < T < 33{\cdot}5°C$); (*c*) filament domains ($T > 33{\cdot}5°C$).

both on the doped sample and on a pure nematic reference sample of equal thickness and temperature. The ratio $\tilde{I}$ between the measured OFT threshold in the doped and pure samples as a function of the chiral parameter $\tilde{q}_0$ is shown in figure 1. The two samples were placed in the same position along the beam, so that the threshold intensity ratio could be safely replaced by the threshold power ratio as measured by a suitable power meter. The data refer to samples 110 μm thick. The C15 concentration was 0·542 per cent in weight. The laser power at the threshold for the OFT in the pure sample was 280 mW at 22°C, corresponding to an intensity of 1·3 kW cm^{-2} at the sample. In plotting the data, we assumed the linear relation $\tilde{q}_0 - 1 = a(T - T_c)$. The parameter a was found by a best fit to the data. The agreement with theory (full line) is satisfactory. The actual chirality q_0 in figure 1 can be evaluated dividing $\tilde{q}_0$ by $(k_{22}L/\pi k_{33})$. The elastic constants of E7 in our range of temperatures (20–27°C) are $k_{22} = 5{\cdot}8 \times 10^{-7}$ dyne, $k_{33} = 16 \times 10^{-7}$ dyne [8].

We checked in a separate experiment, by detecting the laser-induced change in the liquid crystal ordinary refractive index, that the local temperature and increase due to laser absorption was always less than 1°C.

Figures 3 and 4 refer to the observed optical reorientation. The sample birefringence was estimated by counting the number N of diffraction rings in the laser far-field, and using the approximate relationship $\alpha \simeq 2N\pi$, where α is the phase difference accumulated by the ordinary and extraordinary waves in propagating through the medium. The far-field ring pattern was very neat and regular, confirming the fact that in the case of the optical reorientation we have no breaking into domains.

Figure 3 refers to a very poorly doped sample (0·242 per cent w/w). The threshold for the optical reorientation is very close to $I = 1$ and the transition is manifestly second order as happens in pure nematics [9]. Figure 4 refers to a greater chiral concentration (0·762 per cent w/w). In both cases the temperature was 22°C. The difference is

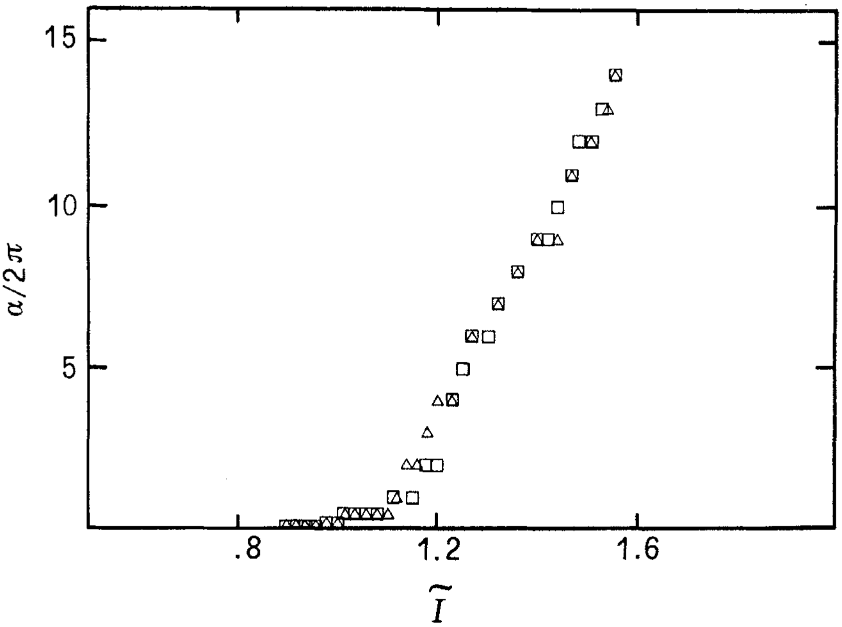

Figure 3. Sample birefringence versus reduced laser intensity. Sample thickness 110 μm; temperature 22°C; C15 0·242 per cent w/w.

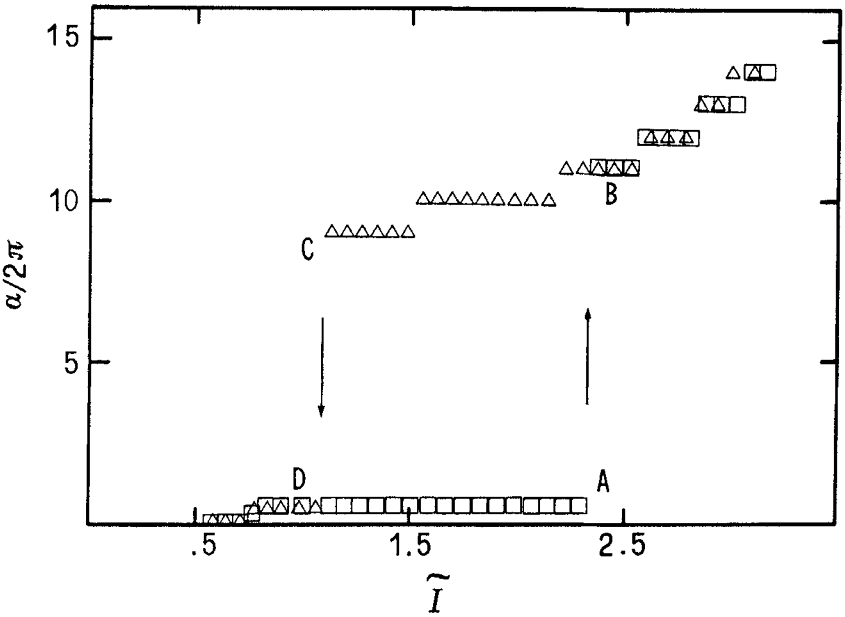

Figure 4. Sample birefringence versus reduced laser intensity. Sample thickness 110 μm; temperature 22°C; C15 0·762 per cent w/w.

surprising. A large hysteresis loop (ABCD) is evident, although a close inspection shows that the true threshold for the OFT is about one half of the threshold for the pure sample and that, after the threshold, there is a very large plateau where the optical phase α remains fixed at the value of π (this corresponds to about half a ring in the laser far-field). When the laser intensity reaches the value corresponding to the point A in the figure, a first order transition to point B is observed, where about 10 rings were observed. This state remained stable even if the intensity was decreased, until, at point C, another first order transition leads the system to the half-ring plateau (point D). Because the birefringence α depends only on the polar (tilt) angle θ formed by the director **n** and the z axis, we may conclude that after the true OFT threshold the angle θ remains almost fixed while the only effect of the light is producing a twist in the sample, varying the azimuthal angle ϕ alone. When the intensity reaches the point A, the free energy due to the twist degree of freedom becomes too high, so that the sample prefers to switch to the state B, characterized by low twist and higher tilt.

4. Conclusions

We showed that the OFT in chiral nematic mixtures is quite different from the case of pure nematics. The main difference are

(i) the threshold intensity for the OFT is lower;

(ii) above the threshold the optical birefrigence remains fixed at about π; the states where the optical phase locking (OPL) occurs are characterized by high twist and poor splay–bend distortion;

(iii) at very large intensity a first order transition drives the system to states characterized by high splay bend and poor twist distortion;

(iv) large hysterisis and intrinsic optical bistability (OB) between these states is observed.

It is evident that the occurrence of a first order transition and associated intrinsic OB is related to non-linear interaction between the molecular distortion and the polarization of the beam inside the sample. In other words, to SISLS. A full analysis of such phenomena cannot be made in the framework of the linearized theory presented here, where the light polarization plays no role. A non-linear model for the whole process and also the study of the dynamics involved in the OFT in chiral nematic mixtures is actually under study and will be presented elsewhere.

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