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DETAILED STUDY NEAR THE THRESHOLD OF THE OPTICAL FREEDERICKSZ TRANSITION: MULTISTABILITY AND OPTICAL PHASE-LOCKING

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Abstract Accurate measurements near the threshold for the Optical Fréedericksz Transition were made using a heterodyne interferometer-polarimeter. Evidence of multistability and Optical Phase Locking to odd multiple of π was obtained. A physical explanation of these phenomena based on total angular momentum conservation was also proposed.

INTRODUCTION

The Optical Fréedericksz Transition (OFT) was first observed in Nematic Liquid Crystals (NLC) by using a linearly polarized laser beam at normal incidence on a homeotropic film.[1] The effect consists in a strong optically induced reorientation of the mean direction **n** of the liquid crystal molecules, when the laser intensity exceeds a characteristic threshold.

It is commonly accepted that the OFT is very similar to the well known Fréedericksz transition induced in nematics by external static fields.[2] It is argued, in fact, that, in view of the linear polarization of the light inside the sample, the liquid crystal molecules feel an average optical field which is constant both in intensity and in direction. Consequently, all models proposed in the literature to explain the phenomenon retain this picture and lead to results that are very similar to the ones obtained in the static-field case.[3]

Nevertheless, recent experiments[4] have shown that the interaction between nematics and light is essentially different from the interaction between nematics and static fields because angular momentum can be transferred from the optical field to the medium. In simple geometries, the transfer of angular momentum from the radiation to the liquid crystal leads to Self-Induced Stimulated Light Scattering.[5]

This suggests that also in the simplest case, namely in the optical Fréedericksz transition with linearly polarized light at normal incidence, the coupling between the angular momentum of the light and the LC medium cannot be neglected so that features should appear having no analog in the static-field case. As shown in a recent theoretical work, these features should include Multistability and Optical Phase-Locking (OPL).[6]

In this paper we present a detailed study on the phenomena occurring near the threshold for the OFT in nematics made with the aid of a very sensitive heterodyne interferometer-polarimeter. Although our results confirm some of the effects foreseen in Ref.[6], we observed also some discrepancies that seem to require future investigations.

MULTISTABILITY AND OPTICAL PHASE-LOCKING

Before presenting the experimental observations, it is worth while to discuss briefly some of the results of the simple model of Ref.[6].

The main difference between the model proposed in Ref.[6] and all other reported previously in the literature is that the molecular director **n** is now allowed to precess around the direction of propagation of the laser beam, even if the laser beam is linearly polarized. This new degree of freedom leads in a natural way to the occurrence of peculiar phenomena, as OPL, intimately related to angular momentum transfer.

Consider, in fact, the typical configuration for OFT. A linearly polarized laser beam is impinging at normal incidence onto a homeotropic nematic film of thickness L. Let ϑ and ϕ denote the polar and azimuthal angles of the director **n** with respect to the z-axis directed along the laser beam. The laser intensity is slightly above the threshold for the OFT, so that a small reorientation is induced in the film. Initially, the reorientation takes place with **n** in the polarization plane of the beam.

Assume now small rigid rotation $\delta\phi(t)$ around the z-axis of the plane containing the director, due to fluctuations. Since the birefringence axis of the medium is now out of the polarization plane, the polarization of the light emerging from the sample will be elliptically polarized, eventually with very small ellipticity. Angular momentum is then transferred from the beam to the medium, producing a small nonzero torque $\delta\tau_z$ along the z-axis. Due to the linear polarization of the input light, the sign of $\delta\tau_z$ may be positive as well as negative, depending on the sign of the "elicity" of the light after having traversed the sample. Accordingly, the torque $\delta\tau_z$ may either damp out or enhance the initial fluctuation $\delta\phi$, leading to instability of the initial state.

The sign of the output elicity depends crucially on the phase difference $\Delta\psi$ accumulated between the ordinary and extraordinary waves in traversing the film. For film thicknesses $L \gg \lambda$, the optical wavelength, the phase $\Delta\psi$ is a very sensitive function of the laser-induced birefringence and, hence, of the laser intensity. We expect, therefore, that, as the input power is increased, the output elicity should change in sign many times, driving the system through a sequence of several stable and unstable states.

Moreover, total angular momentum conservation requires that, at steady-states, no net angular momentum is lost by the radiation field, so that the output polarization should eventually rotate, but still remain linear. For $\phi \neq 0$, this implies that the overall birefringence of the sample must adjust so that the phase difference $\Delta\psi$ is $n\pi$, with integer n. The model presented in Ref.[6] actually shows that two sets of steady states exist: one corresponding to even n (λ-retardation) which are unstable, and one to odd n ($\frac{1}{2}\lambda$ retardation), which are stable. In consequence, the laser-induced sample birefringence $\Delta\psi$ cannot change in a continuous way, when the incident power is increased, but it remains locked to some integer multiple of $\frac{1}{2}\lambda$ until it jumps to a different value. Correspondingly, also the ϑ-distributions along the sample are "quantized". We will refer to this phenomenon as to Optical Phase Locking (OPL).

For film thicknesses $L \gg \lambda$ the number n changes rapidly with the light intensity, so that very small steps in intensity are required to evidentiate the OPL. The intensity change ΔI corresponding to a change $\Delta n = 1$ is approximately given by[6]

$$\Delta I = I_{th} \frac{4\pi\delta^2}{\tilde{L}(\delta + 1)} (2A - k), \tag{1}$$

where $\delta=n_e/n_o$, $\tilde{L}=2\pi n_o(\delta-1)(L/\lambda)$, $A=(9-5\delta^2)/(8\delta^2)$, $k=1-k_{11}/k_{33}$, and

$$I_{th}=\frac{c\pi^2\delta^2k_{33}}{n_oL^2(\delta^2-1)} \tag{2}$$

is the threshold intensity for the OFT. In Eqs.(1) and (2) n_o and n_e denote the ordinary and extraordinary refractive indices of the liquid crystal and k_{11} and k_{33} the elastic constants for bend and splay, respectively. Typically, ΔI results of the order of 1% of the threshold intensity I_{th}.

Also the dynamics is affected by angular momentum transfer. In general, an oscillating approach to equilibrium is found from our model, when steady states with **n** out of the polarization plane are reached. Spiraling paths ending into multiple steady states are shown in Fig.1.

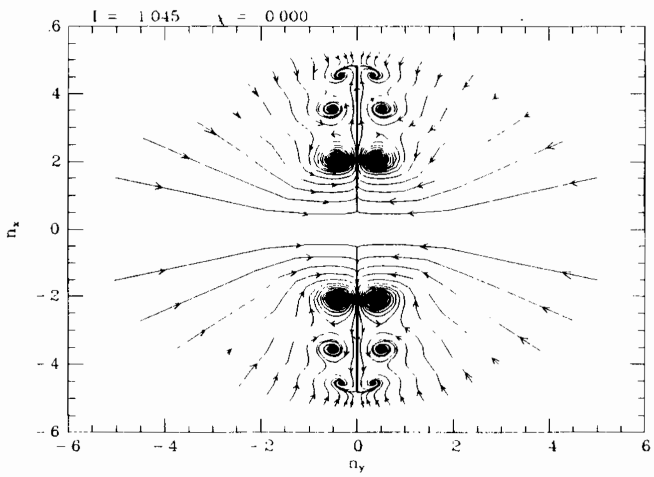

FIGURE 1 Trajectories of the director **n** in the (x,y)-plane for linear polarization and normal incidence on a homeotropic film. The intensity normalized to the threshold is $\tilde{I} = I/I_{th} = 1.045$.

EXPERIMENTAL RESULTS

As said before, different steady states are very close in the intensity scale, so that they can be evidentiated only by exploiting a very accurate control on the power of the laser source. Due to the very low relaxation time of liquid crystals near the OFT, a good long term power stability is also required. In the experiment, we used an INNOVA-90 Argon Ion laser in "light" mode. The long term power stability was found to be ≃ 0.1% in the absence of the sample. With the sample present, the stability lowered to 0.2%, due to the unavoidable backward light reflection into the laser itself. The power was changed in small steps (1 mW each) using a $\frac{1}{2}\lambda$-plate and polarizer variable attenuator driven by a stepping motor. After each step, we waited for 15' to reach equilibrium, but data were taken also during the transient.

On the optical phase scale, different steady states are spaced by π, so a much better phase detection sensitivity is needed. To detect the optical phase, we used a He-Ne counter-propagating probe beam injected into a Mac Zehnder heterodyne interferometer-polarimeter, working at 40 Mhz.[7] The data were collected by a HP-8508A vector voltmeter and sent to a PC for storage and processing. The overall acquisition rate for data was 10Hz. With this apparatus we obtained a sensitivity of about ±1° on the optical phase and we were able to follow the optical phase over many π.

All measurements were made on a E63 nematic liquid crystal film by BDH, 90μm thick. The sample walls were coated with HTAB for homeotropic alignment. The laser spot radius was 81μm (e^{-1} intensity) as measured by moving a small pinhole across the beam.

The threshold for the OFT was measured by monitoring the time constant τ of the initial growing up of the reorientation and by fitting the theoretical Landau slowing down relationship $\tau \propto (I-I_{th})^{-1}$. Due to our high sensitivity in detecting the optical phase, this method is very accurate. We found $I_{th}= 1.44\pm0.08$ kW/cm^2.
From E63 data at room temperature ($k_{11}=1.8\cdot10^{-6}$ dyne; $k_{33}=0.9\cdot10^{-6}$, $\varepsilon_o=2.32$; $\varepsilon_e=3.02$[8]) we obtain $I_{th}=1.86$ kW/cm^2. The agreement with the experimental value is good, having regard on the uncertainties on the material constants. The observed decreasing of the threshold could also be ascribed to finite anchoring forces at the walls.

Although a quantitative comparison with the model of Ref.[6] cannot be done at present, some of the expected dynamic features were indeed observed. For example, the existence of oscillating approach to equilibrium is shown by the large overshoot in the optical phase visible in Fig.2.

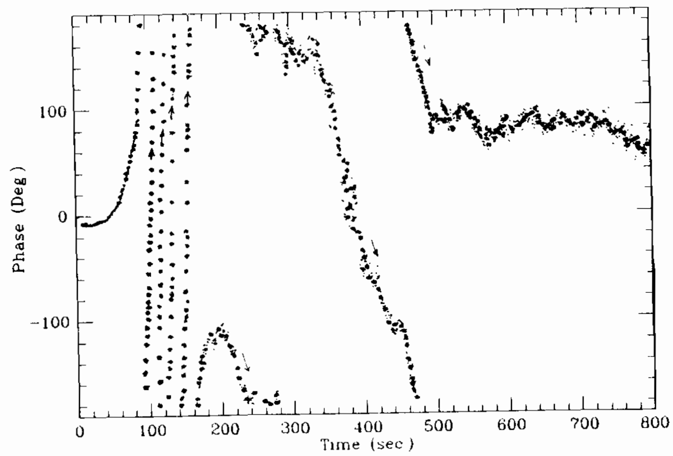

FIGURE 2 Optical phase overshoot in approaching a steady-state.

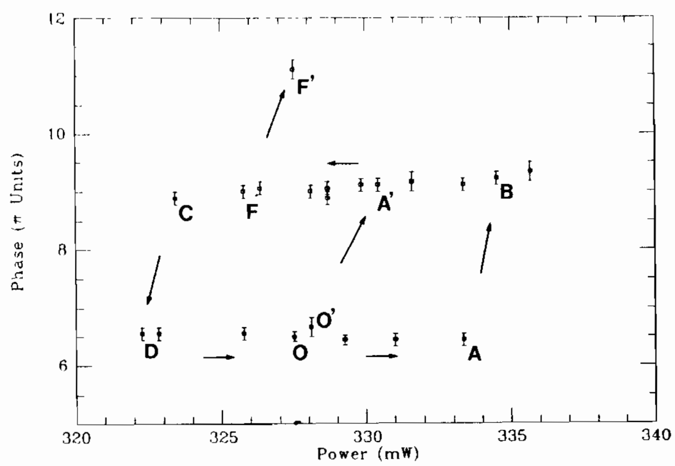

FIGURE 3 Probe phase change $\Delta\alpha$ as a function of pump power.

Multistability was clearly observed too, as shown in Fig.3, where the phase difference $\Delta\alpha$ between the ordinary and extraordinary components of the He-Ne probe beam is plotted as a function of the argon laser power.

The data were taken as follows. Starting from the power P_0 corresponding to $\Delta\alpha \cong 7\pi$ (point O)[9], the power was slowly increased until the point A is reached. A small further increase in power drew the system to point B, having $\Delta\alpha \cong 9\pi$. The power was first increased to point B' and then decreased to C No appreciable change in phase was observed. Decreasing the power below D, made the phase jump back to about 7π (point D). The power was then increased to reach O', where a new jump to 9π was observed (point A'). At this point, the power was lowered to point F, where a new phase jump occurred to point F', corresponding to $\Delta\alpha \cong 11\pi$.

All measurements reported here have been made with a dielectric polarizing cube in front of the argon laser, leading to an intensity extinction ratio of about 1/100. We repeated the measurements using a high quality Glan-Thompson polarizer with a measured intensity extinction ratio 1/5000. Neither multistability nor OPL were observed in this case. The state with **n** in the argon polarization plane was found to be always stable in the range of available light intensities. We have no explanation for this phenomenon. It is surprising, however, that using linear polarized light having an extinction ratio of 1/100 yields so qualitatively different results. A detailed theoretical study of the stability of the states having **n** in the polarization plane for linear and "almost linear" polarization is in progress.

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8. Data and samples were kindly provided by dr. I.Janossy, Central Research Institute of Physics, Budapest, Hungary.

9. We also checked that the actual number of diffraction rings in the argon far-field was of about three.