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MULTISTABILITY HYSTERESIS AND OPTICAL PHASE LOCKING IN THE OPTICAL FRÉEDERICKSZ TRANSITION WITH LINEARLY POLARIZED LIGHT AT NORMAL INCIDENCE

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Abstract We report the first experimental observation of hysteresis, multistability and locking of the optical phase in the Optical Fréedericksz Transition induced by a linearly polarized laser beam at normal incidence into a homeotropically aligned nematic film. The occurrence of these unexpected effects can be understood in terms of angular momentum transfer between the optical field and the medium.

INTRODUCTION

The Optical Fréedericksz Transition (OFT) was first observed in nematics by using a linearly polarized laser beam at normal incidence[1]. The effect consists in a strong optically induced reorientation of the molecular director in the sample when the laser intensity exceeds a characteristic threshold.

It is commonly accepted that this effect 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 field which is constant both in intensity and in direction. Accordingly, 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.

Nevertheless, some recent experiments[3] have shown that the interaction between nematics and light is essentially different from the interaction between nematics and static fields because liquid crystal can directly couple with the angular momentum carried by the optical field. In simple geometries, the transfer of angular momentum from the radiation to the liquid crystal may lead to Self-Induced Stimulated Light Scattering[4]. This suggests that also the simplest case, namely the optical Fréedericksz transition with linear light at normal incidence, could manifest completely new features having no analog in the static-field case.

In this work we present the experimental evidence of some of the unexpected effects that may occur when a linearly polarized laser beam is sent at normal incidence onto a homeotropically aligned nematic film. These effects include multistability, hysteresis and birefringence locking. We will present also a very simple model, based on angular momentum transfer, which is in qualitative agreement with the observations. This model starts from different first principles and it is in no way similar to all previous works appeared in the literature to model the Optical Fréedericksz Transition[5].

THE ROLE OF THE ANGULAR MOMENTUM OF THE LIGHT

The main difference between optical and d.c. fields is that the former can carry angular momentum due to the photon spin. Consider a c.w. laser beam of frequency $\omega = 2\pi/\lambda$ impinging at normal incidence onto a thin film of nematic liquid crystal enclosed between two glass walls coated for homeotropic alignment. If the beam is linearly polarized it carries no angular momentum, on the average, along its propagation direction (z-axis). As it is well known, if the beam intensity at the sample exceeds a critical threshold I_{th} for the OFT, a reorientation of the director $\hat{n}$ takes place in the sample. If the optical field remains linearly polarized everywhere in the medium, one expects that $\hat{n}$ will remain always in the plane of polarization of the beam. Denoting with ϑ and ϕ the polar and azimuthal angles of $\hat{n}$ with respect to the polar axis z [$\hat{n}$ = $(\sin\vartheta\cos\phi, \sin\vartheta\sin\phi, \cos\vartheta)$, this means that only ϑ is affected by reorientation, while ϕ remains zero. As shown e.g. by Ong exploiting the plane-wave Geometric Optics Approximation (GOA), above the threshold for the OFT, the distorted steady state [$\vartheta=\vartheta(z)$, $\phi=0$], is stable against fluctuations in the polar angle ϑ[6]. The stability against ϕ-fluctuations, instead, was simply assumed in Ong's and related papers.

Now, a simple argument, based on angular momentum conservation, shows that this, indeed, is not the case and that the system may well become unstable with respect to fluctuations of the azimuthal ϕ-angle.

Assume, in fact, a small fluctuation $\delta\phi(t)$ of the ϕ-angle. Due to the homeotropic anchoring at the walls, we may assume that this perturbation corresponds to a small rotation, without twist, of the plane containing the steady-state distorted director $\hat{n}(z)$. The absence of twist in the perturbation is expressed mathematically by the fact that $\delta\phi(t)$ does not depend on the z-coordinate[7]. Since the birefringence axis of the medium is now out of the polarization plane of the beam, the polarization of the light emerging from the sample will be elliptically polarized, even if with a very small ellipticity. Angular momentum is therefore transferred from the beam to the medium, producing a nonzero small torque $\delta\tau_z$ along the z-axis. The input light being linearly polarized, the sign of $\delta\tau_z$ may be positive as well as negative, depending on the sign of the "elicity" of the light beyond the sample. This means that the torque $\delta\tau_z$ may either damp out or enhance the initial fluctuation $\delta\phi$. We expect, therefore, that the steady state $\hat{n}(z)$ may be stable or unstable, depending on the elicity value of the light beyond the sample. Roughly speaking, when the light beam crosses the sample, its polarization suffers a change in the elicity sign for a change of π in the phase retardation α between the ordinary and extraordinary wave in the medium. Owing to the laser-induced optical reorientation, for nematic films having thicknesses $L \gg \lambda$, the phase difference α may vary over many π as the laser intensity I is increased, so that the stability of the state $\hat{n}(z)$ is reversed many times in a real experiment. Multistability and eventually hysteresis are therefore expected.

A SIMPLE MODEL

In this section an oversimplified model is presented describing the effects produced by the angular momentum transferred from the optical field to the medium near the threshold for the OFT, in the case of linearly polarized light and normal incidence. In this model we used the plane-wave approximation (all fields depend on one spatial coordinate) and the GOA. Moreover, we will consider steady states only.

Just above the threshold for the OFT, the polar angle ϑ of the molecular director is very small and , for homeotropic alignment at the sample wall (planes z=0 and z=L), its steady state value can be approximated by[8]

$$\vartheta(z) = \vartheta_m \sin(\pi z/L) + o(\vartheta_m^3), \tag{1}$$

with

$$\vartheta_m \cong \sqrt{[I(\phi)/I_{th} - 1]/2B} \tag{2}$$

and

$$B = \tfrac{1}{4}(1 - k - 9u/4)$$
$$k = 1 - k_{11}/k_{33} \tag{3}$$
$$u = 1 - (n_o/n_e),$$

k_{ii} being Franck's elastic constants and n_o, n_e the ordinary and extraordinary indices of the material (we assume $n_o<n_e$). Finally, the threshold I_{th} for the OFT is given by

$$I_{th} = [ck_{33}/(n_o u)](\pi/L)^2. \tag{4}$$

As mentioned above, in all previous works it was tacitly assumed that, during the laser-induced reorientation, the director $\hat{n}$ is bounded to lie in the plane $\phi = 0$. But we know that, due to angular momentum transfer from the optical field to the medium, this is not the case and we must account also for variations of ϕ.

A first consequence is that, since only the component of the optical field in the plane containing $\hat{n}$ is effective in changing the polar angle ϑ, the intensity I in Eq.(2) will depend in general on the azimuthal angle ϕ, as explicitly indicated in Eq.(2).

Denoting with I_0 the intensity of the incident beam at the sample, we have, in fact

$$I(\phi) = I_0 \cos^2\phi. \tag{5}$$

On the other hand, the total (radiation + medium) angular momentum balance, yields, for homeotropic alignment[9]

$$\gamma \int_0^L (\partial\phi/\partial t)\sin^2\vartheta \, dz = (I_0/\omega)\Delta e, \tag{6}$$

where γ is a viscosity coefficient and Δe is the ellipticity change suffered by the polarization of the light in traversing the sample. We notice also that $(I_0/\omega) = N\hbar$, N being the photon flux in the incident beam.

From this equations we see that at steady states, where $\partial\phi/\partial t = 0$, we must have $\Delta e = 0$ and the light emerging from the sample is still linearly polarized as the incident light, eventually in a different direction. We notice that this conclusion arises from angular momentum conservation only, and it should be true, at steady states, even in the presence of very large optically induced distortions in the sample.

Near the threshold for the OFT, however, the distortion is expected to be very small. In particular, we may assume that the twist induced by the optical field into the sample is negligible, at least in the first approximation[10].

Then, we assume $\phi = \phi(t)$, independent of the spatial coordinate z. (Notice that, in view of the homeotropic anchoring at the walls, a rigid rotation of $\hat{n}$ throughout the sample without twist does not requires any spent of elastic energy, so that, unlike ϑ, we cannot assume the ϕ-angle itself small.)

When twist is neglected, the distorted sample is optically equivalent to a retardation plate having its faster axis forming an angle $\phi(t)$ with the polarization plane of the incident beam, the instantaneous phase retardation of the plate being

$$\alpha(t) = (2\pi/\lambda)\int_0^L \left[\frac{n_o n_e}{\sqrt{n_e^2\cos^2\vartheta + n_o^2}} - n_o\right] dz. \tag{7}$$

Then, a very simple calculation shows that the instantaneous ellipticity of the light emerging from the sample is given by

$$e(t) = -\sin 2\phi(t)\sin\alpha(t). \tag{8}$$

For steady states we must have $\Delta e = e = 0$, because of angular momentum conservation ($e=0$ at the input plane), so we see that steady states are characterized by

$$\sin 2\phi \sin\alpha = 0. \tag{9}$$

Moreover, when ϑ is small, we may use Eq.(1) to get an approximate expression for the integral appearing in Eq.(7), obtaining

$$\alpha = \tfrac{1}{4}(\pi L/\lambda)n_o u\vartheta_m^2 + o(\vartheta_m^4). \tag{10}$$

so that, using Eqs.(5) and (2), we get

$$\alpha = \tfrac{1}{8}\pi n_o(u/B)(L/\lambda)[(I_0/I_{th})\cos^2\phi - 1]. \tag{11}$$

Equations (9) and (11) are a set of two transcendental equations which determine the steady state values of the azimuthal ϕ angle and the phase retardation α. Once α is known, ϑ_m is found from Eq.(10).

It is worth noting that in practical samples the ratio (L/λ) is very large, so that even if ϑ_m is small, the phase retardation may vary over many π[11]. For this reason, we cannot consider α small in Eq.(9).

From Eqs.(11) and (12) we see that the system may have many steady states. One steady state always exists where $\phi = 0$ and $\alpha = \alpha(I_0)$, as obtained from Eq.(12), with ϕ set to zero. This steady state was extensively studied in all previous works. But now we find out a number of new steady states corresponding to a phase retardation $\alpha = n\pi$, with integer n, and with azimuthal angles ϕ_n given by the real roots of

$$\cos^2\phi_n = (I_{th}/I_0)[1 + 8n(B/u)(\lambda/n_oL)] \quad (n = 1,2,\ldots). \qquad (12)$$

In these states the director $\hat{n}$ is out of the plane of polarization of the incident light and the phase retardation of the sample is locked at an integer number of π. In these states the sample behaves as a $\frac{1}{2}\lambda$-plate for odd n and as a λ-plate for even n. From this we see that the polarization of the light emerging from the sample, being always linearly polarized at steady states, may be either rotated by an angle $2\phi_n$ for the states with odd n or remains unchanged for the state $\phi = 0$ and the states $\phi = \phi_n$ with even n.

The steady state values ϕ_n of the azimuthal angle are plotted in fig.1 as function of the incident reduced intensity I_0/I_{th}. We see that the branches $\phi = \phi_n$ coalesce with the state $\phi = 0$ at characteristic threshold intensities I_n, obtained from Eq.(12) by setting $\phi_n = 0$:

$$I_n = I_{th}[1 + 8n(B/u)(\lambda/n_oL)] \qquad (n = 1,2,\ldots). \qquad (13)$$

The spacing $\Delta I = I_{n+1} - I_n$ between two successive thresholds is given by $\Delta I/I_{th} = 8n(B/u)(\lambda/n_oL)$. Due to the very small ratio λ/L, the quantity $\Delta I/I_{th}$ is typically of the order of 2÷3%.

As anticipated in Sec.2, the angular momentum balance Eq.(6) can be also used to investigate the stability of the solution $\phi = 0$, $\alpha=\alpha(I_0)$. Assuming a very small perturbation $\delta\phi(t)$ of this state, in fact, Eqs.(6) and (8) yield $\delta\dot{\phi} = k\delta e = -2k\delta\phi\sin[\alpha(I_o)]$, with k a positive proportionality constant. Then, we see that if the intensity I_0 is so that $\sin[\alpha(I_0)] < 0$, $\delta\dot{\phi}$ has the same sign of $\delta\phi$ and the ϕ-fluctuation is enhanced, yielding instability. A simple inspection shows that the state $\phi = 0$ changes its stability at each branching point $I_0 = I_n$. The unstable states are represented by dotted curves in Fig.1.

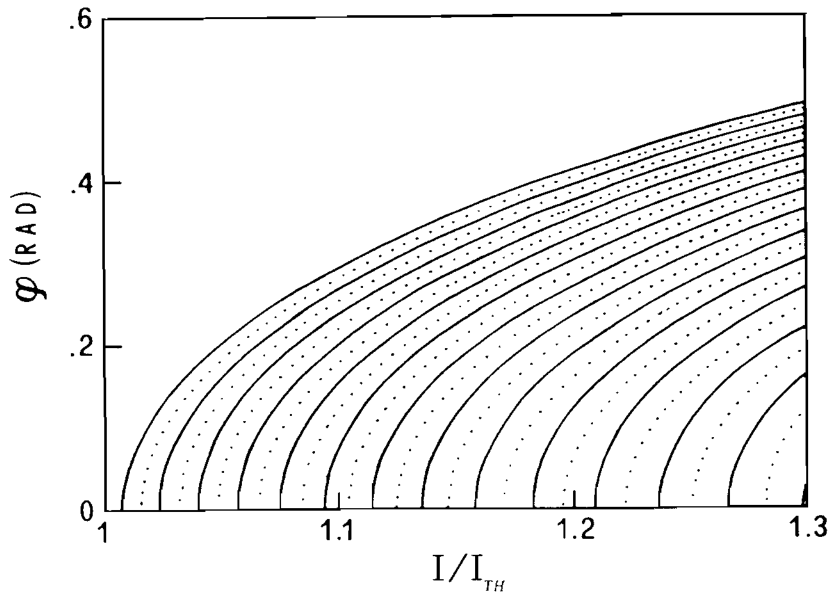

FIGURE 1. Azimuthal angle ϕ as a function of the reduced laser intensity I/I_{th}. The parameter on the curves is the optical phase retardation α. On each ϕ-curve α is locked to an integer number of π.

The simple model presented in this Section assumes a negligible twist in the sample. For this reason, the branches in Fig.1 corresponding to odd n appear to be stable for any value of the intensity I_0. We expect, instead, that when the intensity is increased and ϕ goes out of the plane ϕ=0 following one of the stable branches in Fig.1, the distribution of **n** in the sample becomes more and more twisted. This produces an accumulation of elastic energy that finally relaxes driving the system to a new equilibrium state having a very small angle ϕ. The stability of the branches $\phi = \phi_n$ (n odd) is therefore lost at high enough intensity I_0. This behavior was effectively observed in the experiment.

THE EXPERIMENT

A rough but very usual way of observing the laser-induced reorientation in nematics is to look at the diffraction ring pattern occurring in the far field of the incident beam beyond the sample. Each ring corresponds roughly to a change of 2π in the phase retardation α. Therefore, if one just counts the rings, the sensitivity in measuring α is of the order of π. A more accurate measure of α is obtained by sending a weak laser beam in the sample to probe its birefringence, e.g. by monitoring the light through crossed polarizers. With this method the accuracy on α may be improved, but it still does not allow independent measurements of the optical phase retardation angle α and the azimuthal angle ϕ. In order to verify the provisions of the model presented above, we need an experimental equipment able to give accurate and independent determinations of α and ϕ. We then used a probe He-Ne beam inserted into an heterodyne interferometer/polarimeter scheme, which gives a real-time complete information on the polarization of the probe beam after having traversed the sample. This apparatus was presented elsewhere[12]. Using this apparatus, we may reach an accuracy on the ϕ and α angles of the order of $\pm1°$. Moreover, in order to resolve the structure shown in Fig.1, we should control the intensity of the pump laser beam within an accuracy of a few percent. For the sample used in the experiment (nematic E7 film , 120μm thick) the threshold power needed to induce the OFT was found of about 180mW, so that, in the measurements, we varied the power in steps of 1mW each (the long term stability of our argon laser Coherent Innova 90/3 was of about 5‰).

In the experiment, we used two different polarizations of the probe beam: linear polarization at 90° with respect to the polarization of the Ar^+ laser and circular polarization. In the first case, we observed that the probe light emerged from the sample elliptically polarized. On the other hand, if ϕ remained zero, no variation of the polarization state of the He-Ne beam should be observed, only an ordinary wave being excited by the probe in the sample. The fact that the probe beam was found elliptically polarized, while not per-

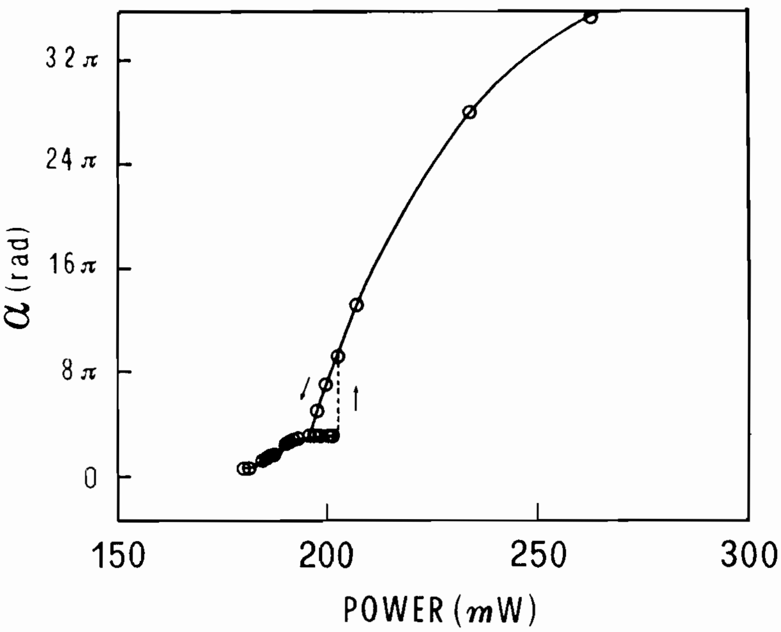

FIGURE 2. Birefringence angle α as a function of the power of the pumping laser beam.

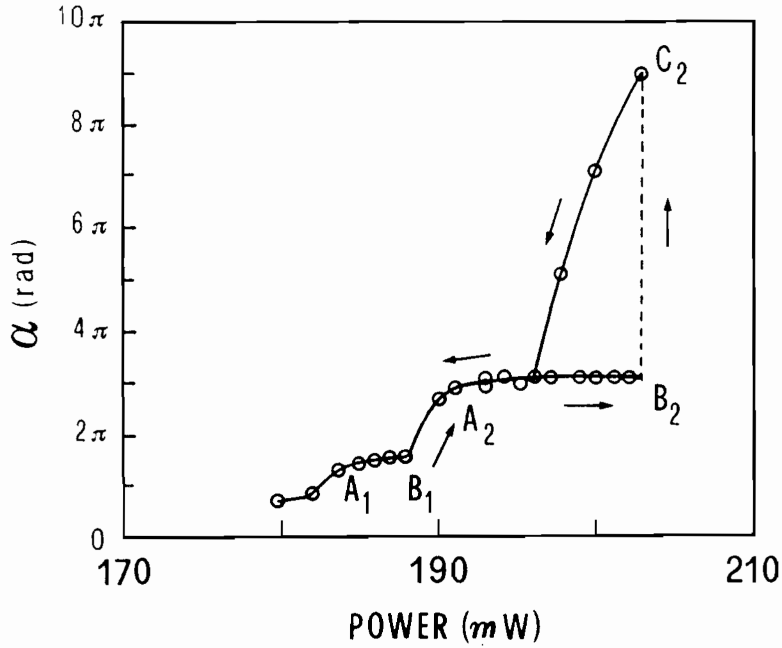

FIGURE 3. Birefringence angle α as a function of the power of the pumping laser beam.

mitting an accurate estimate of α, gives a qualitative evidence that the director $\hat{n}$ is moved out of the argon polarization plane.

In order to obtain a more accurate measure of the optical phase retardation α, we made the probe beam circularly polarized. The results are presented in Figs.2 and 3, where the measured birefringence angle α is reported as a function of the power of the argon beam.

Figure 2 shows the large-scale behavior of $\alpha(I_0)$, when the intensity is varied in large steps and the accuracy on the measure of α is not very high. We see that the overall behavior of $\alpha(I_0)$ follows closely the development of the rings in the far-field pattern, as reported by many other authors[1]. In Fig.3, instead, we expanded the near-threshold zone, on a smaller scale. The occurrence of bistability, hysteresis and optical phase-locking is evident. In particular we observed two plateaux corresponding to a locking of the birefringence angle α at roughly π and 3π (the discrepancy is due to the difference in wavelength between the pump and probe beams). The optical phase locking phenomenon was also supported by the fact that at all points on the plateaux A_1B_1 and A_2B_2 the ring pattern in the far-field of the pump beam was "frozen" to only an aloe and to one ring in the two cases, respectively. When the pump intensity was slightly increased above the value corresponding to the point B_2 in the Figure, the system switched to a state characterized by four rings in the Ar^+ laser far-field (point C_2). If the intensity was slowly decreased the rings in the far-field collapsed to the center of the pattern and the corresponding birefringence α lowers until point A_2 was reached again. Further decreasing the argon intensity, a new phase locking was observed, corresponding to an aloe in the Ar^+ laser far-field, after which the system decayed below the threshold to the undistorted state. The jump from point B_2 to point C_2 was not provided by the simple model outlined in Sec.3, but we believe that it is due to accumulation of twist elastic energy that sudden is delivered.

In conclusion, we think that the results reported in Figs.2 and 3 give the first experimental evidence that also in the simplest case, namely in the case of linear polarized light at normal incidence, the OFT in homeotropically aligned nematics may present peculiar aspects intimately connected to the angular momentum carried by photons.

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[10] A more accurate model accounting also for small nonzero twist distortions will be presented elsewhere.

[11] As it is well known, a rough measure of $\alpha/2\pi$ is the number of diffraction rings appearing in the far field of the incident beam beyond the sample. In usual experiments this number may be large (10 or more).