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EXPERIMENTAL PROBLEMS IN THE OBSERVATION OF MULTISTABILITY IN THE OPTICAL FRÉEDERICKSZ TRANSITION IN NEMATICS.

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Abstract We present an experimental apparatus which permits to have a complete and accurate real-time information on the birefringence of a nematic film during the Optical Fréedericksz Transition. A simple way to compensate for spurious effects due to diffraction and self-focusing is also outlined. An interesting physical problem (multistability in the OFT) that could be observed only by using this kind of apparatus is is also discussed.

Keywords: multistability, optical Freedericksz, transition, nematics

INTRODUCTION

When an intense laser beam propagates inside a liquid crystal a strong reorientation of the molecules of the medium may result. The molecular reorientation is usually monitored either by counting the number of diffraction rings produced in the laser far-field beyond the sample or by probing the sample birefringence with another laser beam.

In some cases, depending on the boundary conditions at the sample walls and on the incidence angle of the beam, the molecular reorientation is induced only when the laser intensity exceeds a characteristic threshold, called the Optical Fréedericksz Threshold (OFT). The OFT was first observed by using a linearly polarized laser beam at normal incidence onto a homeotropically aligned nematic film.[1].

In many respects, the phenomenon is similar to the well known Fréedericksz transition induced in nematics by external static fields[2]. Nevertheless, in a recent paper it was pointed out that also the simplest and most studied case, namely the OFT with linearly polarized light at normal incidence onto a homeotropic cell, could manifest novel and yet unobserved features as multistability and hysteresis that are absent in the static-field case[3]. It was also realized that these peculiar effects are ultimately related to the intrinsic angular momentum (photon spin) carried by the optical radiation[4].

As it will be shown below, the practical observation of these new effects requires an information about the laser-induced molecular distortion more complete and accurate than the one usually achieved in standard experiments on the OFT. The problems the experimenter is faced with in this kind of measurements are generally met whenever one wants to obtain a very accurate determination of the molecular distortion across a liquid crystal film by exploiting optical methods. We think that a discussion about the experimental problems arising in high precision measurements in liquid crystals as well as a detailed description of the technical solutions we actually adopted in our experiment may be of some utility for researchers in the field. For this reason, we devoted this paper to an extensive discussion about the experimental aspects of our measurements and refer to paper I for a detailed exposition of the underlying physical problem. For the sake of completeness, however, the physical problem we are interested in is briefly exposed in the next section.

THE PHYSICAL PROBLEM

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 parallel glass plates coated for homeotropic alignment. If the beam is linearly polarized it carries no angular momentum, on the average, along its propagation direction (z-axis). When the beam intensity I_0 at the sample exceeds the OFT threshold I_{th}, a reorientation of the director $\hat{n}$ takes place in the sample. If the optical field would remain linearly polarized everywhere in the medium, one will expect the molecular director $\hat{n}$ be bound always in the plane of the beam polarization. Denoting with ϑ and ϕ the polar and azimuthal angles of $\hat{n}$ with respect to the polar axis z [$\hat{n} = (\sin\vartheta\cos\phi,\ \sin\vartheta\sin\phi,\ \cos\vartheta)$], only ϑ is then affected by the reorientation, while ϕ remains zero.

Now, a simple argument, based on angular momentum conservation, shows that this, indeed, cannot be 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 assimilate this perturbation to a small rotation, without twist, of the plane containing the distorted director field $\hat{n}(z)$. The absence of twist in the perturbation is expressed mathematically by the fact that $\delta\phi(t)$ is taken independent of the z-coordinate[5]. 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, eventually with a very small ellipticity. Net 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. Accordingly, the torque $\delta\tau_z$ may either damp out or enhance the initial fluctuation $\delta\phi$. The steady state $\hat{n}(z)$ is therefore stable or unstable, depending on the elicity value of the light emerging from the sample. A simple calculation shows that the ellipticity e of the light emerging from the sample is related to the ϕ-fluctuation $\delta\phi(t)$ by

$$e(t) = -\sin 2\delta\phi(t)\sin\alpha(t), \qquad (1)$$

where α is the phase difference between the extraordinary and ordinary waves accumulated in traversing the medium:

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

In Eq.(2) n_o and n_e denote the ordinary and extraordinary refractive indices of the liquid crystal, respectively, and L the sample thickness. Because the incident light is linearly polarized, the net torque along z acting on the sample is given by

$$\delta\tau_z(t) = (I_0/\omega)e = -\sin 2\delta\phi(t)\sin\alpha(t). \qquad (3)$$

We see that $\delta\tau_z$ has the same sign of $\delta\phi$ (and hence tends to enhance the ϕ-fluctuation producing unstability) if $n\pi<\alpha<(n+1)\pi$, with odd n. The stability of the $\phi=0$ state is reversed at intensities $I_0=I_n$ so that $\alpha = n\pi$ (n integer). For nematic films having thicknesses $L \gg \lambda$, the phase retardation α may change over many π as the laser intensity is varied. Consequently, the stability of the steady state $\hat{n}(z)=(\sin\vartheta(z),0,\cos\vartheta(z))$ can be reversed many times in a real experiment. Multistability and eventually hysteresis are therefore expected.

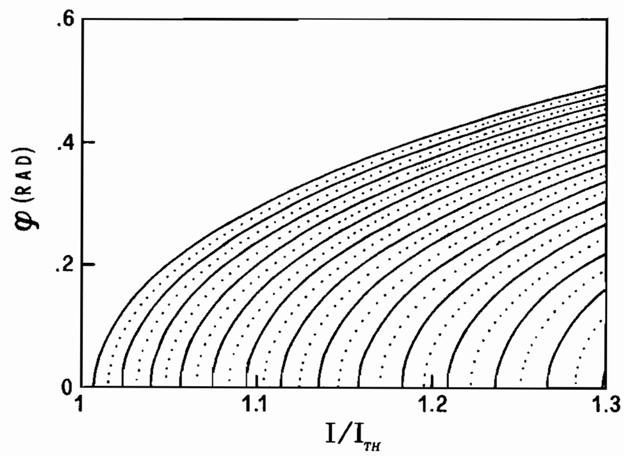

FIGURE 1 The director ϕ-angle vs. the incident light intensity. The solid lines refer to $\alpha=n\pi$ with odd n and are stable states; the dashed ones refer to $\alpha=n\pi$ with even n and are unstable states

Although we remand to paper I for detailed calculations, the main results are shown in Fig.1, where the steady state values ϕ_n of the azimuthal angle ϕ are plotted as functions of the incident reduced intensity I_0/I_{th}. The branches $\phi = \phi_n$ coalesce with the state $\phi = 0$ at characteristic threshold intensities I_n, given by

$$I_n = I_{th}[1 + 8n(B/u)(\lambda/n_o L)] \qquad (n = 1,2,\ldots), \tag{4}$$

where

$$\begin{aligned} B &= \tfrac{1}{4}(1 - k - 9u/4) \\ k &= 1 - k_{11}/k_{33} \\ u &= 1 - (n_o/n_e), \end{aligned} \tag{5}$$

k_{11} and k_{33} being Franck's elastic constant for splay and bend respectively. It is worth noting that along each ϕ-branch in Fig.1 the optical phase retardation α is fixed to $n\pi$, with integer n (n=1 on the first branch). Only branches corresponding to odd n are stable, however. Along these branches the sample behaves as a fixed retardation $\frac{1}{2}\lambda$-plate rotated at an angle ϕ_n with respect the polarization direction of the incident beam.

THE EXPERIMENTAL PROBLEM

The practical observation of these interesting effects poses particular problems. Firstly, we need to measure both the optical phase retardation α (which is related to the distribution of the director angle ϑ across the sample) and the azimuthal angle ϕ, because, unlike in the static-field case, we expect $\phi \neq 0$. Secondly, the values of α and ϕ must be determined with great accuracy (a few degrees) and, finally, the laser power must be controlled very carefully.

Although the first two points are evident, a brief discussion about the third one may be useful. The spacing $\Delta I = I_{n+1} - I_n$ between two successive branching points in Fig.1 is given by $\Delta I/I_{th} = 8n(B/u)(\lambda/n_o L)$. Due to the very small ratio λ/L, the quantity $\Delta I/I_{th}$ is very small, typically of the order of 2÷3‰, which is of the same order of magnitude of the fluctuations of commercial argon lasers and certainly much smaller that the steps in intensity performed in typical measurements on the OFT. Therefore, in order to resolve the multistable structure shown in Fig.1, a very stable laser source is needed and very small and accurate intensity steps must be performed.

On the other hand, in standard experiments on the OFT, only the phase α is usually monitored and, what's more, with a very poor accuracy (of the order of π). This is certainly true in the experiments where the sample birefringence was deduced from the number of diffraction rings in the far-field, but applies also (at least as order of magnitude) to the experiments where a probe laser beam was sent into the sample between crossed polarizers. As long as it concerns the ϕ-angle, all the experiments on the OFT performed so far have been not designed to evidentiate an eventual departure of the director $\hat{n}$ out of the initial plane, because it was simply assumed that this event was impossible. In our opinion this explains why the occurrence of a multistable structure of the OFT was not observed in previous experiments.

In our experiment, we used a COHERENT INNOVA-90 argon laser in "light control" mode. The power incident onto the liquid crystal was controlled by means of an electro-optic modulator. One hour was waited for laser thermal stabilization. This procedure ensured a long term stability of the power incident onto the sample better that 1‰.

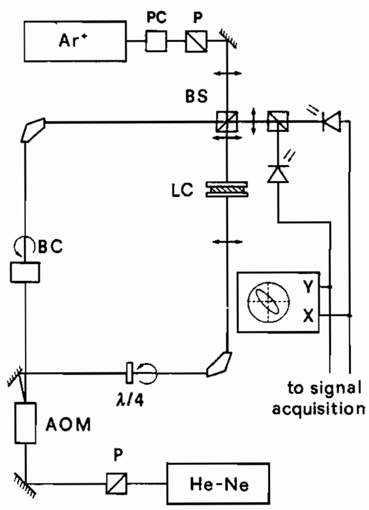

FIGURE 2 The experimental set-up. (AOM-acoustooptic modulator; BC-Babinet compensator; BS-beamsplitter; LC-liquid crystal cell; P-polarizer; PC-Pockel's cell

A real-time simultaneous measurement of the optical phase retardation α and the director azimuthal angle ϕ was obtained by detecting the phase difference and amplitudes of the horizontal and vertical components of the optical field of a probe He-Ne laser beam sent through the sample. To improve sensitivity, the liquid crystal cell was inserted into the signal arm of an heterodyne Mach-Zehnder interferometer. The apparatus is shown in Fig.2. The acousto-optic modulator shifted the frequency of the laser beam in the reference arm of 40MHz. The beat signals at 40MHz were detected by two photodiodes for both the horizontal and vertical polarization components. The phase difference and amplitudes of the beat signals were measured by an HP Vector Voltmeter and stored into an IBM-PC for further elaboration. Alternatively, the beat signals could be sent on the x- and y-axes of a digital oscilloscope to have a real-time visual image of the polarization ellipses of the light in the signal arm of the interferometer[6]. A Babinet-Soleil compensator was inserted in the reference arm for initial calibration of the apparatus. During the calibration, a rotating retardation plate was inserted in the signal arm to produce a known sequence of polarization states. The calibration curve is shown in Fig.3. With this apparatus the optical phase difference ψ and the amplitudes A and B of the horizontal and vertical components of the probe optical field emerging from the sample was measured with good accuracy. Our estimation was about 2° for ψ and about 1% for A and B.

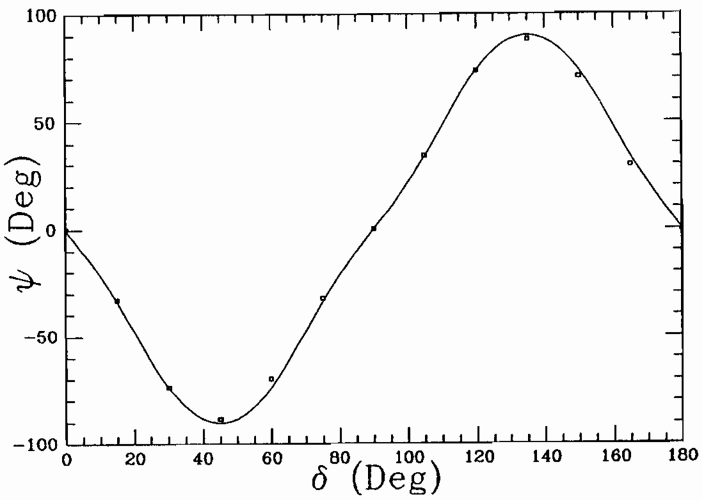

FIGURE 3 Calibration plot of the interferometer. The optical phase difference ψ is plotted vs. the angle of the axis of the calibration plate. The dots are the experimental point and the solid line is the theoretical curve.

The quantities ψ, A, and B, which are directly accessible to the experiment, are related to the phase α [Eq.(2)] and to the director azimuthal angle ϕ in a complicated way that it is worth of discussion.

Assume that the molecular director $\hat{n}$ is reoriented with negligible twist, so that it is bounded to a plane forming an angle ϕ with respect the horizontal plane. Then, from the optical point of view, the nematic film can be considered as a birefringent plate of retardation α oriented at the angle ϕ with respect to the horizontal plane. If the incident light were a plane wave, the distribution of $\hat{n}$ would be uniform in the x,y-plane and the measured quantities ψ, A, B will be related in a simple way to α and ϕ. Due to the finite cross-section of the pump beam, however, the distribution of $\hat{n}$ is not uniform and a lensing effect occurs so that part of the incident power is scattered away from the beam z-axis, forming a characteristic diffraction ring pattern in the far-field. The pump beam is usually focused over a spot few hundred of micron wide, so that this diffraction effect is sizable. In most cases, the power losses at the center of the beam exceed 50% of the total. The diffraction effect may be lower for the probe beam if it is focused tightly, so that its spot size at the sample is much smaller than the region over which $\hat{n}$ varies appreciably. In our experiment, the argon and He-Ne spot sizes at the sample were 100μm and 20μm, respectively; nevertheless an appreciable fraction (≧30%) of the probe power was still diffracted away from the interferometer.

A detailed description of the evolution of the polarization of a coherent light beam in the presence of diffraction due to aberrational lensing is difficult in isotropic media and also more difficult in birefringent media. As a rough approximation, however, we may say that, in our case, only the extraordinary component of the beam suffers the diffraction losses, while the ordinary component experiences an index distribution which is almost uniform. Then we may account for the diffraction losses just by supposing that the amplitude of the extraordinary component of the incident beam is attenuated of a factor τ in traversing the sample, the ordinary component remaining unaffected. The value of τ depends on the underlying diffraction mechanism, and, therefore, together with α and ϕ, it must be determined experimentally for each value of the pump intensity I_0. Since below the OFT no reorientation can be induced in the sample, τ=1 when $I_0 \leqq I_{th}$. In our experiment we used a circularly polarized probe beam. Then, for $I_0 > I_{th}$ we have $I_1 = A^2 + B^2 = 2A^2$ for the probe incident wave and $I_1' = A^2 + \tau^2 B^2 = \frac{1}{2}(1+\tau^2) I_1$ for the intensity of the light transmitted beyond the sample (absorption and scattering can be neglected). For circular polarization of the probe beam, a simple relationship between the factor τ and the apparent transmittance $T = I_1'/I_1$ of the sample can be deduced:

$$\tau = \sqrt{2T - 1}. \tag{6}$$

For other polarizations the formula is more complicated and depends on α and ϕ. The transmittance $T(I_0)$ can be directly measured, so that

$\tau(I_0)$ can be deduced as a function of the intensity I_0 of the pump beam. Once τ is found, a straightforward calculation based on Jones' matrices shows that the phase α and the angle ϕ are related to the measured quantities ψ , A, and B by

$$\sqrt{\frac{1+\tau^2}{1+\rho^2}}\begin{pmatrix}1\\ \rho^{i\psi}\end{pmatrix} = R(-\phi)\begin{pmatrix}\tau & 0\\ 0 & 1\end{pmatrix}\begin{pmatrix}e^{-\frac{1}{2}i\alpha} & 0\\ 0 & e^{\frac{1}{2}i\alpha}\end{pmatrix}R(\phi)\begin{pmatrix}1\\ -i\end{pmatrix}, \qquad (7)$$

where $\rho=B/A$ is the ratio between the vertical and horizontal components of the probe beam emerging from the sample and $R(\phi)$ is the 2x2 rotation matrix of an angle ϕ.

Relations (6) and (7) have been obtained by assuming that the laser-induced reorientation has negligible twist. Moreover, the effects of diffraction on the probe polarization have been accounted for in an approximated way. Nevertheless, we think that Eqs.(6) and (7) retain their practical validity and should be used in experiments where a complete information about the sample birefringence is required.

CONCLUSIONS

We presented an experimental apparatus which permits to have a complete information on the birefringence of a nematic film during the Optical Fréedericksz Transition. Spurious effects related to diffraction and self-focusing have been considered and a simple way to compensate for these effects was pointed out. Our experimental setup was specifically designed for the observation of the multistability and hysteresis expected during the OFT, but we believe that the experimental technique outlined here may be of general interest for people working in liquid crystals. The physical origin of multistability in the OFT was also briefly discussed. The measurements are actually in progress and will be presented in a forthcoming paper.

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