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COLLECTIVE ROTATION OF THE MOLECULES OF A NEMATIC LIQUID CRYSTAL DRIVEN BY THE ANGULAR MOMENTUM OF LIGHT

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Abstarct The experimental evidence of laser-induced collective rotation of the molecules of a nematic liquid crystal is presented. The effect is shown to have resulted by angular momentum transfer from the light to the medium.

INTRODUCTION

Optical-field induced molecular reorientation in nematic liquid crystals has received a great deal of attention in recent years[1]. When a c.w. laser beam is focused onto a homeotropically aligned nematic film at normal incidence, it was proven experimentally that the molecular reorientation can be effectively induced only if the light intensity exceeds the threshold characteristic

for the optical Freedericksz transition[2]. The underlying physical mechanism is the same, in essence, as in the Freedericksz transition induced by d.c. static fields. In fact, the light remains linearly polarized, during the transition, so that the liquid crystal molecules feel an average d.c. field proportional to the light intensity and directed along the polarization direction. A completely different mechanism arises if a c.w. elliptically polarized laser beam is used in the experiment. In this case, in fact, the optically induced molecular reorientation leads to a change in the beam polarization, as it traverses the sample. Since the polarization ellipticity varies, in general, angular momentum is transferred from the radiation to the birefringent medium. This phenomenon has no analogue in the case of d.c. external fields.

In this work, we proved experimentally that angular momentum can be continuously transferred from a circularly polarized laser beam to a homeotropically aligned nematic liquid crystal film. The angular momentum transfer results in a torque applied to the sample that, therefore, starts to rotate. The angular velocity of the rotation is determined by angular momentum conservation.

In our experiment, we measured the angular velocity of the laser-induced rotation of the liquid crystal molecules and compared it with the value predicted by angular momentum conservation in a simple theoretical model. The agreement between theory and experiment was

found very good near the threshold, where the model applies. The transition to the rotating regime was found to be first-order. the phenomenon is therefore intrinsically bistable. To our knowledge, this is the first time that a purely optically induced first-order transition is reported in nematics. For higher laser power, other interesting phenomena were observed, as nonlinear self-oscillations and also "deterministic cahos". We are still working to find a quantitative explaination for these effects. We believe, however, that angular momentum transfer should have an essential role.

EXPERIMENT

The rigid rotation of the liquid crystal structure is manifested, above the threshold, by a simultaneous rotation of the polarization ellipse of the light transmitted through the sample. We used a heterodyne polarimeter/interferometer experimental scheme that permits a real-time visual displaying of the light polarization ellipse on the screen of an oscilloscope[3]. The experimental details will be presented elsewhere[4]. As shown in Fig.1, a circularly polarized argon laser beam (λ = 514.5 nm) was focused by a 10cm. focal lens onto a homeotropically aligned 5-CB nematic liquid crystal film 65 μm thick at normal incidence. A fraction of the beam was splitted off and frequency-shifted by an acousto-optic modulator driven at 10 MHz. A cube beam-splitter and two crossed polarizers made the horizontal (H) and ver-

tical (V) linear components of the signal beam (coming from the sample) and the reference beam (coming from the modulator) to interfere. The heterodyne beating signals between the H- and V-components were detected by two photodiodes and sent to a sampling oscilloscope in XY mode for the visual displaying of the polarization ellipse.

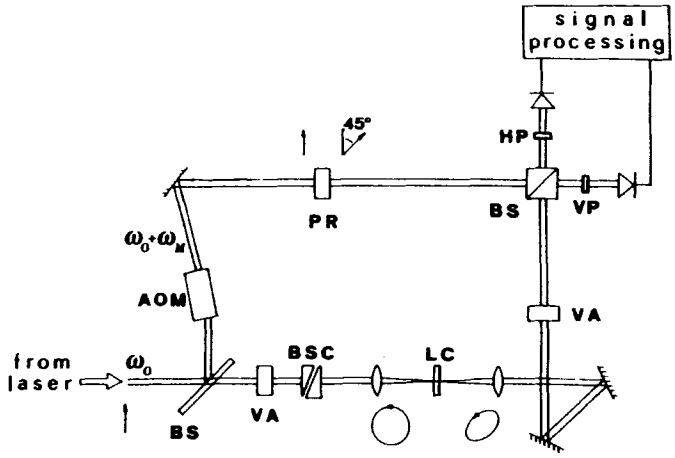

FIGURE 1 The experimental apparatus.

No effect was observed as long as the light intensity was below a critical threshold and the polarization of the light transmitted through the sample remained circularly polarized. Above the threshold, however, the transmitted light became elliptically polarized and the polarization ellipse was seen continuously rotate on the oscilloscope

screen. The period of the rotation ranged from 40 to 50 seconds. The experimental value of the threshold intensity was $I_{th} = 2.1$ kW/cm^2., in good agreement with thevalue 2.2 kW/cm^2. calculated from the expression

$$I_{th} = (ck_{33}/n_o)(2n_e^2/(n_e^2-n_o^2))(\pi/L)^2 \qquad (1)$$

yielding the threshold for the optical Freedericksz transition for circularly polarized light[5]. In Eq.(1) n_o and n_e are the ordinary and extraordinary refractive indeces of the liquid crystal and k_{33} is the elastic constant for bend. In the calculation we used the values of the material constants of 5-CB, as found in the literature ($n_o = 1.53$, $n_e = 1.70$, $k_{33} = 4.4 \times 10^{-7}$ dyne).

The transition from the undistorted state to the rotating ellipse regime was found to be first-order. This is clearly shown in Fig.2, where the ellipticity change Δs_3 suffered by the beam in traversing the medium is reported as a function of the normalized light intensity I/I_{th}. A bistability loop is clearly visible. This intrinsically bistable optical phenomenon does not require a resonant cavity or a static external field. Provided the incident power is changed very slowly, the hysteresis cycle cycle is well reproducible. The occurrence of a hysteresis loop is due to the fact that the threshold for the optical Freedericksz transition is lower for elliptically polarized light than for circularly polarized light[5]. Therefore, above the threshold, where the

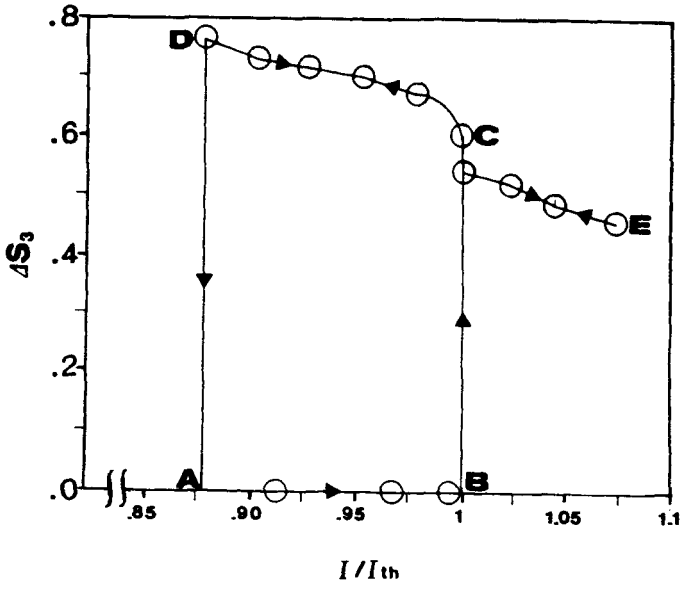

FIGURE 2 Ellipticity change of the beam vs. the normalized light intensity I/I_{th}.

light becomes elliptically polarized, the Freedericksz threshold is reduced. Then, once the rotation regime is reached, the incident power must be reduced substantially below I_{th} before the elliptical polarization in the medium becomes insufficient to sustain the molecular reorientation. This explains the up transition BC and the down transition DA shown in Fig.2. The BC and CE branches were available in both directions; but, while the former was stable, the latter was metastable. After a certain time (from 10 to 60 minutes), in fact, a many ring diffraction pattern showed up

in the far-field. During the formation of the pattern, the evolution of the polarization was quite complicate, until a stationary regime was reached where the the system started again a constant rotation of the polarization ellipse, but at the much lower rate of one turn in about 10 minutes. We noted, however, that, if the same power level was reached suddenly, the number and diameter of the far-field diffraction rings started to oscillate in time. This fact suggests that in this regime the liquid crystal molecules undergo a precession and a nutation motion simultaneously. We are still in the process of finding a quantitative explaination for these phenomena. We have compared, however, the angular velocity of the rotation of the polarization ellipse as measured by our apparatus with the value predicted by angular momentum conservation[4]:

$$\Omega = \pm (I/I_{th})(k_{33}/\gamma_1)(\pi/L)^2(\Delta s_3/2\arcsin(\Delta s_3/2)). \quad (2)$$

In Eq.(2) Ω is the angular velocity of the rotation, L is the sample thickness, γ_1 is the Leslie viscosity coefficient (γ_1 = 0.38 poise for 5-CB) and Δs_3 is the ellipticity change suffered by the laser beam in traversing the sample. Expression (1) was obtained from a simple model where the liquid crystal molecules are supposed to lie in a plane rotating with angular velocity Ω. This kind of motion is made possible by the homeotropic alignment of the

molecules at the sample walls. It should be stressed that Eq.(2) is a direct consequence of angular momentum conservation.

In Fig.3 the measured angular velocity Ω of the polarization ellipse rotation (normalized to the value I/I_{th}) is plotted as a function of the ellipticity change Δs_3 as measured by our apparatus. The continuous line is obtained from Eq.(2).

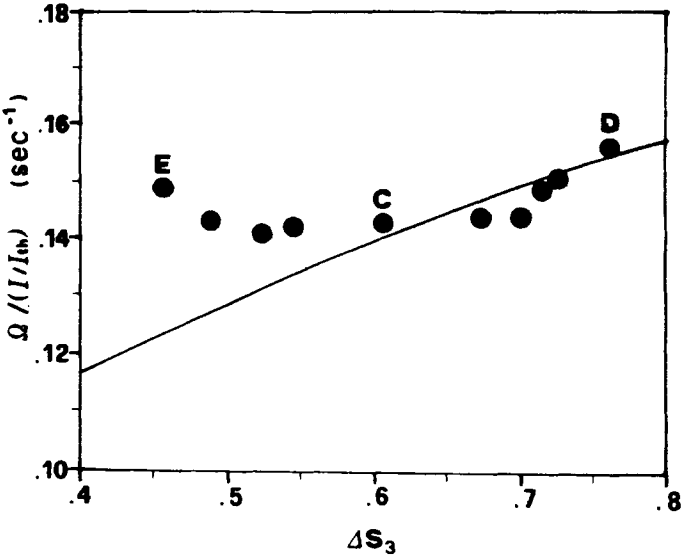

FIGURE 3 Measured angular velocity Ω of the polarization rotation vs. the ellipticity change Δs_3.

We see that the agreement is good for the points on the CD branch in Fig.2, while it is poor for the points on the CE branch. Although the angular momentum conservation in the

process should be valid in general, the result (2) is based on the assumption of a negligible twist in the rotating structure. We conclude, therefore, that only the near-threshold CD branch corresponds effectively to a negligible twist, while for the CE branch the twist is large enough to invalidate relation (2).

We used both right- and left-handed circular polarization for a fixed power of the incident light. Equal angular velocities were observed, in the ellipse rotation regime, but the sense of the rotation was reversed, according to the $\pm$ sign in Eq.(2). From all these observations we conclude that indeed angular momentum is continuously transferred, in this experiment, from the circularly polarized laser beam to the liquid crystal, above the threshold for the optical Freedericksz transition.

We also realize that the induced molecular precession requires a deposition of the beam energy in the medium. This energy is dissipated by the viscous forces. Since the medium is transparent, this can only happen if part of the beam is down-shifted in frequency. The rotation of the polarization ellipse of the output light means that its right and left circular components have a frequency difference 2Ω. Since the input light has frequency ω and right-handed circular polarization, say, we conclude that left-handed circularly polarized photons at the new frequency $\omega' = \omega - 2\Omega$ are coherently generated in the process. With a slight modification of our apparatus we were able to measure direc-

tly the frequency shift of the ω' component in the transmitted light. A $\lambda/4$ retardation plate beyond the sample has been inserted in order to separate the right and left circular components of the transmitted beam. The beating signals detected by the photodiodes (see Fig.1) were compared with the signal coming from the local oscillator. Since the heterodyne signal was near to the zero frequency, the noise was comparatively large. For this reason, we reported in Fig.4 a,b the power spectrum of the beating signal coming from the left (a) and right (b) circular components of the transmitted beam. The frequency shift (68mHz) of the spectrum of the left circular component is evident.

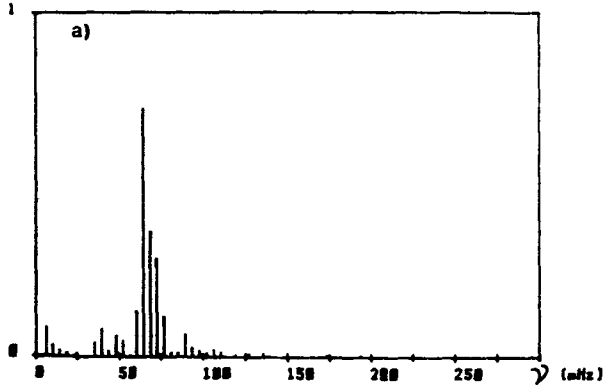

FIGURE 4a Power spectrum of the beating signal coming from the left circular component of the beam transmitted through the sample. The spectrum is centered at 68 mHz.

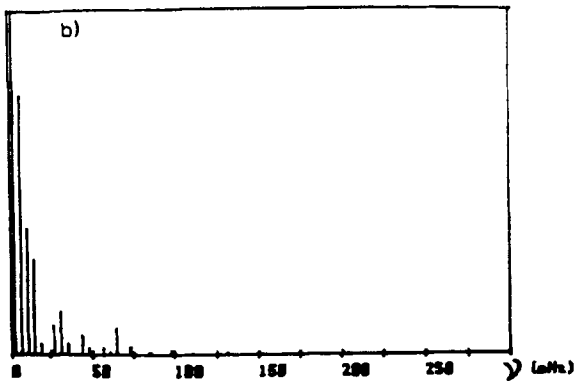

FIGURE 4b Power spectrum of the beating signal coming from the right circular component of the beam transmitted through the sample. The spectrum is centered at the zero frequency.

In this respect, we can also regard the present nonlinear optical effect as a stimulated scattering process where right circular photons of frequency ω are changed into left circular photons of frequency ω'. The details of the theoretical description of this scattering process will be presented elsewhere.

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