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PHOTODYNAMICAL EFFECTS INDUCED BY THE ANGULAR MOMENTUM OF LIGHT IN LIQUID CRYSTALS

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ABSTRACT

We report the observation of a very rich reorientational dynamics ofthe molecular director of a 5CB nematic film, induced by an elliptically polarized cw-laser beam above the threshold for the Optical Fréedericksz Transitionas well as the occurrence of multistability and of locking of the optical phase in the case of linear polarization of the incident beam. In despite of the very complicated character of the laser-induced reorientational motions, a simple model, based on continuum theory and angular momentum conservation reproduces very well the experimental findings.

1.INTRODUCTION

Laser-beam propagation in nematic liquid crystals exhibits very unique and fascinating nonlinear optical effects. It has been demonstrated that a sufficiently strong laser field can induce a Fréedericksz transition (an orientational transition) in a nematic film[1]. In a recent experiment[2]it was shown that a time-dependent polarization rotation is induced in a homeotropically aligned liquid-crystal film by a circularly polarized cw-laser beam at normal incidence. This remarkable effect was later ascribed to a new nonlinear optical effect, Self-Induced Stimulated Light Scattering[3]. The phenomenon is caused by the constant deposition of angular momentum from the field to the medium, inducing a collective uniform precession of the liquid crystal molecules about the beam propagation direction. In the process, energy is dissipated by viscous forces in the medium. Since the medium is transparent, the energy loss appears as a red shift on part of the beam. This red shift was also measured[4].

Self-Induced Stimulated Light Scattering occurs also when an elliptically polarized laser beam is sent onto a homeotropically aligned liquid-crystal film, but, in this case, the process becomes much more complicated, because both circular components σ^+ and σ^- are present in

the incident beam. Photons σ^+ and σ^- transfer opposite angular momenta to the medium so that the actual torque on the molecules of the liquid crystal depends on their delicate balancing. Moreover, the azimuthal symmetry of the system (field and matter) is broken. This yields characteristically different results in the two cases. The elliptically polarized input can lead the system through various dynamic regimes: torsional oscillation, nonuniform precession, nutation superimposed on precession and others. Experimental observations are in good agreement with teoretical prediction from a continuum model of laser-beam propagation in an anisotropic fluid with reorientational internal degrees of freedom[5].

The case of linear polarization at normal incidence is exceptional in the sense that no angular momentum is transferred to the medium along the beam direction. This results in a simpler dynamics in which the system always reaches a final equilibrium state[1]. Nevertheless, both theory and experiment demonstrate that also in this case angular momentum transfer from the optical field to the medium plays an important role, producing the occurrence of remarkable nonlinear optical phenomena like multistability, hysteresis and optical phase locking[6].

In this work we report the first experimental evidence of the various dynamical regimes induced in a nematic liquid crystal by an elliptically polarized cw-laser beam as well as the occurrence of hysteresis and optical phase-locking when linearly polarized light at normal incidence was sent onto the sample.

2.THE EXPERIMENT

In our experiments we used a 75μm-thick nematic film of 4-cyano-4'-pentyl-biphenyl (5CB) sandwiched between two glass plates coated with DMOAP for homeotropic alignment (molecular anchoring normal to the walls).

The experimental arrangement is shown in Fig.1. The pump argon-laser beam (λ=0.515μm) propagating along the z-axis was focused at normal incidence onto a 5CB sample by means of a 15-cm focal lens. The polarization and the intensity of the argon laser could be changed independently by a Pockels cell and a variable attenuator. The optically-induced time-dependent molecular reorientation was probed through a counter-propagating He-Ne beam focused at the same point as the argon beam. The spots of the He-Ne and of the argon laser at the sample were $\simeq$ 70μm and $\simeq$120μm diameter, respectively. The probe He-Ne beam was circularly polarized by a $\lambda/4$ plate put in front of the sample and inserted in a heterodyne interferometer/polarimeter scheme[7]. The heterodyne polarimeter was already described in Ref.2, to which we refer for details. This apparatus permits a real-time monitoring of the complete polarization state of the He-Ne beam after its passage through the sample. The polarization ellipse of the He-Ne beam could be either observed directly on the screen of a digital oscilloscope or sent to a vectorial voltmeter and/or to an IBM-PC computer for data processing. Circular polarization was chosen for the probe beam in order to decouple the polar and azimuthal degrees of freedom of the molecular director during the observations.

The data were taken by fixing the polarization of the argon beam by the Pockels cell and varying its intensity in steps of about 5mW each in the experiment with elliptical polarization and of about 1mW each in the experiment with linear polarization, using the on-line attenuator. The long-term fluctuations of the pump intensity were found to be less than 1%.. After each intensity change, we waited until the final regime was

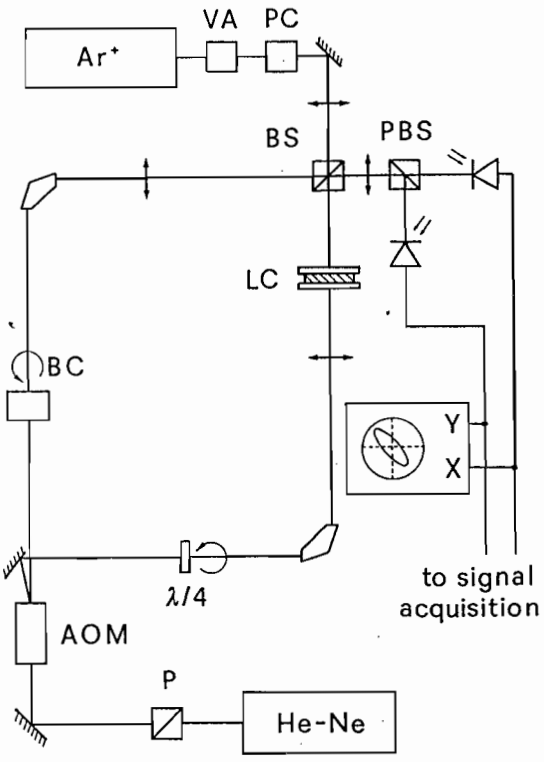

Fig.1 - Experimental set-up [AOM - Acousto-Optic Modulator; BC - Babinet Compensator; BS - Beam Splitter; LC - Liquid Crystal cell; P - Polarizer; PBS - Polarizing Beam Splitter; PC - Pockel's Cell; VA - Variable Attenuator].

attained. The transients ranged from about 30 to 60 sec., a much longer time than the relaxation time at laser switching off, which was of a few seconds. When a given regime was reached, however, it remained stable for hours.

In the case of elliptical polarization, different dynamical regimes were observed, depending on the polarization ellipticity s_3 and on the intensity I of the pump beam. Our observations are summarized in Fig.2, where the plane (s_3,I) of the control parameters has been divided in several regions, pertaining to different kind of motions induced in the sample. In the figure, the intensity I is normalized to the intensity $I_{th}(0)$ needed to induce the optical Fréedericksz transition for linear polarization of the argon beam. The advantage is that the ratio $I/I_{th}(0)$ was determined experimentally by the corresponding input power ratio, without any need of accurate determination of the beam cross-section at the sample, which usually introduces large errors in the measurements.

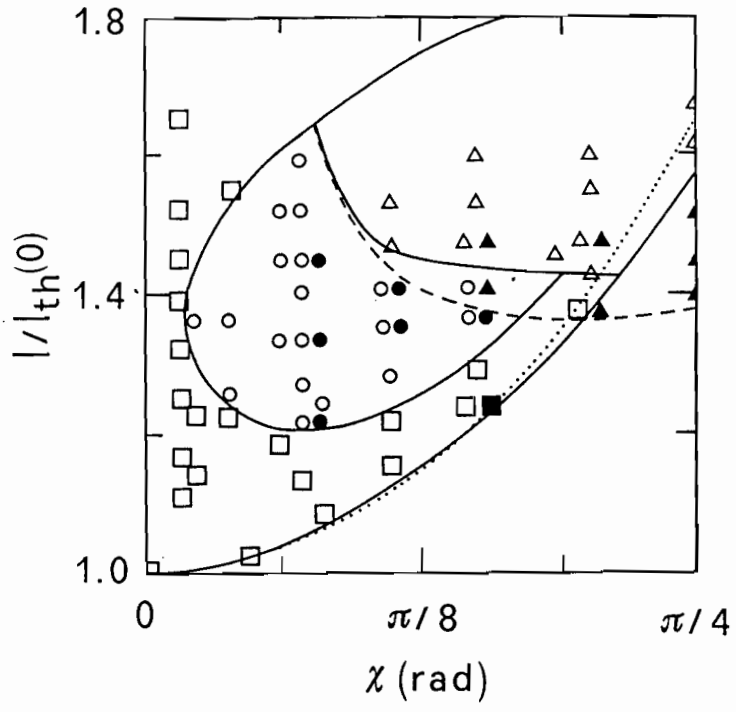

Fig.2 - Zone diagram in the (χ,I) plane of the observed dynamical regimes: χ is the pump beam polarization ellipticity angle defined by $\sin 2\chi = s_3$ and $I/I_{th}(0)$ is the normalized beam intensity. Squares refer to steady-state regime, circles to oscillations, triangles to rotations; filled-in symbols refer to regimes observed by decreasing the intensity. Dashed line is the temperature corrected theoretical threshold intensity.

Let us describe the laser- induced reorientation in the liquid crystal sample by the polar angles (ϑ, ϕ) of the molecular director **n**. For a fixed ellipticity, no reorientation was observed in the sample until the intensity threshold I_{th} for the optical Fréedericks transition was reached. Let us consider the case of general elliptical pump polarization first. Slightly above the threshold, the azimuthal and the polar angles ϕ and ϑ of **n** were found to reach a final steady-state. At higher argon intensities, the molecular director undergoes a small irregular fluctuation around its equilibrium position, that gets stronger and more regular increasing the pump power, until persistent neat oscillation of ϕ and ϑ occurs. The transition from the "noisy" steady-state and the persistent oscillations regime is smooth. An example of these oscillations is shown in Fig.3. At higher intensities, a new critical value of I is attained, beyond which the director **n** starts to

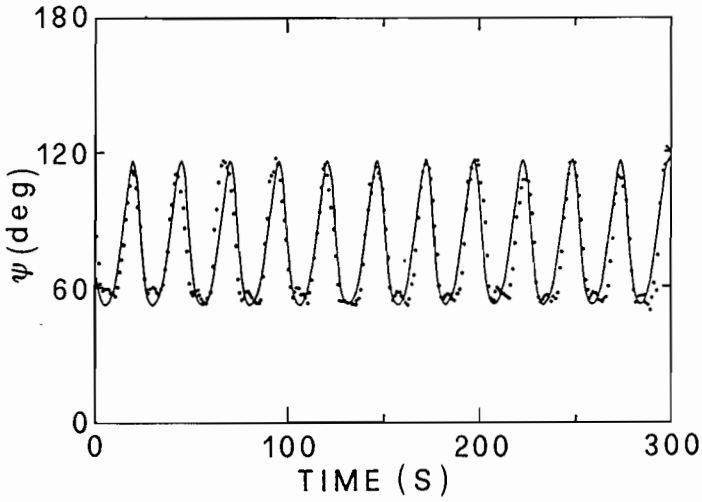

Fig.3 - Typical oscillatory regime observed for $\chi = 0.35$ rad and $I/I_{th}(0) = 1.41$ [ψ is the angle between the polarization ellipse major axis and a reference direction].

precess continuously around the beam propagation direction. The switching to precession is discontinue. The angular velocity of the precession motion is not uniform, in general, and it is always associated to nutation. The presence of nutation is manifested by the oscillation in time of the ellipticity s_3 of the probe beam emerging from the sample. The presence of precession is manifested by the rotation of the polarization ellipse of the probe beam, as shown in Fig.4.

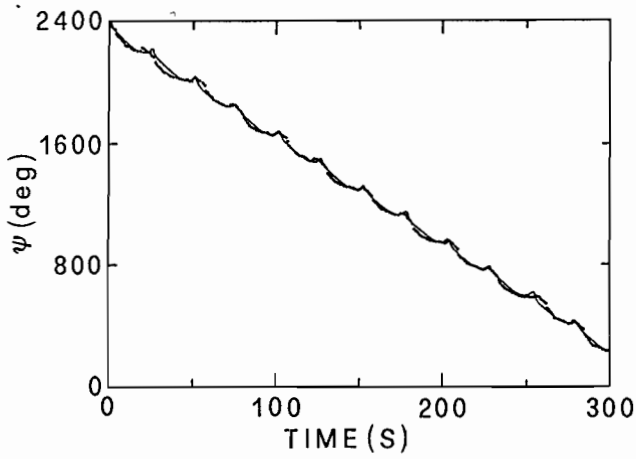

Fig.4 - Typical rotatory regime observed for $\chi = 0.35$ rad and $I/I_{th}(0) = 1.53$ [ψ is the angle between the polarization ellipse major axis and a reference direction].

When the pump intensity is lowered, the persistent oscillation regime is reached again, but at a lower transition intensity. In other words, hysteresis was found between the two time-dependent stable states of persistent oscillations and of precession/nutation motion. This is shown by the dashed region in Fig.2. As long as it concerns the transition back from the persistent oscillation regime to the steady-state regime, we observed regular oscillation at intensities where before irregular fluctuations were found. Unlike the case of increasing intensities, the transition to the steady-state regime is now sharp.

In the case of circular polarization, oscillations were absent and, above the threshold, the system was put directly into a uniform rotatory motion, as already reported in Ref.1. The direct transition to rotation was observed also for ellipticity values close to the circular one. Finally, in the case of linear pump polarization, a monotonic decay towards a final steady-state was always observed, above the threshold. All these regimes are also shown in Fig.2.

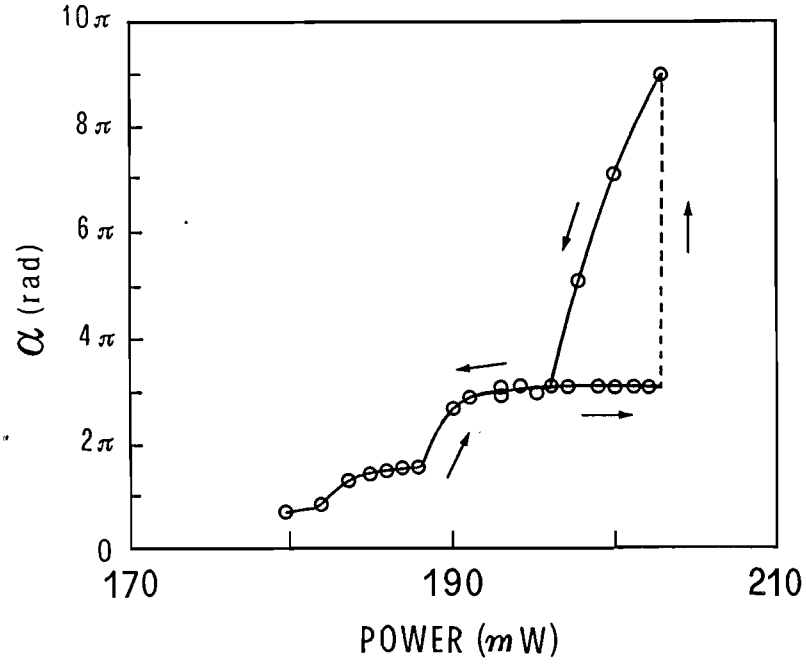

Fig 5 - Birefringence angle α as a function of the power of the linearly polarized incident laser beam.

The case of linear polarization of the incident beam, however, exhibits particular features. In Fig.5 we plotted the otpical phase retardation between the ordinary and extraordinary wave of the probe beam as a function of the argon intensity. A large hysteresis loop is found, having a characteristic plateau where the optical phase α is locked to a nearly constant value of about π. When the argon laser intensity was varied across the range corresponding to the plateau, the sample behaved as a fixed retardation plate of $\frac{1}{2}\lambda$ whose orientation changes with the

pump intensity. The output polarization of the pump beam, in fact, was linear as in the input light, but rotated of an intensity dependent angle. The locking of the optical birefringence of the sample was confirmed also by the fact that, in the plateau, the number of diffraction rings in the far-field beyond the sample was "frozen" to a constant value even when the pump intensity was increased.

2.DISCUSSION

We now consider some quantitative features of the observed phenomena. The threshold I_{th} for the Fréedericksz transition is given by[8]

$$I_{th} = \frac{2I_{th}(0)}{1 + \sqrt{1 - s_3^2}}, \qquad (1)$$

where $I_{th}(0)$ is the threshold for linear polarization and s_3 is the ellipticity of the incident beam. Relation (1) shows that for circular polarization the threshold should be twice $I_{th}(0)$. From our measurements we found a ratio $I_{th}(1)/I_{th}(0) \cong 1.6$. The discrepancy with respect to the theoretical ratio of 2 is far beyond the experimental errors. We ascribe this to thermal effects due to laser heating. The material constants [which are encoded in $I_{th}(0)$] are temperature dependent, in fact, since the order parameter S of the nematic material changes with temperature according to the empiric law (for 5CB)[9]

$$S = (1 - T/T^*)^{0.22}. \qquad (2)$$

where T^* is an effective temperature very close to the nematic-to-isotropic transition temperature. T^* was experimentally related to the intensity I^* to which the clearing of the sample was observed. The resulting temperature corrected threshold curve as a function of ellipticity is drawn in Fig.2. The agreement with experiment is good.

The equation of motion governing the molecular director **n** are obtained by balancing the elastic, viscous and optical torques τ_e, τ_v, and τ_o in the medium[10] and have been numerically integrated by using a library routine for parabolic partial differential equations on a Digital VAX 2000 station. In the numerical computation, tabulated values were used for the material constants of 5CB [11] ($k_{11} = .7\ 10^{-6}$dyne, $k_{22} = .5\ 10^{-6}$dyne, $k_{33} = .9\ 10^{-6}$ dyne, $n_o = 1.52$, $n_e = 1.7$), without any fitting procedure. The results are in excellent agreement with the experimental observations (see full-line curves in Figs.3 and 4). The model yields also the correct succesion of the observed dynamical regimes as shown in Fig.2 (see Ref.5).

We conclude with a few words about the optical phase-locking phenomenon in the case of linear polarization of the incident beam. For intensity above the threshold, the director **n** can rotate out of the plane containing the beam polarization. If this occurs, the output light will be elliptically polarized and net angular momentum is exchanged with the medium. Depending on the intensity, the resulting torque on the sample may drive **n** further away or restoring it towards the plane. Only if the light emerging from the sample is still linearly polarized, no torque is

exerted and equilibrium can be reached. Now, the output polarization is linear only if the optical phase retardation is $n\pi$, with n integer. This explains why at equilibrium the optical phase is found locked at π. Multistability arises because steady-states exist for any n. A more detailed inspection shows that only states with odd n are stable. This also is confirmed by the experiment.

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