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Photodynamical Effects Induced by Laser Radiation in a Nematic Liquid Crystal

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The observation of a very rich reorientational dynamics of the molecular director of a nematic liquid crystal film, induced by an elliptically polarized cw laser beam, is reported. Despite of the complicated laser-induced motions, a simple model, based on angular momentum conservation reproduces fairly well the experimental findings.

Keywords: nonlinear optics, nematic liquid crystal, dynamics

INTRODUCTION

In a recent experiment[1] it was shown that a time-dependent polarization rotation can be induced by a circularly polarized cw-laser beam in a homeotropic liquid-crystal film with molecules aligned parallel to the surface normal. The laser beam, indeed, not only can produce a distortion in the homeotropic alignment of the molecules but it can also transfer part of its angular momentum to the medium inducing a collective uniform precession of the molecules about the normal to the sample walls. In the case of an elliptically polarized incident laser beam, the process can become even more complicated. The laser-induced molecular rotation will not be uniform, in general, and, in some cases, it may reduce to an oscillatory motion rather than a rotating one. Moreover, unlike the case of circular polarization, the angle between the molecular director $\hat{\mathbf{n}}$ and the major axis of the polarization ellipse at a fixed point in the medium may also change with time. Since the transverse optical torque on the molecules is maximum when $\hat{\mathbf{n}}$ is parallel to the major axis of the polarization ellipse and minimum when it is orthogonal, a nutation motion superimposed to the precession of the molecules may occur.

Here we report the observation of the various dynamical regimes induced by an

elliptically polarized cw-laser beam in a homeotropic nematic film. The experimental observations are in good agreement with theoretical predictions based on a continuum model describing the laser-beam propagation in an anisotropic fluid.

THE EXPERIMENT

The experimental set-up is shown in Figure 1. An Ar^+-laser beam was focused to a spot of 120 μm at normal incidence onto a 75 μm homeotropic film of 4-ciano-4′-pentyl-biphenyl (5CB). The intensity and the polarization of the pump laser could be changed independently by means of a variable attenuator and a Pockels cell, respectively. A counterpropagating He–Ne laser beam, focused to 70 μm in the same region of the pump laser, was used to probe the optically-induced time-dependent molecular reorientation of the liquid crystal molecules. The He–Ne beam was circularly polarized and its polarization state at the output from the nematic sample was analyzed by a heterodyne interferometer/polarimeter.[2] By means of this apparatus we had a real-time monitoring of the probe beam polarization after transmission by 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 an IBM personal computer for data processing.

The probe beam was circularly polarized in order to decouple the polar and

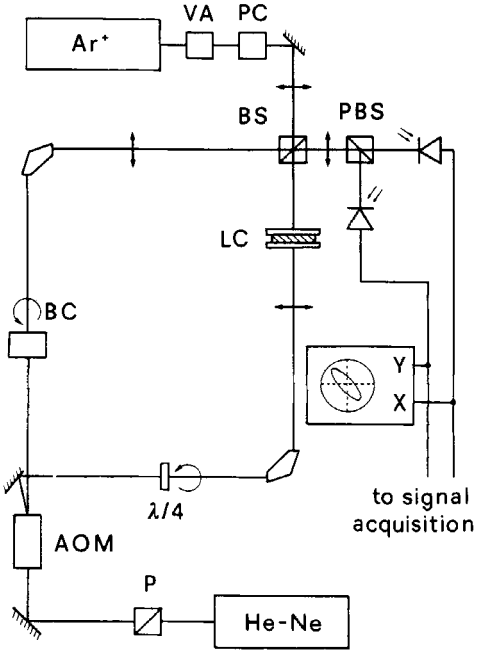

FIGURE 1 Experimental set-up [AOM-acoustooptic modulator; BC-Babinet compensator; BS—beamsplitter; LC—liquid crystal cell; P—polarizer; PBS—polarizing beamsplitter; PC—Pockel's cell; VA—variable attenuator].

azimuthal degrees of freedom of the molecular director during the observations. Assuming, indeed, a negligible twist in the molecular orientation of the film, for a circularly polarized incident beam, the output light from the sample is elliptically polarized with the ellipse major axis directed at 45° with respect to the plane formed by the director $\hat{\mathbf{n}}$ and the surface normal. The ellipticity S_3 of the output probe beam is given by

$$S_3 = \pm\cos(\Delta\alpha),$$

where $\Delta\alpha$ is the phase difference accumulated by the extraordinary and ordinary waves in traversing the sample. The double sign accounts for the right- or left-handed character of the circularly polarized light of the probe. Taking the $\hat{\mathbf{z}}$-axis directed along the normal to the sample walls, $\Delta\alpha$ is given by

$$\Delta\alpha = \frac{2\pi}{\lambda}\int_0^d \left[\frac{n_0 n_e}{\sqrt{n_e^2\cos^2\vartheta + n_0^2\sin^2\vartheta}} - n_0\right] dz \tag{1}$$

In Equation 1, n_0 and n_e denote the ordinary and extraordinary indeces of the liquid crystal, $\vartheta(z, t)$ the director polar angle and d the sample thickness. The ellipticity S_3 as well as the angle ψ, formed by the polarization ellipse major axis and a reference $\hat{\mathbf{x}}$-axis fixed in the laboratory, are measured by our apparatus, allowing a real-time tracking of the liquid crystal reorientational motion.

Once the polarization state of the argon beam was fixed by the Pockel's cell, the data were taken varying the power of the pump beam in steps of 5 mW using the on-line attenuator. For each intensity change, the system went into a steady state after a transient period ranging from 30 to 60 sec. Different dynamical regimes were observed at steady state, depending on the polarization ellipticity $S_{3p} = \sin 2\chi$ and on the intensity I of the pump beam. The observations are summarized in Figure 2(a), where the plane (χ, I) of the control parameters has been divided in different regions, each pertaining to different kinds of motions induced in the sample. To avoid inaccuracy in the determination of the beam cross-section at the sample, the intensity I is normalized to the intensity I_{th}^0 needed for the linearly polarized argon beam to induce the optical Freédericksz transition in the sample.

Let us consider the case of a general elliptical pump polarization first. Slightly above the threshold the azimuthal and polar angles ϕ and ϑ of $\hat{\mathbf{n}}$ were found to reach a final steady-state. At higher pump intensities, the molecular director was found to reach an equilibrium state with small random fluctuations around it. These fluctuations became stronger and more regular increasing the pump power, until a regular and persistent oscillation of ϕ and ϑ occurred. This corresponds to a libration of $\hat{\mathbf{n}}$. The transition from the "noisy" steady-state to the persistent oscillations regime is smooth. At higher intensities, another critical value of intensity, say I_c, was reached, beyond which the director $\hat{\mathbf{n}}$ starts to precess continuously around $\hat{\mathbf{z}}$. The switching to precession is rather abrupt. Moreover, this precession motion has an angular velocity which is not uniform and it is always associated to nutation. The presence of precession is manifested by the rotation of the polari-

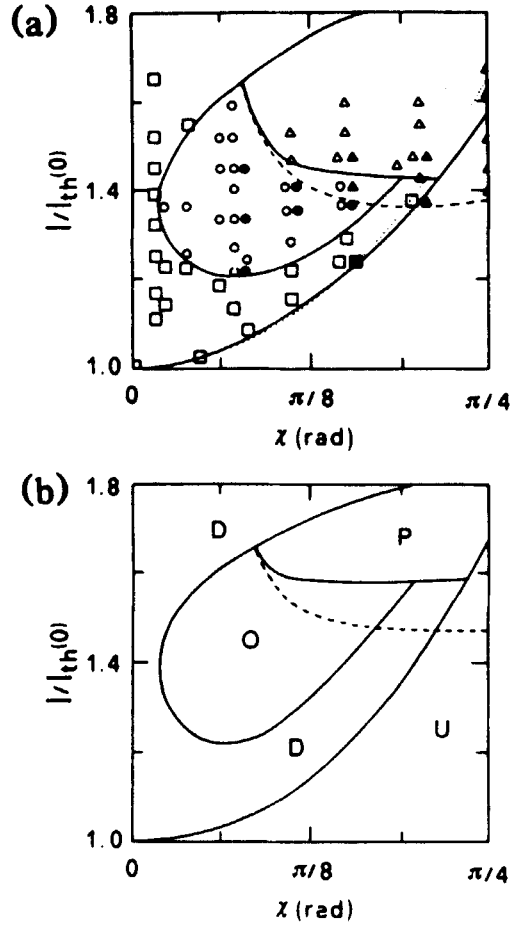

FIGURE 2 "Phase" diagrams of various dynamic regimes in the (χ, I) plane. (a) Experimental observations: squares refer to distorted equilibrium, circles to persistent oscillation, triangles to precession-nutation. Open symbols refer to states obtained with increasing laser intensity and solid symbols to states with decreasing intensity. (b) Theoretical simulation. U, D, O and P refer to undistorted, distorted-equilibrium, persistent-oscillation and precession-nutation regimes, respectively. Solid lines describe boundaries between different dynamic regimes. The dashed line describes the boundary at which from the P state the system switches back to U, D, O as the pump intensity decreases.

zation ellipse while the nutation is manifested by the oscillation in time of the probe output polarization ellipticity S_3.

When the pump intensity is decreased, the persistent oscillation regime is reached again, but at a lower transition intensity I_c'. This means that an hysteresis was found between the two states of persistent oscillation and precession/nutation.

Reducing the polarization ellipticity of the pump beam, we found that at high intensities, the system went no longer in the precession-nutation regime but changed from persistent-oscillation to stable distorted-equilibrium state. If the pump polarization ellipticity is further reduced close to linear, a monotonic decay towards a final equilibrium state was always observed. On the other hand, for pump po-

larization very close to circular, oscillations were absent and, above threshold, the system was directly put into a uniform rotatory motion, as reported in Reference 1.

THEORETICAL MODEL AND CONCLUSIONS

The equation of motion governing the molecular director $\hat{\mathbf{n}}$ are obtained balancing the elastic, viscous and optical torques τ_e, τ_v and τ_0 in the medium.[3] Backflow effects are neglected and $\tau_v = \gamma\,(\hat{\mathbf{n}} \times \partial\hat{\mathbf{n}}/\partial t)$ is assumed, with γ being an effective viscosity coefficient.[4] The torque balance equation must be coupled to Maxwell's equations, that, in the slow-envelope approximation and for transparent birefringent media, reduce to a simple precession equation for the reduced Stokes vector $\mathbf{S} = (S_1, S_2, S_3)$ which describes the polarization state of the pump laser beam

$$\frac{\partial \mathbf{S}}{\partial z} = \Omega \times \mathbf{S} \tag{2}$$

with $\Omega = (\omega\Delta n/c)\,(\cos 2\phi, \sin 2\phi, 0)$, ω being the pump frequency and $\Delta n(\vartheta)$ the integrand in Equation 1. A consequence of the torque equations and of Equations 2 is the angular momentum equation

$$\int_0^d \sin^2\vartheta(\partial\phi/\partial t)\,dz = (I/\omega)\Delta S_3, \tag{3}$$

where $\Delta S_3 = S_3(d) - S_3(0)$ is the ellipticity change suffered by the laser beam in traversing the sample. We notice that for an equilibrium state $\partial\phi/\partial t = 0$, so that $\Delta S_3 = 0$ is expected. This was checked experimentally by measuring the pump beam ellipticity beyond the sample.

The torque balance equations and the polarization Equation 2 have been numerically integrated by means of a library routine for parabolic partial differential equations on a Digital VAX Station 2000. In the numerical computation, tabulated values were used for the material constants of 5CB[5] ($k_{11} = 0.7 \times 10^{-6}$ dyn, $k_{22} = 0.5 \times 10^{-6}$ dyn, $k_{33} = 0.9 \times 10^{-6}$ dyn, $n_0 = 1.52$, $n_e = 1.7$) without any best-fit optimization. The angular momentum relation (3) was used as a global check for the numerical routine.

In order to simulate the experimental conditions, the intensity I was varied in small steps for a fixed polarization ellipticity and the resulting final distributions of $\vartheta(z, t)$, $\phi(z, t)$ and $\mathbf{S}(z, t)$ were taken as initial conditions for the successive run. The numerical results are reported in Figure 2(b). A comparison with the experimental observations shows that all the dynamic regimes observed in the experiment were reproduced in a correct successive order and that the matching between the two diagrams was qualitatively good. Also the hysteresis between the precession/nutation and the persistent-oscillation regime was reproduced by the model.

In summary we have found that, above the Freédericksz transition, an elliptically

polarized laser beam can induce very complicated time-dependent motions in a nematic film. The effects are correlated to the deposition of angular momentum from the beam to the medium. Unlike the case of circular input polarization, the transfer of angular momentum may be in both directions, resulting, eventually, in a persistent collective oscillation of the liquid crystal molecules. Depending on the pump beam intensity and input ellipticity, several dynamic regimes were observed: distorted equilibrium state, persistent-oscillation and precession-nutation regimes. These findings are in substantial agreement with the predictions of a numerical model consistent with angular momentum conservation.

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References

1. E. Santamato, B. Daino, M. Romagnoli, M. Settembre and Y. R. Shen, *Phys. Rev. Lett.*, **57**, 2423 (1986).
2. R. Calvani, R. Caponi and F. Cisternino, *Opt. Comm.*, **54**, 63 (1985).
3. An explicit expression of the elastic and optical torques in terms of the director angles ϑ and ϕ is given in E. Santamato, G. Abbate, P. Maddalena, *Phys. Rev. A*, **38**, 4323 (1988).
4. F. M. Leslie, *Quart. J. Mech. Appl. Phys.*, **19**, 357 (1966).
5. K. Skarp, S. T. Lagerwall and B. Stebler, *Mol. Cryst. Liq. Cryst.*, **60**, 215 (1980).