*Mol. Cryst. Liq. Cryst.*, 1990, Vol. 179, pp. 189–200
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Printed in the United States of America

# UNDAMPED NONLINEAR OSCILLATIONS OF THE DIRECTOR OF A NEMATIC LIQUID CRYSTAL INDUCED BY AN ELLIPTICALLY POLARIZED LASER BEAM

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Abstract Undamped nonlinear oscillations of the molecular director in a homeotropically aligned nematic liquid crystal have been observed in the field of an elliptically polarized laser beam at normal incidence. This new nonlinear optical phenomenon has been studied with a sensitive heterodyne interferometer/polarimeter. A physical explanation of the results based on the total angular momentum conservation and on Self-Induced Stimulated Light Scattering is also presented.

## EXPERIMENT

When a linearly polarized optical wave is sent onto a homeotropically aligned Nematic Liquid Crystal (NLC) film at normal incidence, a steady-state reorientation of the molecular director is induced, provided the light intensity is above the threshold for the Optical Fréedericksz Transition (OFT)[1].

A completely different phenomenon is observed if an elliptically polarized wave is sent onto the sample: the characteristic far-field diffraction pattern due to self-

phase modulation becomes unstable and the number of the aberrational rings varies periodically in time. This effect is to be ascribed to the onset of nonlinear oscillations of the molecular director in the sample under stationary illumination conditions. To our knowledge, this effect was not reported before.

In this work, we present preliminary quantitative measurements on this phenomenon, obtained with a heterodyne ellipsometric technique. The experimental setup is shown in Fig.1. A focused argon laser beam was used to reorient a homeotropically aligned NLC film (5CB, 75μm thickness) and a He-Ne circularly polarized beam was used to monitor the sample birefringence.

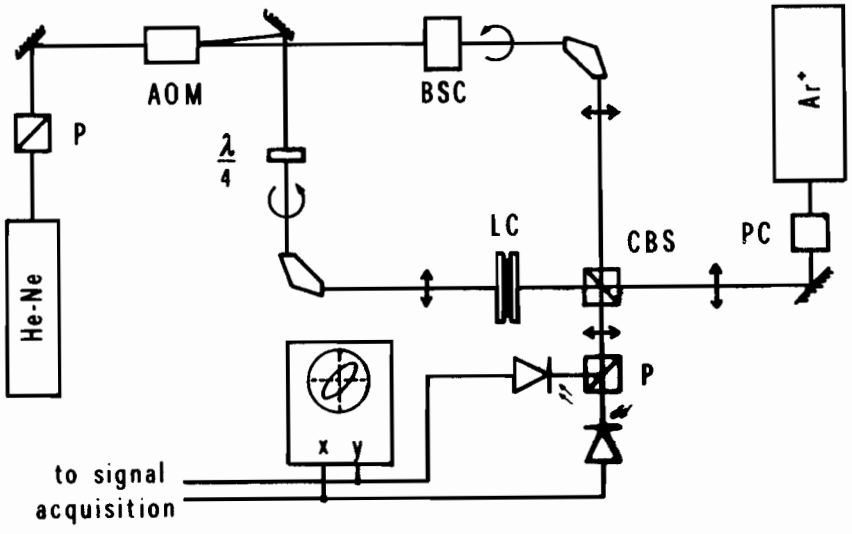


FIGURE 1. Experimental set-up [AOM, Acousto-Optic Modulator; BSC, Babinet-Soleil Compensator; CBS, Cube Beam Splitter; LC, Liquid Crystal film; P, Polarizer; PC, Pockel Cell].

The polarization state of the probe beam emerging from the sample was recorded with use of a heterodyne polarimeter scheme[2]. As shown in Fig1, a fraction of the linearly polarized He-Ne beam was split off and frequency shifted by an acousto-optic modulator driven at 40 MHz and sent into the reference arm of a Mach-Zehnder-like interferometer. A Babinet-Soleil compensator was inserted in the reference arm for calibration. A $\lambda/4$ plate was inserted in the other (signal) arm of the interferometer to obtain circular polarization and then focused onto the liquid crystal film. The signal and reference beams was recombined by a cube beam splitter. As shown in the Figure, the beam splitter was used also for inserting the pump argon beam. The recombined beams were further divided into a Horizontal (H) and Vertical (V) components of equal intensities by a Glan prism polarizer. In this way, the H and V components of both the signal and reference beams fell separately on two photodiodes and generated an heterodyne beat signal at 40 MHz from each of them. The two beat signals from the photodiodes were then sent to the x and y terminals of a sampling oscilloscope for visual display of the output polarization ellipse, and/or to an IBM-PC computer for data processing.

The He-Ne probe beam was circularly polarized and tightly focused on the sample to a spot much smaller than the argon pump beam so that the liquid crystal reorientation could be assumed almost uniform across the probe cross-section. The proper dimensions of the probe and pump beam at the sample was checked visually by a microscope. The circular polarization was checked by the null-reflection method.

Circular polarization was chosen for the probe beam so that, assuming negligible twist in the sample, the major axis of the polarization ellipse, as seen on the oscilloscope screen, yielded a measure of the instantaneous projection of the optical axis of the sample on the plane xy, parallel to the sample walls and the ellipticity of the polarization ellipses yielded a measure of the overall birefringence of the sample. In fact, when circularly polarized light is sent onto a uniaxial birefringent film, having the local optical axis contained in a plane $\pi$, say, the output polarization ellipses is oriented with its major axis at an angle of 45° with respect to the plane $\pi$. Then, if the plane $\pi$ happens to rotate, the output polarization ellipse rotates accordingly. Moreover, the ellipse ellipticity e (defined as the ratio between the minor and major axis of the polarization ellipse) is simply related to the phase difference $\Delta\Psi$ between the ordinary and extraordinary waves, accumulated in traversing the sample:

$$e = (1 - \cos\Delta\Psi)/\sin\Delta\Psi$$

$$\Delta\Psi = (2\pi/\lambda)\int_0^d [n_o n_e/(n_e^2\cos^2\theta + n_o^2\sin^2\theta)^{\frac{1}{2}} - n_o]dz$$

where $n_o$ nd $n_e$ are the ordinary and extraordinary indices of the material, respectively, $\lambda$ is the vacuum wavelength, d is the sample thickness and $\theta$ is the polar angle between the optical axis and the z-axis. We notice that the ellipticity e depends only on the distribution of the polar angle $\theta$ in the liquid crystal film. Since both the ellipticity and the orientation of the polarization ellipse are directly measured by our apparatus, we were able to obtain information on the rotation of the optical axis and on the polar angle $\theta$, separately. The sensitivity of our apparatus in measuring the phase difference $\Delta\Psi$ was of the order of

0.1°. This is to be compared with the sensitivity of $\cong 2\pi$ rad in usual observation based on the count of the diffraction rings produced in the far-field beyond the sample.

Below the threshold for the optical Fréedericksz transition, the polarization of the probe beam remained circular after having traversed the sample. The threshold was found by increasing the pump laser intensity until the probe polarization beyond the sample was seen to become elliptical. Due to the pre-transitional time slow-down, more than 10 minutes was waited after each change of the laser intensity to be sure that the steady state was really reached. The long-term stability of the argon laser light intensity was also checked to be less than .05% during the whole experiment.

In the case of circular polarization of the argon pump beam and for intensities not too far from the threshold, we observed only a rigid rotation of the output polarization ellipse of the probe beam, indicating a uniform rotation of the molecular director of the NLC, as already reported [3].

For elliptical polarization of the pump beam, a much more complicated dynamics was observed.

As a first step we measured the threshold intensities $I_{th}$ for the OFT as a function of the ellipticity $s_3(0)$ of the pump argon beam. Due to the very high sensitivity of the ellipsometer, two different thresholds are usually found for each fixed pump ellipticity. Below the lowest threshold no orientation occurs and above the highest threshold oscillations of the molecular director is always observed. For intermediate intensity, the system finds a stationary equilibrium final state. The difference between the lowest and the highest threshold becomes larger as the

pump polarization tends to be linear and vanishes for pump ellipticities greater than .85. This corresponds to the experimental fact that for a pump polarization close enough to the linear one a final state of equilibrium is always attained, while for a pump polarization very near to the circular one no equilibrium is possible and the NLC molecules start to continuously rotate around the beam propagation direction. It is worth to mention the following result between the threshold measurements: although previous theoretical calculations for an incident plane wave indicate that the threshold intensity for the circularly polarized light is exactly twice the one expected for the linearly polarized light[3], we definitely obtain a ratio of 1.4 (±.2). We have no explanation for this, but we strongly suspect that the lowering of the observed circular-to-linear threshold ratio may be due to the finite cross-section of the focused pump beam.

Above the threshold for oscillations, a very complicated dymamics is observed, depending on the pump intensity and polarization ellipticity. For intensity not too far from the threshold, we can distinguish three principal regimes:

a) pump ellipticity close to linear:
after a transient ranging from 2 to 5 sec., where the molecular director moves in the plane formed by the beam propagation direction and the major axis of the pump ellipticity (initial reorientation stage), the molecular director starts to oscillate rotating back and forth of the initial plane. The period of this oscillatory motion is of the order of 50-100 sec., much longer than the initial reorientation time. As long as we approach the linear

polarization the oscillation period becomes longer and longer until, for linear polarization, the molecular director stays in the beam polarization plane. An example of this regime of motion is reported in Fig.2.

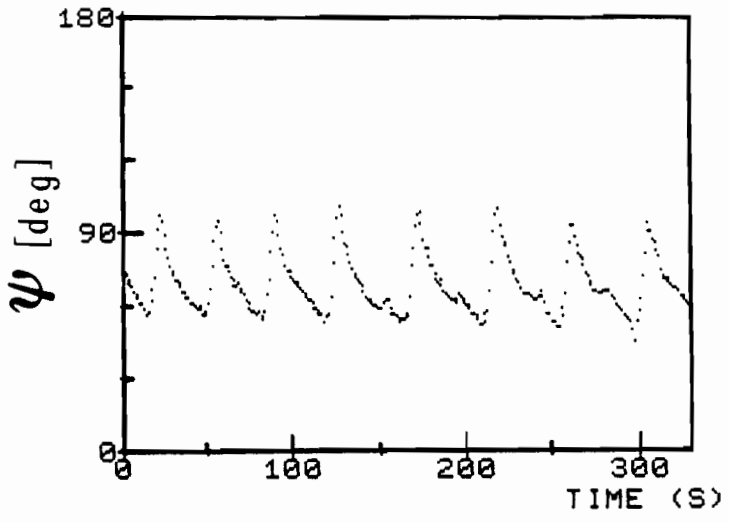


FIGURE 2. Plot of the angle $\psi$ (the ellipse major axis orientation) of the probe beam vs. time. The pump power is P=109 mW and ellipticity $s_3(0)$=.40.

b) pump ellipticity close to circular:

after a transient ranging from 2 to 5 sec., where the molecular director moves in the plane formed by the beam propagation direction and the major axis of the pump ellipticity (initial reorientation stage), the molecular director starts to continuously rotate around the beam propagation direction. The rotation angular velocity is not uniform, in general, and also the phase retardation between the ordinary and extraordinary waves changes periodically in time. This suggests that the rotation of the director is

accompained by nutation.

The polar angle $\vartheta$, which the molecular director forms with

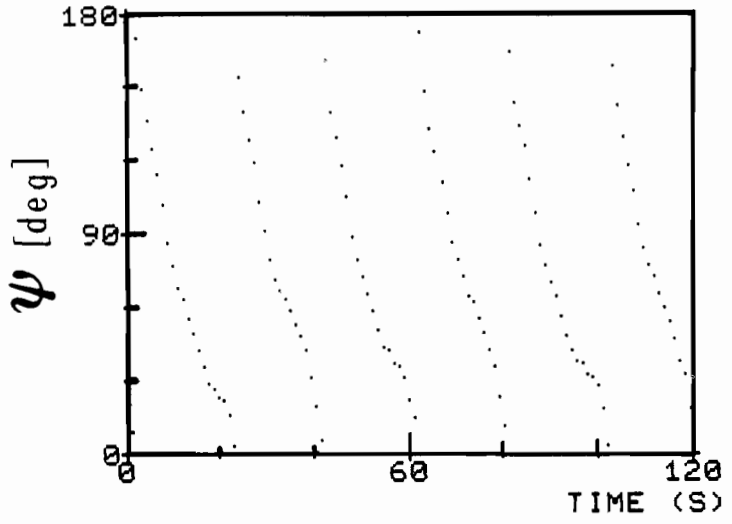


FIGURE 3. Plot of the angle $\psi$ of the probe beam vs. time. The pump power is P=116 mW and ellipticity $s_3(0)$=.95

the beam propagation direction, oscillates but never reaches the value zero.

The period of both rotation and nutation is in the range of 50-100 sec., much longer that the transient initial orientation time. As long as we approach the circular polarization, the nutation becomes negligible and the rotation becomes more uniform, reproducing the results already reported in Ref.2. The rotation is reversed if the elicity of the pump beam is reversed, accordingly. An example of this regime is reported in Fig.3.

c) <u>intermediate pump ellipticity:</u>

after a transient similar to the previuos ones, the molecular director starts to rotate around the beam propagation

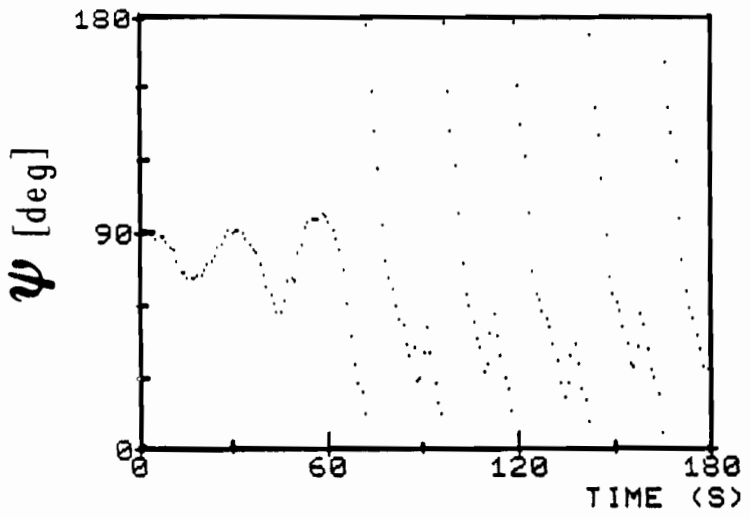


FIGURE 4. Plot of the angle $\psi$ of the probe beam vs. time. The pump power is P=111 mW and ellipticity $s_3(0)=.81$

direction and, simultaneously, the director polar angle $\vartheta$ diminuishes. After some time (of the order of 50-100 sec.), the angle $\vartheta$ becomes zero and the whole process (including the initial transient stage) is repeated. An example of this intermediate regime is reported in fig.4.

We have also investigated the case of pump intensity very highly above the oscillation threshold. Much more complicated motions have been observed, in this case, resembling deterministic cahos. The study of this kind of motions will be reported in a separate work.

## PHYSICAL INTERPRETATION

For an elliptically polarized wave, the electric field $\underline{E}$ rotates at the optical frequency $\omega$ around the beam propaga-

tion direction, that we take as z-axis. The average optical torque on the homeotropically aligned liquid crystal molecules is zero, initially. No molecular reorientation can be induced, therefore, until the intensity of the wave exceeds the threshold for the OFT. Above the threshold, the undistorted state becomes unstable and the molecular director $\underline{n}$ starts to move in the plane formed by the polarization ellipse major axis and the z-axis. Then, because of the birefringence of the medium, besides the ordinary wave, also an extraordinary wave is generated in the medium. Due to the interference between the o- and e-waves, the polarization of the optical beam varies strongly as it propagates in the liquid crystal and, therefore, the optical torque on the NLC is different from point to point in the material. This produces, in general, a spatially complicate reorientation in the NLC, involving twist, splay and bend degrees of freedom.

The polarization of the optical beam varies also in a complicated way throughout the medium and, in general, the light emerging beyond the sample will be elliptically polarized, but with a different ellipticity. Angular momentum is therefore gained or lost by the optical field in the process. This angular momentum must be transferred to the liquid crystal. Because of the homeotropic alignement at the sample walls, no mechanical torque can be exchanged between the liquid crystal and the walls. Then, the torque on the NLC molecules, due to angular momentum transfer from the optical field, cannot be compensated by the surface torque at the walls (that are fixed in the laboratory frame) and the molecules of the liquid crystal are forced to rotate around the z direction.

In the case of circular polarization of the pump beam, a uniform rotation of the liquid crystal director is induced[3]. In this simple case, the process can be understood as a new kind of Self-Induced Stimulated Light Scattering[5], where right-handed circularly polarized photons are transformed into left-handed circularly polarized photons, having a lower frequency. The red-shift of the created photons accounts for the energy dissipation in the medium.

In the case of an elliptically polarized incident wave, the process is much more complicated, since now the wave is a mixture of both right- and left- handed circularly polarized photons having the same frequency. Both components undergo the Self-Induced Stimulated Scattering Process and also one component may be scattered into the other. The frequency shift in the scattering process may be now both toward the Stokes and the antiStokes regions, even if the total energy carried by the photons emerging from the sample must be lower, since, in any case, energy is dissipated in the medium. Because of the frequency difference between the circular components of the wave generated in the medium, the average optical torque on the NLC molecules varies in time and the local director $\underline{n}$ is forced to move continuously, producing the observed motion.

## CONCLUSIONS

We presented the first quantitative experimental study on the onset of spontaneous nonlinear oscillations of the director of a NLC under the action of an elliptically polarized laser beam at normal incidence.

Several dynamical regimes were observed and described,

depending on the polarization ellipticity of the incident beam. In the measurements a very sensistive heterodyne interferometer/polarimeter was used.

Even if we believe that the fundamental mechanism governing the process can be understood in terms of a Self-induced Stimulated Light Scattering process, we have not yet a detailed mathematical model to be compared with the experimental results. A plane-wave model, that can be handled by numerical methods, is actually under study and will be compared soon with the experiment.

ACKNOWLEDGEMENTS

This work was supported by MPI. ES also aknowledges support by NATO Grant n.0463/87.

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