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Dynamical Regimes and Motion Intermittence Induced by a Laser Beam in a Nematic Liquid Crystal Film

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Dynamical Regimes and Motion Intermittence Induced by a Laser Beam in a Nematic Liquid Crystal Film

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When an *s*-polarised laser beam shines on a homeotropically aligned film of nematic liquid crystal at small incidence angle, undamped oscillations of the molecular director may be produced. At high beam intensities, the oscillations break up into deterministic chaos. Although this effect has been known for a long time and the route to chaos has been qualitatively analysed, no detailed study of the director motion has been carried out. We designed an experimental apparatus to monitor the dynamics of the director, as described by its two polar angles. We observed different dynamical regimes, depending on laser intensity: steady states, oscillations, rotations, and, at the highest laser intensities, deterministic chaos. Moreover, the transitions between the oscillation and rotation regimes are characterized by intermittency.

Keywords: optics of liquid crystals; optical reorientation; intermittence; deterministic chaos

INTRODUCTION

It has been known since a long time that liquid crystals can be reoriented by laser optical fields [1, 2, 3]. It was soon realized, however, that the laser induced reorientation in liquid crystals differs from the molecular reorientation induced by external electric or magnetic static fields, because the light beam can transfer a part of its intrinsic angular momen-

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tum to the medium because of the Self-Induced Stimulated Light Scattering (SISLS) [4]. When the polarization ellipticity of the incoming beam changes in traversing the medium, a net torque is produced inducing a rotation of the liquid crystal molecular director around the beam propagation direction. The sign of this torque depends on the phase difference that the ordinary and the extraordinary waves accumulate in traversing the sample. Because the sample thickness L is usually much larger than the optical wavelength λ, the sign of the optical torque along the beam direction may change many times during the reorientation process, leading to an oscillating motion of the molecular director. This happens for example when an elliptically polarized laser beam is sent onto a liquid crystal film at normal incidence. Depending on the beam intensity and polarization ellipticity, steady-states, oscillations, or rotations of the molecular direction are induced [5, 6, 7]. A linearly polarized laser beam cannot produce SISLS at normal incidence, because the light polarization does not change in passing through the sample: in this case, a steady distorted state is reached, provided that the laser intensity exceeds the threshold for the Optical Fréedericksz Transition (OFT) [8, 9]. A steady state distortion is reached also for oblique incidence angle, when p-polarization is used. A simple inspection shows, indeed, that the beam polarization remains unchanged in this case too. A completely different situation arises if the laser is switched to s-polarization. In this case, in fact, the light polarization becomes elliptical in traversing the sample, yielding SISLS. Oscillating dynamical regimes of the molecular director were indeed observed in such geometry long time ago [8, 10, 11] and the occurrence of deterministic chaotic motion was also reported and studied quantitatively in recent times [12]. It should be noticed, however, that, although SISLS may provide a satisfactory qualitative explanation of the director oscillation phenomenon, no quantitative model has been worked out hitherto to describe the oscillating regime. Having more detailed experimental data is therefore advisable to acquire a deeper insight in the oscillation process. Unfortunately, no detailed information about the director motion can be

found in the literature, because previous experiments were based on the measurement of physical quantities that cannot be related to the director orientation in a simple way. In Refs. [8, 10, 11] the number of rings and the diameter of the far field diffraction pattern was determined. These quantities are related in a simple way to the mean polar angle θ of the molecular director $\boldsymbol{n}$, but provide no information about its ϕ azimuthal angle*. In Ref. [12] the transmitted intensities $I_{\|}$ along the incident laser polarization direction and $I_{\perp}$ perpendicular to it were measured as functions of time. The light intensities $I_{\|}$ and $I_{\perp}$ were measured at the center of the far field self-diffraction pattern. Although $I_{\|}$ and $I_{\perp}$ may depend on both the director polar angles θ and ϕ, their actual dependence cannot be worked out until a detailed model is available.

THE EXPERIMENTAL APPARATUS

In this work, in order to determine the director dynamics, we singled out two physical quantities related in a simple way to the two polar angles θ and ϕ. We chose to measure the total angular aperture Θ (or, equivalently, the maximum diameter) and the average polarization direction of the far field self-diffraction pattern. The former quantity is, in fact, related to θ only [13], while the latter provides a direct information on ϕ, because only the extraordinary component of the laser beam is scattered into the outer region of the ring pattern and, therefore, it is roughly polarized at an angle ϕ with respect to the polarization of the incident beam.

Our setup is shown in Fig. 1. The sample was a L=50 μm thick E7 commercial nematic film. The sample walls were coated with DMOAP surfactant for homeotropic alignment. Because of the very large temperature range of the nematic phase of E7, no temperature stabilization was necessary. An s-polarized argon laser beam (λ = 514.5 nm) was sent onto

*θ and ϕ are defined with respect to polar axes x and z directed along the incident beam polarization direction and along the normal to the sample walls, respectively

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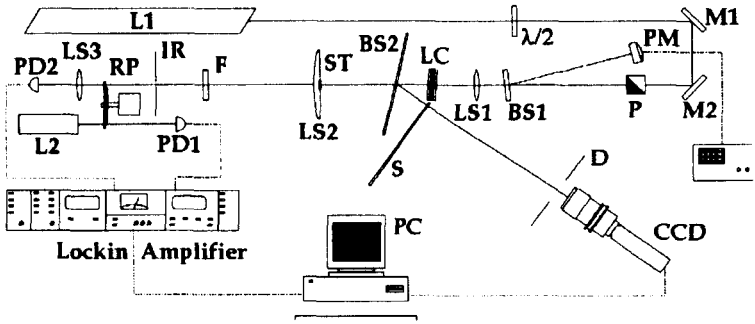

FIGURE 1 The experimental setup. L1 Ar^+ laser; L2 He-Ne laser; M1, M2 mirrors; P polarizer; PM power meter; BS1, BS2 beam splitters; LS1, LS2, LS3 lenses; LC nematic sample; S screen; D diaphragm; ST beam stop; F neutral filter; IR iris diaphragm; RP rotating polarizer; PD1, PD2 photodiodes; PC personal computer.

the film at incidence angle $\alpha = 5°$ to excite a pure extraordinary wave in the undistorted medium. A 150 mm converging lens focused the beam to a Gaussian spot having waist $w = 22$ μm at the sample position. Above the threshold for OFT, the liquid crystal molecules are reoriented and the characteristic circular ring pattern due to self-diffraction is formed in the far field. The ring pattern was monitored by a CCD camera and a suitable PC program extracted in real time its total angular aperture $\Theta(t)$ from the CCD data. In our experimental conditions ($w \ll L$) the angle $\Theta(t)$ is roughly proportional to the square of the maximum polar angle $\theta_m(t)$ that $\boldsymbol{n}$ may assume in the sample [Ref. [13], Eq. (26)]. The rest of the beam was collected by the large aperture lens LS2 onto the photodiode PD, after having passed through the rotating polarizer RP, driven by a 50 Hz synchronous motor. A low power He-Ne laser beam was made to pass through the same rotating polarizer and then sent to the photodiode PD1. The phase difference $\Phi(t)$ of the two 100 Hz signals coming from PD1 and PD2 was measured by a two channel lock-in amplifier. The

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phase difference $\Psi(t)$ is twice the angle $\phi(t)$ between the mean polarization direction of the ring pattern and of the reference laser L2. The two signals $\Theta(t)$ and $\Phi(t)$ were processed simultaneously by our acquisition program. A small stop placed at the center of the large aperture lens L2 prevented the central part of the ring pattern reaching the rotating polarizer. This trick was needed to improve the sensitivity of the polarization measuring chain, because in the central part of the pattern the ordinary and extraordinary waves generated in the medium add together producing unwanted elliptical polarization. The outer part of the self-diffraction ring pattern is almost linearly polarized, instead, because it was produced by the extraordinary wave only. The overall time resolution of our apparatus is $\approx 200\ \mu$s.

THE DYNAMICAL REGIMES

One of the most useful features of our apparatus is that different dynamical regimes of the molecular director $\boldsymbol{n}$ can be observed in detail so that, for instance, rotations of $\boldsymbol{n}$ can be visually distinguished from oscillations. This was impossible to achieve with all experimental techniques reported in literature†. We tested our apparatus at normal incidence where we measured also the threshold I_{th} for the OFT. We found $I_{th} = 6.7 \pm 0.3$ kW/cm^2, in good agreement with the value $I_{th} = 6.4 \pm 0.2$ kW/cm^2 obtained from theory [13, 14]. We checked also that the response time was proportional to the incident power, as predicted by theory. At normal incidence, only steady distorted state were observed, as expected. Passing to the incidence angle α=5° and raising the incident power in very small steps ($<$ 5 mW each) the following sequence of dynamical regimes was suddenly observed: (i) for $I/I_{th} < 1.00$, no distortion is induced in the sample; (ii) for $1.00 < I/I_{th} < 1.41$, the system reaches a distorted steady

†The heterodyne technique exploited in Refs. [5, 6] can distinguish, in principle, rotations from oscillations, but it is no longer reliable when many rings are present in the far field pattern

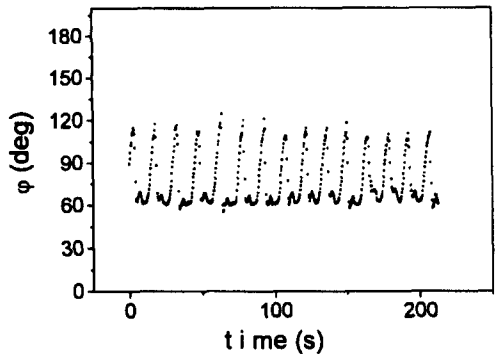

FIGURE 2 Azimuthal angle ϕ of $\boldsymbol{n}$ as a function of time. Oscillations are about the beam polarization directions. $I = 2.14 I_{th}$.

state; (iii) for $1.41 < I/I_{th} < 2.19$, the molecular director oscillates about the laser incidence plane (ϕ=90°); (iv) for $2.19 < I/I_{th} < 2.40$, the molecular director undergoes an intermittent, possibly chaotic, switching between oscillations and rotations; (v) for $2.40 < I/I_{th} < 2.81$, the molecular director rotates around the z-axis perpendicular to the sample walls; (vi) or $2.81 < I/I_{th} < 2.92$, the molecular director shows a second intermittent dynamical regime between rotation and oscillation; (vii) for $2.92 < I/I_{th} < 3.65$, the molecular director oscillates again about the laser incidence plane; (viii) for $I/I_{th} > 3.65$, the oscillations break into apparently chaotic oscillating motion. Examples of these dynamical regimes are reported in figures from Fig. 2 to Fig. 5. Just above the threshold, the molecular director has a stationary initial orientation along the beam s-polarization direction (regime 2), but, as the incident power is increased, the steady-state ϕ angle rotates more and more toward the laser incidence plane until the nonlinear oscillations start (regime 3).

We note that in the oscillation regime 3 (see Fig. 2) no rings were formed in the far field, showing that the angle θ remained frozen to a very small value, in spite of the high laser intensity (more than twice the

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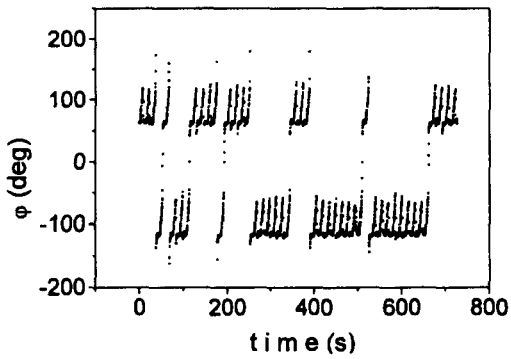

FIGURE 3 Azimuthal angle ϕ of $\boldsymbol{n}$ as a function of time, showing intermittence between oscillations and rotations. The "flipping" phenomenon is evident. $I = 2.19 I_{th}$.

threshold intensity). In the same intensity range Cipparrone et al. [12] – using a different experimental apparatus – claimed to have observed a period doubling and a subharmonic cascade, successively frustrated, that they interpreted as a precursor to chaos. This conclusion seems to be in disagreement with our observations, although a more careful comparison between our and their data would be desirable. We observed, instead, a transition to an intermittent motion, where oscillations around the incidence plane alternate with complete rotations around the z-axis (regime 4). This regime is shown in Fig. 3. The rotation period was found to be about twice the period and this circumstance may have led Cipparrone et al. to interpret it as the onset of a period doubling cascade.

Increasing the laser power, the rotations become unstable and a second irregular intermittent motion sets in (regime 6), alternating rotations with oscillations around the laser incidence plane. During intermittence, the period of the ring diameter pulsation remains almost constant, so that there is no way to detect intermittency just looking at the ring pattern. Further increasing of the incident power stabilizes the oscillations around the incidence plane. The oscillations around the incidence plane are ini-

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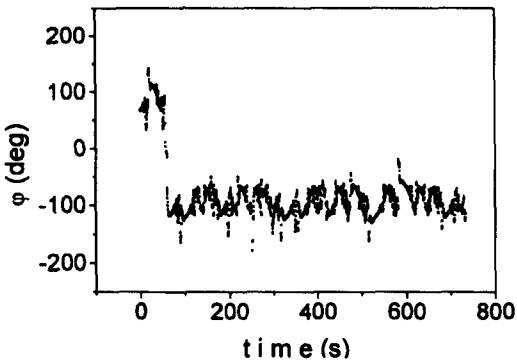

FIGURE 4 Azimuthal angle ϕ of $\boldsymbol{n}$ as a function of time, showing chaotic motion. $I = 3.75 I_{th}$.

tially regular in time, but they become more and more irregular as the incident power is increased, until, for $I > 3.65 I_{th}$ the oscillations around the incidence plane become irregular in time with the onset of deterministic chaos. A similar irregular behavior is observed in the ring diameter, as shown in Figs. 4 and 5.

We have not analyzed the transition to chaos quantitatively as made in Ref. [12], and we cannot be sure of the existence of a quasiperiodic (mode-locked) regime having frequency ratio close to 26/27 as the precursor of chaos. We observed, instead, an apparently direct transition from periodic oscillations to chaos.

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