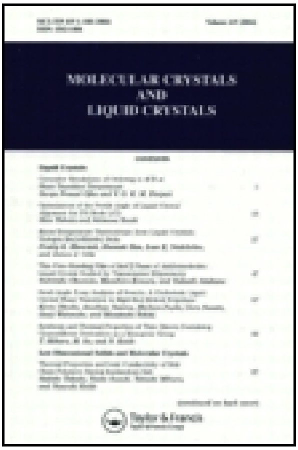

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Analysis of the Dynamical Regimes Induced by a Laser Beam in Nematic Liquid Crystal Films

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We report the analysis of the dynamical regimes induced in a homeotropically aligned nematic liquid crystal layer when a s- polarised laser beam acts at a small incident angle. For increasing light intensities these regimes are studied through an analysis performed on the variation of the polarisation state of a light probe beam. To measure with a good resolution the four Stokes parameters we use an experimental apparatus that include a pump-probe technique and a Four-Detector-Polarimeter (FDP). A preliminary description of the dynamical regimes, based on the analysis of the Fourier spectra, the autocorrelation functions and the attractors of the measured time series has been discussed. Moreover we calculated also the maximum Lyapunov exponent. For the last series of measurements this analysis suggest the presence of a transition towards chaos.

INTRODUCTION

The molecular director reorientation induced by a light field in a nematic liquid crystals (NLC) films has been extensively studied by several authors in different experimental geometry [1-5]. Nevertheless for some of this geometry [3-5] a very interesting and rich dynamics can be observed. The dynamics depends on the unperturbed director orientation, the light polarisation, the light intensity and the incidence angle. In particular, evidences for oscillations of the diffraction rings, observed in the far field pattern of the transmitted beam under certain conditions, indicate the presence of this interesting dynamical behaviour which is only the result of the highly nonlinear coupling between the NLC birefringence and the intense optical field.

Time variations of the reorientation state of the director (rotation around the beam propagation direction, oscillations, etc.) have been observed and studied in the

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case of circular and elliptical polarisation at normal incidence; a very exhaustive study of these dynamical regimes has been reported in [4]. A similar behaviour has been observed using a different experimental geometry, namely in the case of linearly polarised light beam impinging on an homeotropically aligned nematic liquid crystal sample. The time variations are observed when the following conditions are fulfilled: i) the polarisation is perpendicular to the incidence plane and ii) the incidence angle is small but different from zero [3, 5].

In our previous works [5] the phenomenon of self-oscillations of the diffracted ring pattern has been characterised in the framework of the interesting route to a chaotic state, which occurs when the incident intensity is increased [5], in the case of the last experimental condition. In these works the analysis was performed on the time series of the intensities of the two components in the center of the transmitted inducing beam, polarised parallel and perpendicular to the polarisation of the incident beam. The inadequacy of the measured quantities to be related to the local molecular dynamics persuaded us to change the experimental apparatus.

In a recent paper [6] we propose a different experimental method to study the induced dynamics, which allows a most complete study of the phenomenon. This method includes a pump-probe technique and a four detector polarimeter (FDP) that allows us to measure the time evolution of the Stokes parameters of a light probe beam. The director dynamics can be retrieved by the analysis of the probe polarisation state.

The idea to improve our experimental apparatus for a better description of that occur in this system, is due to the belief that these systems and their non-linear interaction with the intense optical field show a rich variety of dynamical regimes.

In the first section we report a short description of the experiment and the measured quantities. In section 2 we show a characterisation of the various observed regimes with a description through the Fourier spectra, the autocorrelation functions, the attractors and the determination of Lyapunov exponents of the measured time series.

EXPERIMENT

The used experimental set up is depicted in fig.1. The experiment and the working of the Four-Detector-Polarimeter (FDP) are extensively described in ref. 6.

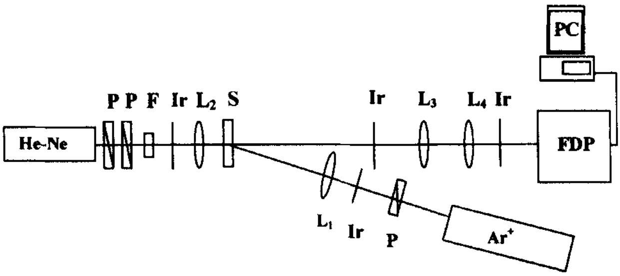

FIGURE 1 Experimental set-up. L_1, L_2, L_3, L_4 lenses; P polarizer; Ir iris; F interference filter; S sample; FDP Four-Detector-Polarimeter.

In our experimental geometry the s-polarised Ar^+ laser beam (λ=514.5nm) is focused (f_1=200mm) on a homeotropically aligned nematic liquid crystal sample (E7 by Merck) with the incidence angle fixed at 5°. The thickness of the sample was 75μm.
The He-Ne laser (λ=632.8nm) probe beam was linearly polarised along the same direction of polarisation of the inducing beam and is sent with normal incidence on the NLC film. In order to investigate a smaller region than those in which molecular reorientation is induced, the probe beam is focused on the sample by a shorter focal length lens (f_2=25mm). The polarisation of the transmitted probe beam is analysed by the FDP, which allows the measurement of the Stokes parameters.
The measurements reported here were performed at fixed incidence angle, using the intensity of the inducing beam as external control parameter. We evaluate the Optical Fréedericksz Transition (OFT) threshold at normal incidence both theoretically and experimentally, and we found an experimental value I_{th} =2.5 ± 0.4 $kWcm^{-2}$, in agreement with the calculated value (I_{th} =2.1 $kWcm^{-2}$). Since the incidence angle is

small, we use the normalised value I/I_{th} in the following. If $I/I_{th} < 1.0$ we didn't observe any variation of the probe beam polarisation state, with respect to the case of undisturbed nematic sample. That is when $I/I_{th} < 1.0$ no director distortion was induced.

The incident laser power is increased with steps of 6 mW (corresponding to 200Wcm^{-2} in intensity). The observation of a sequence of different dynamical regimes in the time evolution of the polarisation state when the beam intensity is increased shows the occurrence in the sample of a very interesting nonlinear dynamics. From an experimental point of view, at a fixed value of I/I_{th} we observe a stationary dynamical regime. When we increase I/I_{th}, we observe either the same kind of behaviour in the time evolution or a different behaviour. When this latter situation is encountered we scan the range of values of I/I_{th} in order to get a precise value of the intensity at the transition towards the new dynamical regime. This transition is generally called *bifurcation*. In this sense our system is very interesting from a dynamical point of view, because it allows us to investigate a sequence of bifurcations in a nonlinear phenomenon. Among all measurements we performed, we show here that corresponding to bifurcations representative of different dynamical regimes. From the acquired time series of the measured Stokes parameters S_0, S_1, S_2, S_3 we calculate [7] the ellipticity e, the azimuthal angle of the major axis Θ and the degree of polarisation P:

$$\Theta = \frac{1}{2}\mathrm{arctg}\left(\frac{S_2}{S_1}\right)$$

$$e = tg\left\{\frac{1}{2}arcsin\left[\frac{S_3}{\left(S_1^2+S_2^2+S_3^2\right)^{\frac{1}{2}}}\right]\right\}$$

$$P = \frac{\left(S_1^2+S_2^2+S_3^2\right)^{\frac{1}{2}}}{S_0}.$$

The degree of polarisation (not reported here) remains always constant (P≅1 within the experimental errors). This means that, for all the dynamical regimes observed at any intensity of the inducing beam, the system don't produce depolarisation of the transmitted light, i.e. no decorrelation is introduced in the components of the electric field of the probe beam.

ANALYSIS OF THE DYNAMICS

As described in [6], the observed dynamical regimes are the same for both e and Θ therefore we choose to report the analysis of the encountered regimes only for the ellipticity e (figs 2-6).

First of all we show the time evolution of $e(t)$ (figs 2a-6a). We report also the usual Fourier Transform (figs 2b-6b) which are commonly used to can get useful information about the dynamics. Then we report the autocorrelation functions $C(\tau) = \frac{1}{T}\int_0^T e(t)e(t+\tau)dt$, where τ is a lag time and T is the duration of the time series (figs 2c-6c). Finally we report a two-dimensional representation of the attractors of the time series e (t) (figs 2d-6d) for most completely information about the dynamics in the phase space.

The attractors are reconstructed using the Takens method [8] that is based on the construction of a vector $\vec{x}(t)$ in the embedding phase space:

$$\vec{x}(t) = [e(t), e(t+\tau), e(t+2\tau), \ldots, e(t+(d_E-1)\tau)]$$

in which τ is an appropriate time delay and d_E is the embedding dimension. Usually τ is the time that corresponds to the first zero of the autocorrelation function.

For I/I_{th} between 1.0 and 1.6, the polarisation state reaches a stable state when the pump beam is switched on, which is different with respect to the case of undisturbed sample. This is a stable stationary state, indicating that the molecular director reaches a distorted steady state. In the phase space a fixed point represents this situation.

After this state a Hopf bifurcation appears. For $1.6<I/I_{th}<1.9$ the variation in time of the polarisation state shows a regular oscillating pattern around negative values of e (fig. 2a). The autocorrelation function shows typical oscillations with a decrease of the amplitude as the lag-time increases, as expected for periodic signals (fig. 2c). The same behaviour can be seen looking at the Fourier spectrum (fig. 2b) in which characteristic frequencies appear ($f_0 \cong 26$mHz). This is a very well defined periodic regime and the attractor of the time series is a limit cycle (fig. 2d).

For $1.9<I/I_{th}<2$, we observe a regime characterised by time intervals during which the oscillations are centred on both positive and negative values of e (fig. 3a).

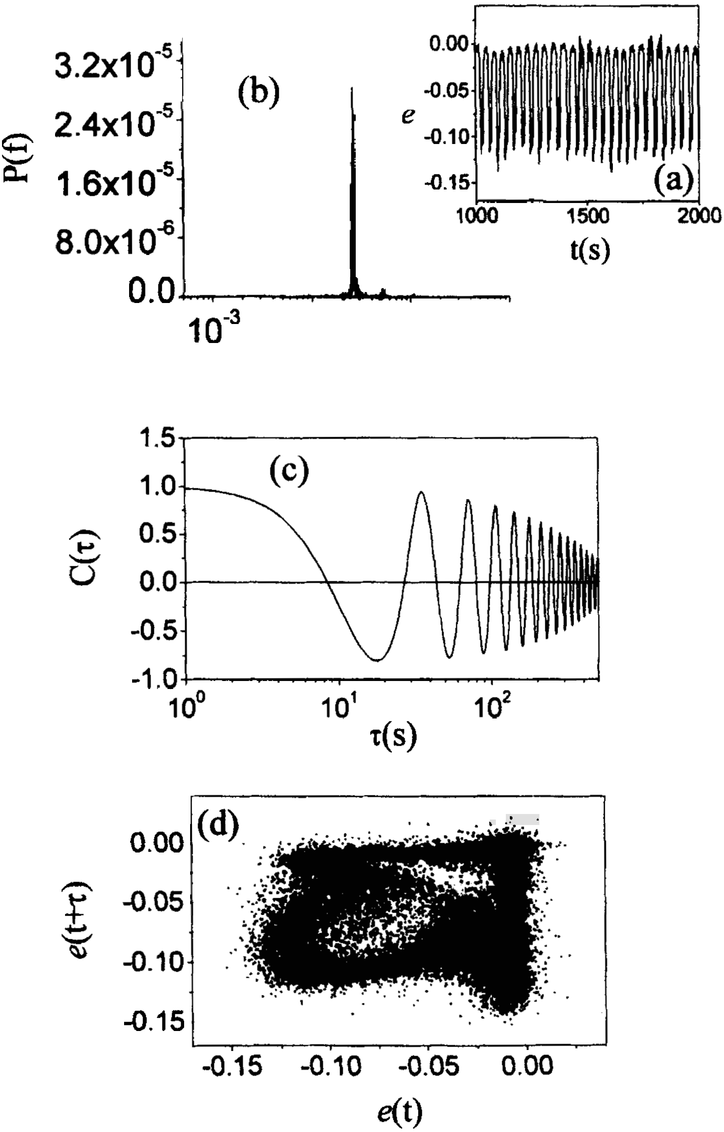

FIGURE 2 **I/I_{th}=1.8; (a) Ellipticity** *e* versus time; (b) Fourier spectrum; (c) Autocorrelation function; (d) Attractor.

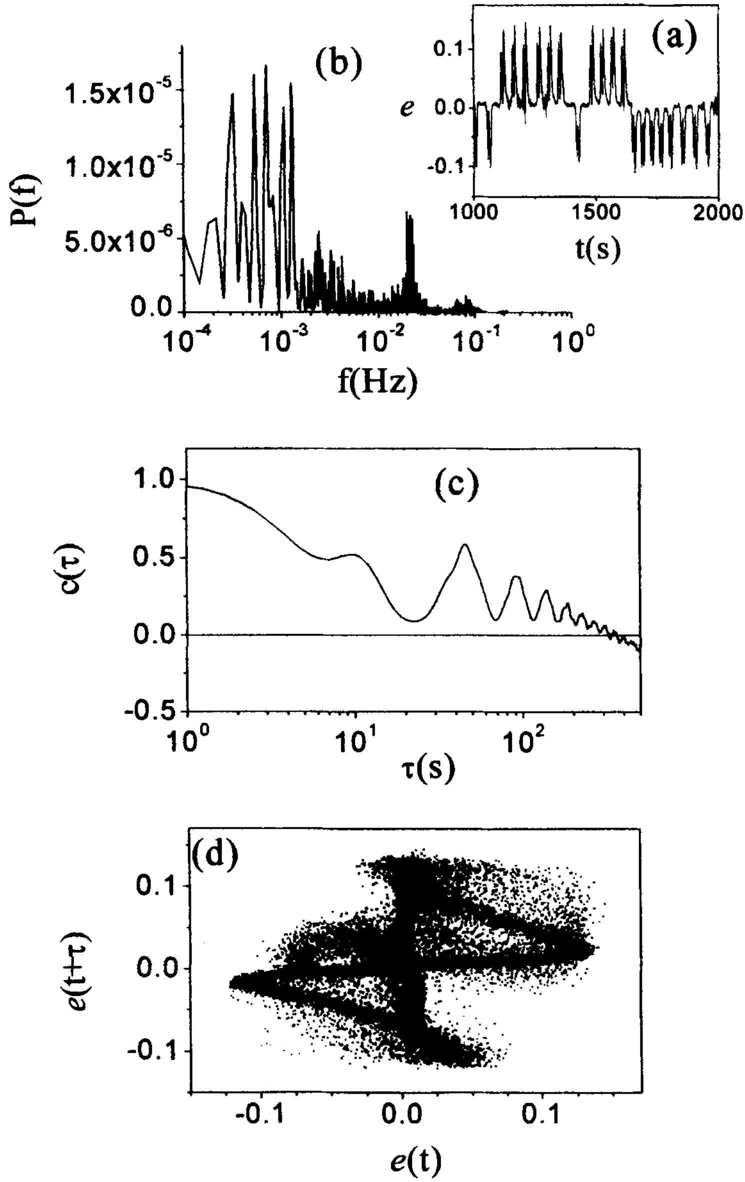

FIGURE 3 The same of Figure2 for I/I_{th}=1.9

During this regime it is possible to observe two equilibrium states characterised by a left elliptical polarisation and a right elliptical polarisation. These states are unstable in the sense that the system oscillates between both states. This is of course a new periodic regime, but looking at the autocorrelation function (fig. 3c) and at the Fourier spectrum (fig. 3b) can better evidence its peculiar characteristics. In fact $C(\tau)$ is positive defined for very long times τ, thus indicating a kind of persistence in the system. This strange behaviour should be due to the fact that there are time intervals where oscillations are positive or negative in a persistent way for long time. For this intervals the signal remains similar to itself and this should be the origin of the persistence (or memory in broad sense) we observe. As regard the Fourier spectrum we can see that a broadening of the main frequency and the birth of a lot of further lower frequencies characterise this regime. Within this lower frequency band we cannot individuate any sub-harmonics of $f_0 \cong 22$mHz. The attractor of the time series is made by two regular regions (fig. 3d) in which the phase trajectories are confined for a certain period; this period corresponds to the duration of the interval in which oscillations are positive or negative.

In this regime the time intervals during which the system reaches a single polarisation state tend to decrease as I/I_{th} increases. This behaviour suggests that we are observing a transition towards a new regime characterised by regular oscillations between the two polarisation states above mentioned, as shown in fig.4a. This new regime is observed in the range $2<I/I_{th}<2.6$. Perhaps this transition is due to a different kind of bifurcation. Looking at the autocorrelation function (fig. 4c) and at the Fourier spectrum (fig. 4b), we can recognise that the transition represents a kind of "self-organisation" of the signals. In fact the measured quantity is periodic for $I/I_{th}=1.8$, looks to be "quasi-stochastic" for $I/I_{th}=1.9$ but is again periodic for $I/I_{th}=2.3$. All frequencies, which are visible in fig. 3b, are now completely suppressed, apart for a single frequency $f_0 \cong 18$mHz. The corresponding autocorrelation function looks to be similar to those reported in fig. 2c, showing that there is a loosing of the persistence observed in the previous regime. In the phase space the two regions are now merged forming a single limit cycle (fig. 4d) more complex and wider than this shown in fig. 2d.

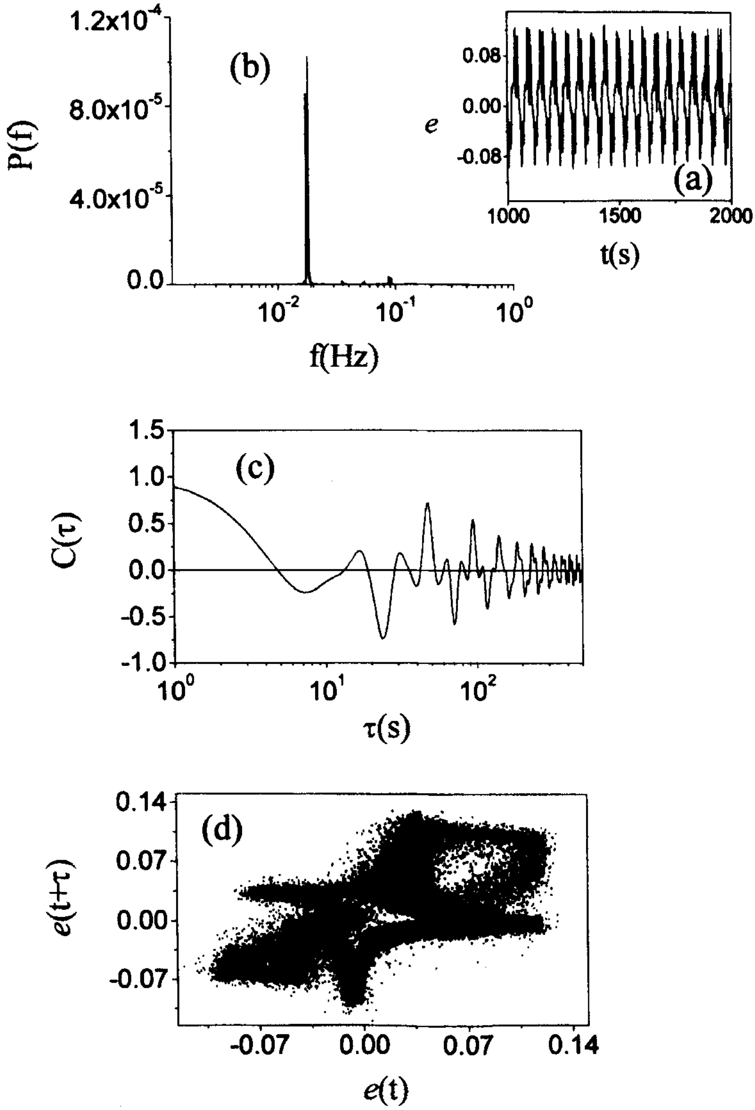

FIGURE 4 The same of Figure2 for I/I_{th}=2.3

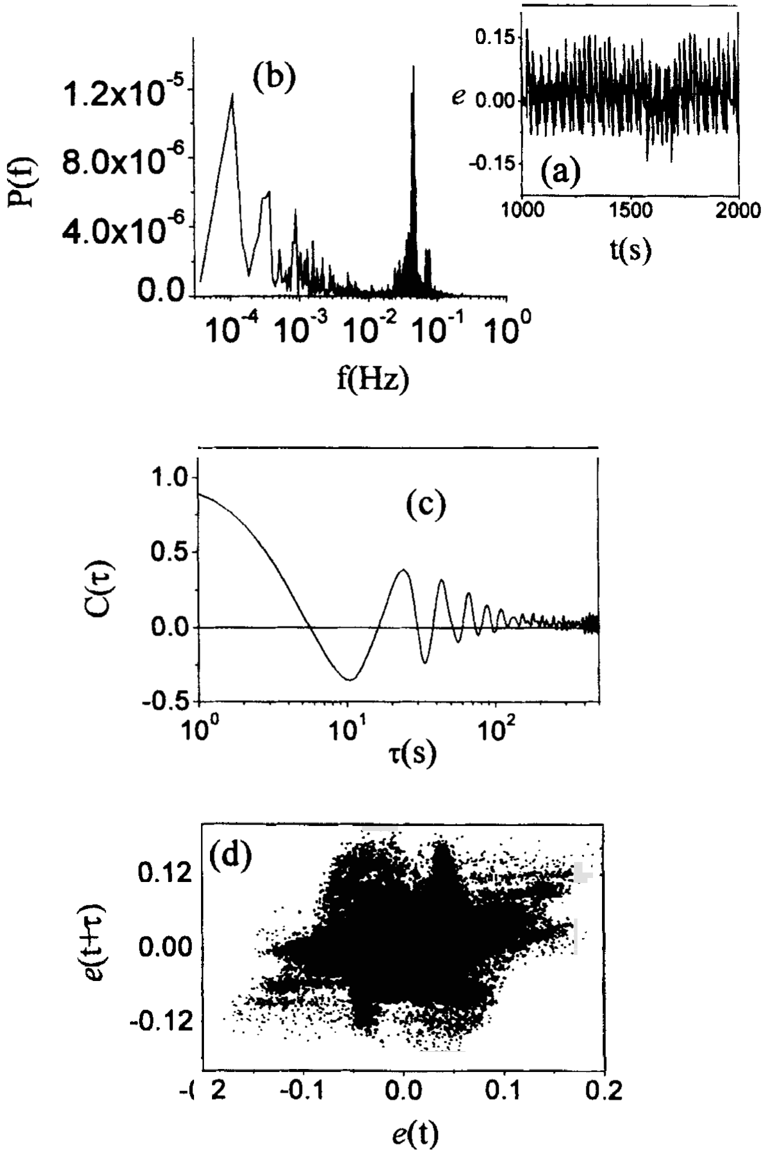

FIGURE 5 The same of Figure2 for I/I_{th}=2.8

This regular regime (figs 4) can be observed up to the value I/I_{th} = 2.6. In this range of frequencies we observe a decreasing of the period of the oscillations and the birth of some harmonics as I/I_{th} is increased up to I/I_{th} =2.6.

For $2.6<I/I_{th}<3.0$ we observe a new irregular sequence of time oscillations, as shown in fig. 5a. Even in this case the Fourier spectrum (fig. 5b) shows broadening of the main frequency and a band of lower frequencies similar to that observed in fig. 3b. The main difference with respect to the regime shown in figs 3 is the fact that in this case the autocorrelation function decreases to zero in few tens of seconds. In fact if increase the intensity, we do not observe the occurrence of a new regular regime, but the appearance of a very stochastic behaviour of *e* (fig. 6a). Even the attractor shows a gradual disruption of the regular structures observed in the previous regimes covering a most large area in the phase space (fig. 5d).

As the stochastic behaviour appears, the autocorrelation function decreases rapidly to zero (fig. 6c) and the Fourier spectrum shows a decrease of the lower frequencies previous observed and the birth of higher frequencies thus generating a continuous spectrum (fig. 6b). For this regime a disordered set of point constitutes the attractor (fig. 6d).

The reported measurements show the evidence of a very rich dynamical behaviour that require a more detailed investigation, mainly as concern the physical description of the sequence of bifurcations. The results of the preliminary analysis, that is the Fourier transforms, the autocorrelation function and the reconstruction of the phase space, suggest the presence of a chaos transition in the measured quantities, as the control parameter increases.

In order to investigate the nature of the observed regimes we calculated the Lyapunov exponent λ of the time series *e (t)* using the Wolf method [9]. This method allows calculating the average rate of the divergence of the trajectories on the attractor.

For regular regimes ($1.6<I/I_{th}<3.0$) the Lyapunov exponent seems to converge towards the value $\lambda \approx 10^{-5} s^{-1}$, close to zero as we expect for periodic signals, whereas for irregular regimes ($I/I_{th}>3.0$) we found a convergence towards $\lambda \approx 10^{-3} s^{-1}$. The last value of λ not suffices to establish whether deterministic chaos or a stochastic random phenomenon drives the dynamic of the polarisation state.

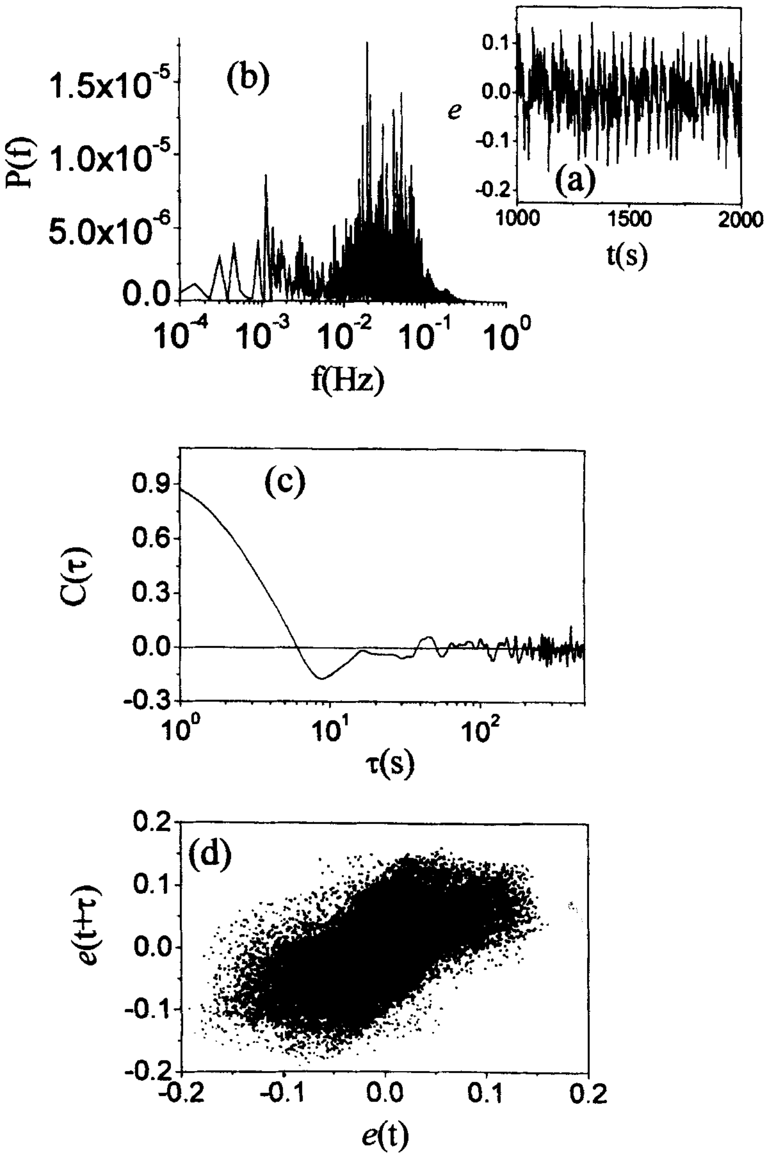

FIGURE 6 The same of Figure2 for I/I_{th}=4.2

In fact two fundamentals problems, depending on the experimental conditions, occurs in the determination of the Lyapunov exponent. They can be resumed in the two following points:

- experimental noise that could mask the real nature of the attractor;
- length of the time series not enough to realise a correct analysis.

The last sentence means that a limited time series with a slow dynamics does not provides a sufficient number of trajectories on the attractors for a correct estimate of the Lyapunov exponent. Unfortunately experimental limits not allow us a longer data set. In fact when the irregular regimes occurs, we observed that the molecular anchoring is broken after few hours. This phenomenon could be attributed to the kind of dynamics induced in the system. In fact, for the same intensity values, in an experimental geometry in which a distortion stable stationary state of the molecular director is observed (the same geometry at normal incidence), the molecular anchoring persists for a period longer than our experimental time series.

CONCLUSION

In conclusion we report the analysis of the dynamical regimes induced in a NLC film as the intensity of a laser beam increases. We use a new experimental set-up based on a pump-probe technique and a Four-Detector-Polarimeter. This apparatus allows us to get information with a very good temporal end spatial resolution on the variation in the polarisation state of the probe laser beam by measuring the time evolution of the Stokes parameters. Measurements show a series of very interesting dynamical regimes as the pump intensity increases. The sequence of bifurcations we observe is very interesting and deserves further studies. The analysis of the observed time series at different intensities suggests the presence of a transition towards a chaotic regime in the system.

Unfortunately an estimate of the maximum Lyapunov exponent does not allows us to confirm whether or not deterministic chaos is present. An efficient investigation of the irregular regimes is prevented by the experimental conditions which do not allows us to achieve sufficiently long time series at high pump intensity.

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