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EXPERIMENTAL INVESTIGATION OF LIGHT INDUCED CHAOTIC DYNAMICS IN NEMATIC LIQUID CRYSTALS

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Abstract We report the characterization of the chaotic behavior in the phenomenon of molecular reorientation induced in nematic liquid crystals by a laser beam in a particular geometry. The conventional approach to the reconstruction of dynamic properties of the measured chaotic signal is used to find the fractal dimension of the attractor and the largest Lyapunov exponent of the system. The intermittency observed in the chaotic regime gives useful informations to develop a satisfactory theoretical framework for the interaction between the light beam and the nonlinear medium.

INTRODUCTION

In recent years, many papers have appeared reporting detailed experimental studies of quasiperiodicity and transition to chaos in different physical systems¹⁻⁵, and a particular experimental interest has been devoted to low - dimensional fluid systems rendered dissipative by viscosity^{6,7}. Nematic liquid crystals (NLC) are ordered fluids in which viscosity plays an important role not only where shear and compression are concerned, but also in the orientational motion of the liquid crystal director $\mathbf{n}(\mathbf{r})$. The NLC responds to external

perturbations giving rise to several collective phenomena that often result in a local distortion of the vectorial field $\mathbf{n}(\mathbf{r})$. Experimental studies on its nonlinear dynamics have been also carried out^{8,9}. The director reorientation can be induced by a static electric or magnetic field^{10,11} as well as by an optical field (LIFT: "Light Induced Freedericksz Transition"¹²); furthermore, the interaction with an intense optical field can exhibit different features depending on the experimental geometries, e.g. the unperturbed director orientation (homeotropic, planar or hybrid), the polarization of the impinging light and the incidence angle. Since the birefringence of the NLC depends on the director orientation, the light modifies the refractive indexes of the medium when reorienting the director. This results in self - phase modulation (SPM) effect¹³ and in changes of the spatial pattern of the transmitted beam. Thus, a NLC interacting with a laser beam represents a dissipative nonlinear system which could exhibit interesting features where the nonlinear dynamics, and eventually the transition to chaos, are concerned. Light, on the other hand, represents a helpful tool to investigate the dynamics of the liquid crystal director since the intensity and polarization of the transmitted beam pattern are determined by the director field¹⁴.

Recently, we have devoted our attention to the fact that, in certain geometries, the SPM results in several oscillating rings in the pattern of the transmitted beam. At present the above mentioned oscillating behavior has been observed in two experimental geometries:

- 1- A circularly or elliptically polarized light beam acts on a homeotropically aligned NLC film at normal incidence¹⁵⁻¹⁷;
- 2- A linearly polarized light beam impinges on a homeotropically aligned NLC film at a small incidence angle and the light polarization is perpendicular to the incidence plane¹⁸⁻²⁰.

The first experimental geometry has been exhaustively studied by Santamato and coworkers¹⁵⁻¹⁷. Time dependent variations of polarization reflecting the orientational dynamic of the director have been observed in the beam outgoing from the sample, for both elliptical and

circular polarizations of the light, when the intensity of the impinging intensity exceeds a threshold value. These effects are interpreted in terms of an angular momentum exchange between the optical and the director fields. For elliptical as well as circular polarization the motion of $\mathbf{n}(\mathbf{r})$ results in a limit cycle in the phase space. Since the angular velocity of $\mathbf{n}(\mathbf{r})$ is a function of the impinging intensity, several limit cycles are possible, but no experimental evidence of an instability of these limit cycles has been reported. The existence of a limit cycle has been also reported for the second experimental geometry²⁰. In this case the self oscillations are strictly periodic only in a well-defined range of laser intensity and incidence angle values. Outside this range, fluctuations become stochastic. The authors investigated experimentally the regime of the periodic oscillations while nothing was reported about the stochastic regime, except the observation of "irregular self oscillations" of the transmitted pattern. For the same geometry (oblique incidence of a linearly polarized ordinary beam), in a previous paper²¹ we reported the observation of various dynamic regimes occurring in the system when the limit cycle becomes unstable.

In this paper we report the characterization of the chaotic regime, showing that the conventional approach to the reconstruction of dynamic properties of the signal and investigations of the peculiar features of the chaotic regime give useful informations for a better understanding of the interaction mechanism of light with a nematic liquid crystal.

EXPERIMENTAL SETUP

When the light intensity exceeds the threshold value, the molecular director reorients out of the incidence plane, so that the medium appears birefringent to the incident light, whose polarization becomes elliptical while passing through the sample. Therefore, the study was

performed by detecting, in the center of the outgoing beam, the intensities $I_{\perp}(t)$ and $I_{//}(t)$ of the two components polarized respectively perpendicular and parallel to the polarization of the incident beam. The experimental setup is shown in Figure 1: The light beam (TEM_{00}) from an Ar^{+} laser ($\lambda = 5145 \text{ \AA}$) is linearly polarized perpendicularly to the incidence plane (ordinary wave) and focused on the sample (150 mm focal length); the incidence angle is fixed at the value $\Phi = 7^{\circ}$. The NLC sample is an eutectic mixture of cyanobiphenyl compounds (E7 by the British Drug Houses) and the sample cell, 50 μm thick, is kept at the temperature of 18 $^{\circ}\text{C}$ by a thermostatic bath.

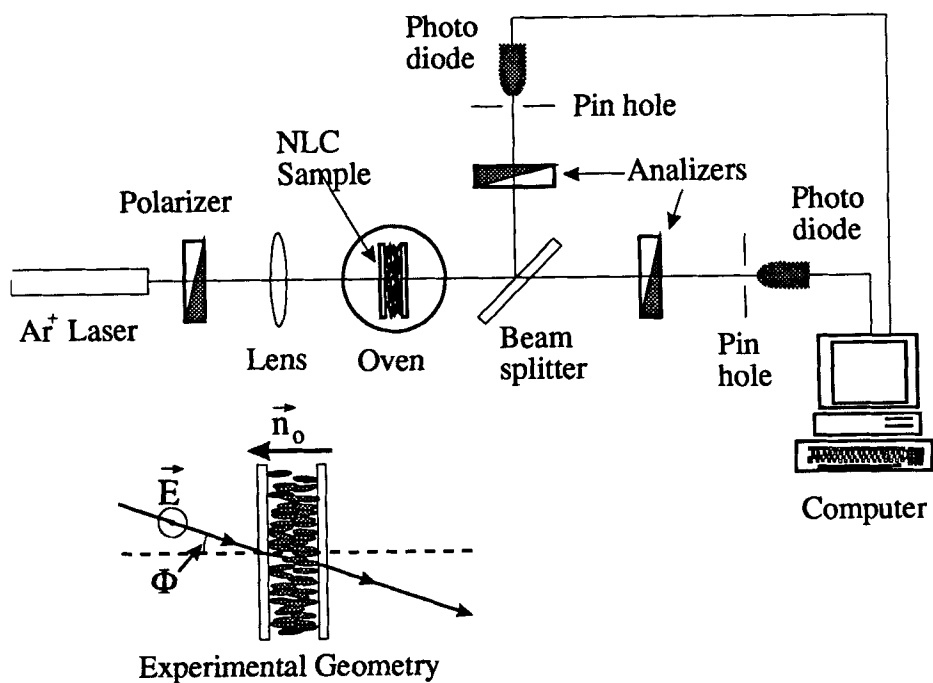


FIGURE 1 Sketch of the experimental setup.

A 200 μm pinhole in the center of the outgoing beam, a beamsplitter, two crossed polarizers and two photodiodes allow us to detect $I_{\perp}(t)$ and $I_{//}(t)$. Signals are sent to a personal computer for the AD conversion and storage. Time series consist of $N = 16384$ points, sampled at the fixed time interval $\Delta t = 0.1$ s.

ANALYSIS OF SIGNALS

We have detected $I_{\perp}(t)$ and $I_{//}(t)$ for three different values of the impinging power P_{inc} , drawing Fourier spectra $S(\omega)$ and projections in the phase space $(I, dI/dt)$. At low impinging power ($P_{\text{inc}} \approx 420$ mW), the spectra $S_{\perp}(\omega)$ and $S_{//}(\omega)$ show a single frequency periodic pulsation which corresponds to a limit cycle in the phase space (Figure 2). At higher impinging power ($P_{\text{inc}} \approx 540$ mW), the spectra exhibit more frequencies, and the limit cycles in the phase space become unstable (Figure 3). At the highest impinging power ($P_{\text{inc}} \approx 700$ mW), the peaks broaden and the gaps between them become shallower. In the phase space we observe a chaotic motion on a strange attractor (Figure 4).

ANALYSIS OF THE CHAOTIC REGIME

At this stage, three questions arise naturally: First of all, can we describe our phenomenon by a low dimensional system of differential equations? The second question is whether neighboring orbits in the phase space exponentially diverge with time, thus characterizing a "deterministic chaos". Finally, can we get from the experiment useful information to develop a satisfactory theoretical model which describes the phenomenon? To answer these questions, we must first of all do the two main steps for the reconstruction of the dynamic properties of an experimentally observed chaotic signal. The first step consists in finding the fractal dimension V of the attractor, which gives an indication of the "complexity" of the physical system.

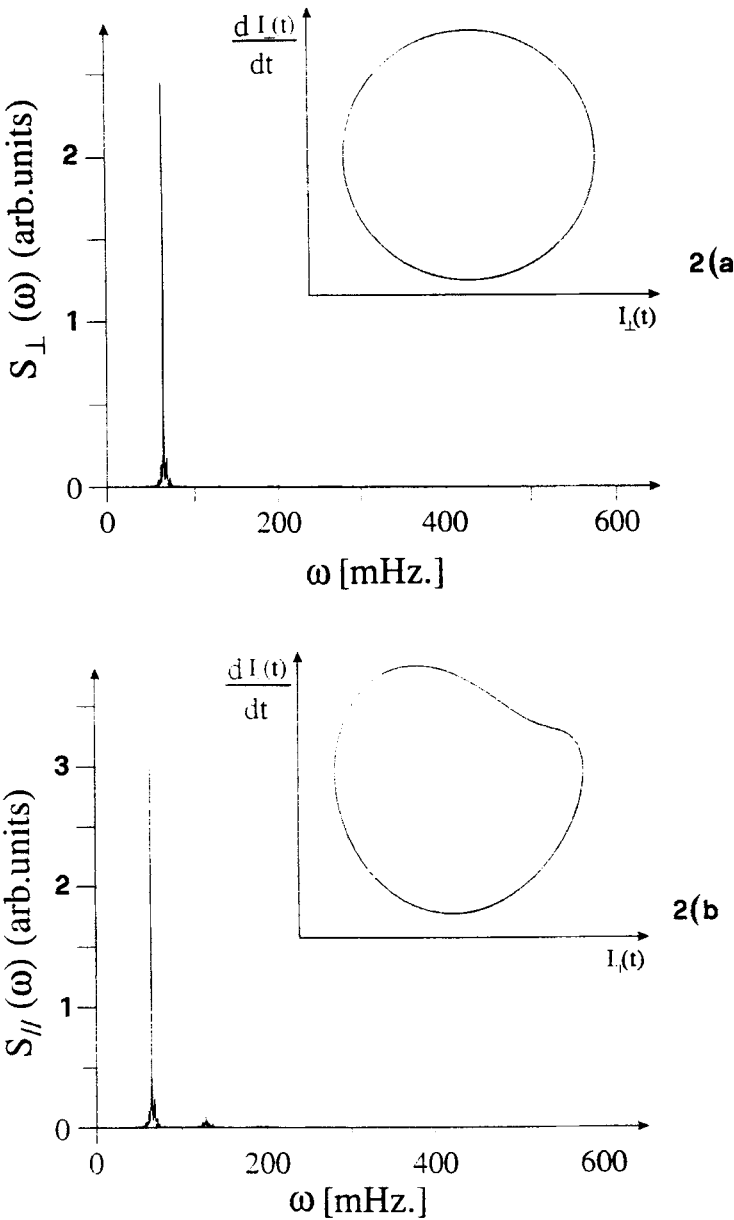


FIGURE 2 Fourier spectra $S_{\perp}(\omega)$ (2a) and $S_{\parallel}(\omega)$ (2b) along with projections in the phase space at impinging power $P_{inc} \approx 420$ mW.

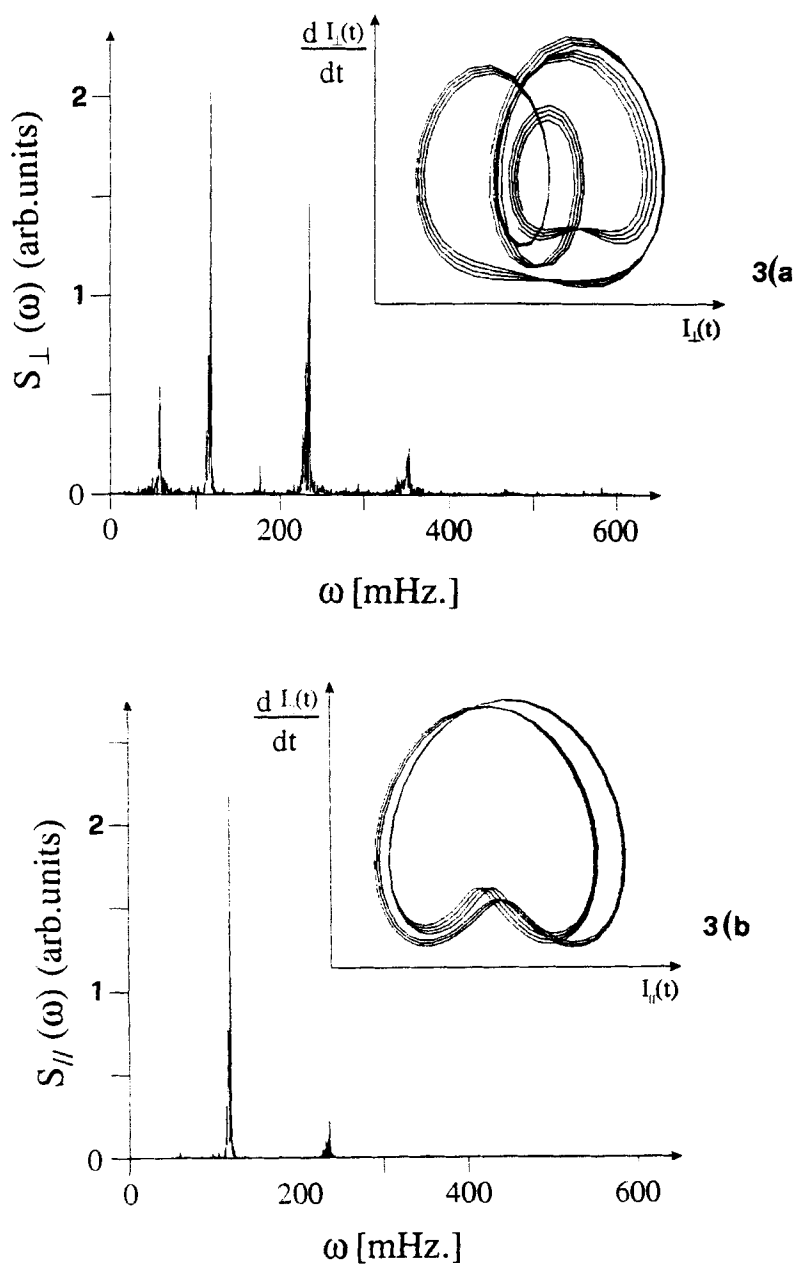


FIGURE 3 Fourier spectra $S_{\perp}(\omega)$ (3a) and $S_{\parallel}(\omega)$ (3b) along with projections in the phase space at impinging power $P_{inc} \approx 540$ mW.

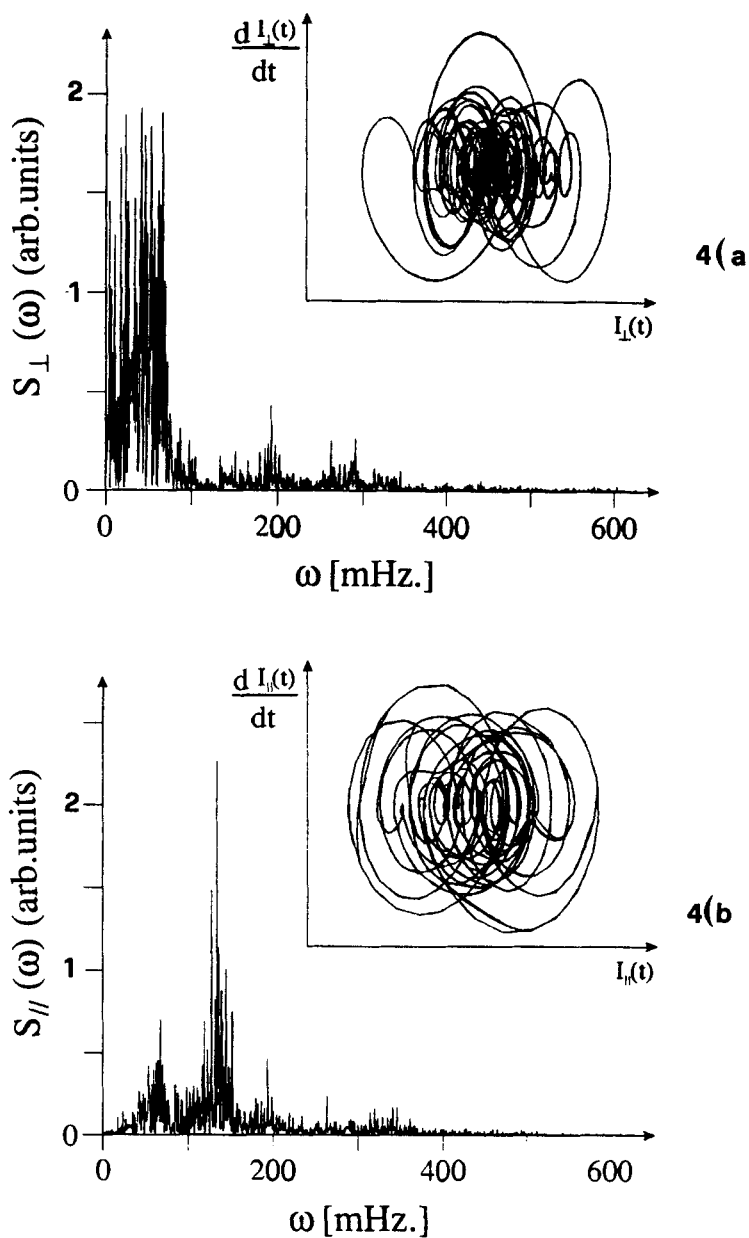


FIGURE 4 Fourier spectra $S_{\perp}(\omega)$ (4a) and $S_{\parallel}(\omega)$ (4b) along with projections in the phase space at impinging power $P_{\text{inc}} \approx 700$ mW.

Measurements of $I_{\perp}(t)$ and $I_{//}(t)$ performed in the chaotic regime ($P_{\text{inc}} \approx 700$ mW) have been used to calculate ν by standard methods. For both $I_{\perp}(t)$ and $I_{//}(t)$ it has been found²²: $\nu = 2.85 \pm 0.05$. This low value indicates that the answer to the first question is positive: it should be possible to describe our phenomenon by a low dimensional system of differential equations; probably, a system of three equations should be enough.

The second step consists in finding, at least, the largest Lyapunov exponent λ of the system. It represents a measure of the divergence of nearby orbits and characterize the sensitive dependence on initial conditions of the physical system. calculation of both $\lambda(I_{\perp})$ and $\lambda(I_{//})$ gives²²:

$$\lambda(I_{\perp}) = (0.1 \pm 0.015) \text{ s}^{-1} \quad (1)$$

$$\lambda(I_{//}) = (0.03 \pm 0.005) \text{ s}^{-1} \quad (2)$$

It is well known from theory²² that the chaoticity of the system is proved if $\lambda > 0$. This is true however only for a theoretical system modelled by a system of differential equations. In our case, the values of λ have been obtained from experimental signals which naturally contain noise. Therefore, values (1) and (2) of λ are meaningful only if compared with the values obtained when the system is in the periodic regime. Calculations performed from experimental data obtained at the impinging power $P_{\text{inc}} \approx 420$ mW give²²:

$$\lambda(I_{\perp}) \approx \lambda(I_{//}) \approx 0.001. \quad (3)$$

Comparison with values (1) and (2) proves that also the answer to the second question is positive: our phenomenon undergoes deterministic chaos. The time interval after which the motion becomes unpredictable is of the order of $1/\lambda$ and can be estimated from (1) and (2): $1/\lambda(I_{\perp}) \approx 10$ sec and $1/\lambda(I_{//}) \approx 33$ sec.

Finally, in order to answer to the third question, we must look at the features of signals in the chaotic regime: A satisfactory theoretical model should exhibit the same features.

EVIDENCE OF INTERMITTENCY

The main feature exhibited by signals in the chaotic regime can be deepened by looking at their time behaviors and Probability Distribution Functions (PDF) on different time scales. First of all, we have averaged both $I_{\perp}(t)$ and $I_{//}(t)$ on different time scales by evaluating the integral

$$\langle S(I, \Delta t) \rangle = (1/\Delta t) \int_t^{t+\Delta t} I(s) ds \quad (4)$$

for different Δt values. In Figure 5 (a, b, c), the time evolution of $\langle S(I_{\perp}, \Delta t) \rangle$ and $\langle S(I_{//}, \Delta t) \rangle$ is reported for three different Δt values. We note some peculiarities of $\langle S(I_{\perp}, \Delta t) \rangle$ which are not present in $\langle S(I_{//}, \Delta t) \rangle$. In fact, $\langle S(I_{\perp}, \Delta t) \rangle$ behaves similar on the different time scales: One peak in the larger time average is resolved into two or more peaks in the smaller time averages; this behavior is typical of a fractal structure. Furthermore, on all time scales, there are regions of high intensity in a small time interval, followed by regions of small intensity in a large time interval. We can say that fluctuations are not homogeneous in time; rather, they are intermittent: rare events have a probability which is greater than the Gaussian PDF. Indeed, we have calculated $P(S_{\perp})$, that is the PDF of $\langle S(I_{\perp}, \Delta t) \rangle$, and $P(S_{//})$, that is the PDF of $\langle S(I_{//}, \Delta t) \rangle$. They are shown in Figure 6 for the same Δt values as in Figure 5. While $P(S_{//})$ looks like a bimodal almost Gaussian PDF on short time scale and an asymmetric unimodal PDF on large time scales, $P(S_{\perp})$ looks like a binomial PDF on large and short time scales, namely it has an intermittent behavior.

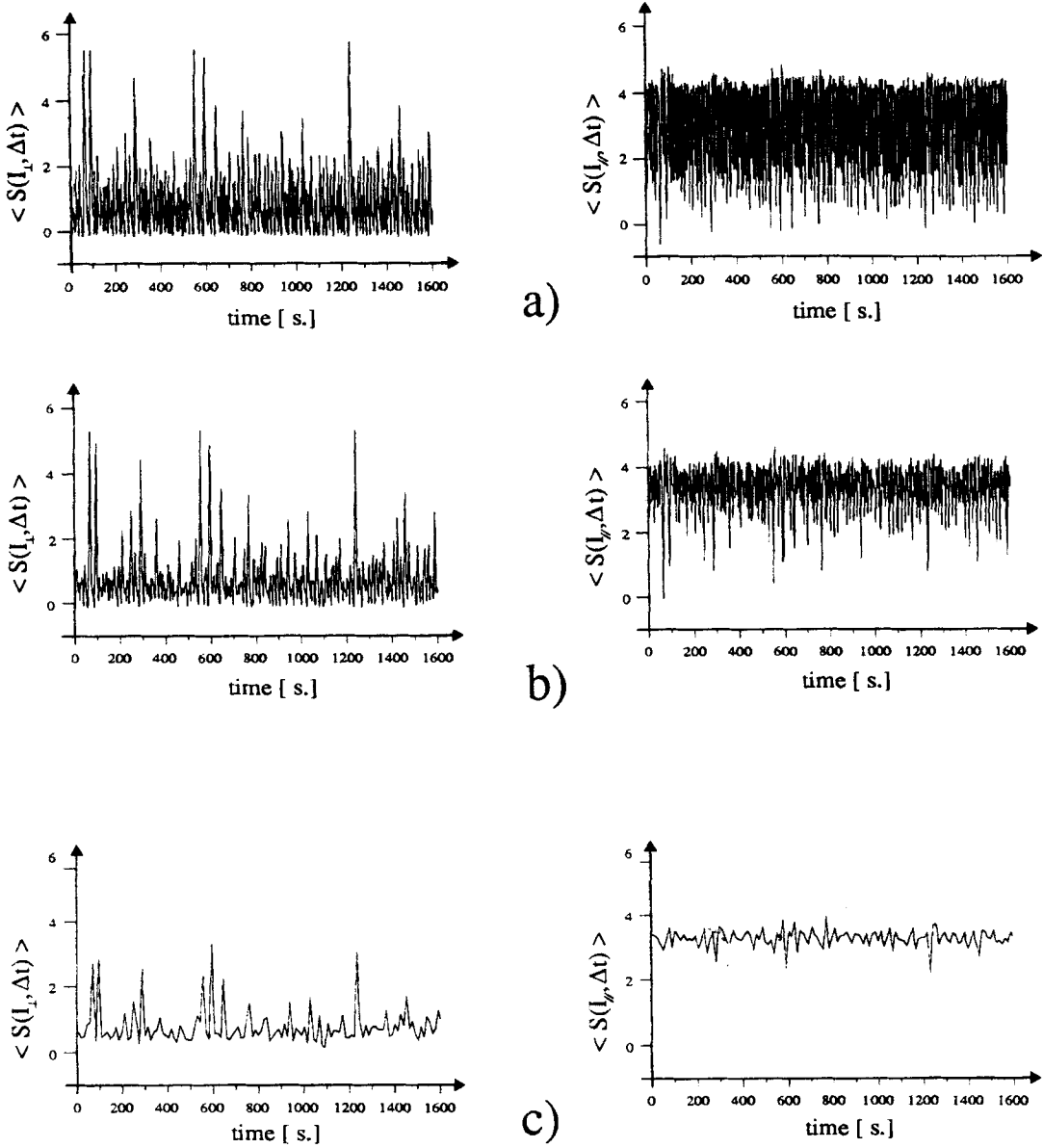


FIGURE 5 Time evolution of $\langle S(I_{\perp}, \Delta t) \rangle$ and $\langle S(I_{//}, \Delta t) \rangle$ reported for three average intervals: a): $\Delta t = 0.4$ sec.; b): $\Delta t = 1.6$ sec.; c): $\Delta t = 6.4$ sec.

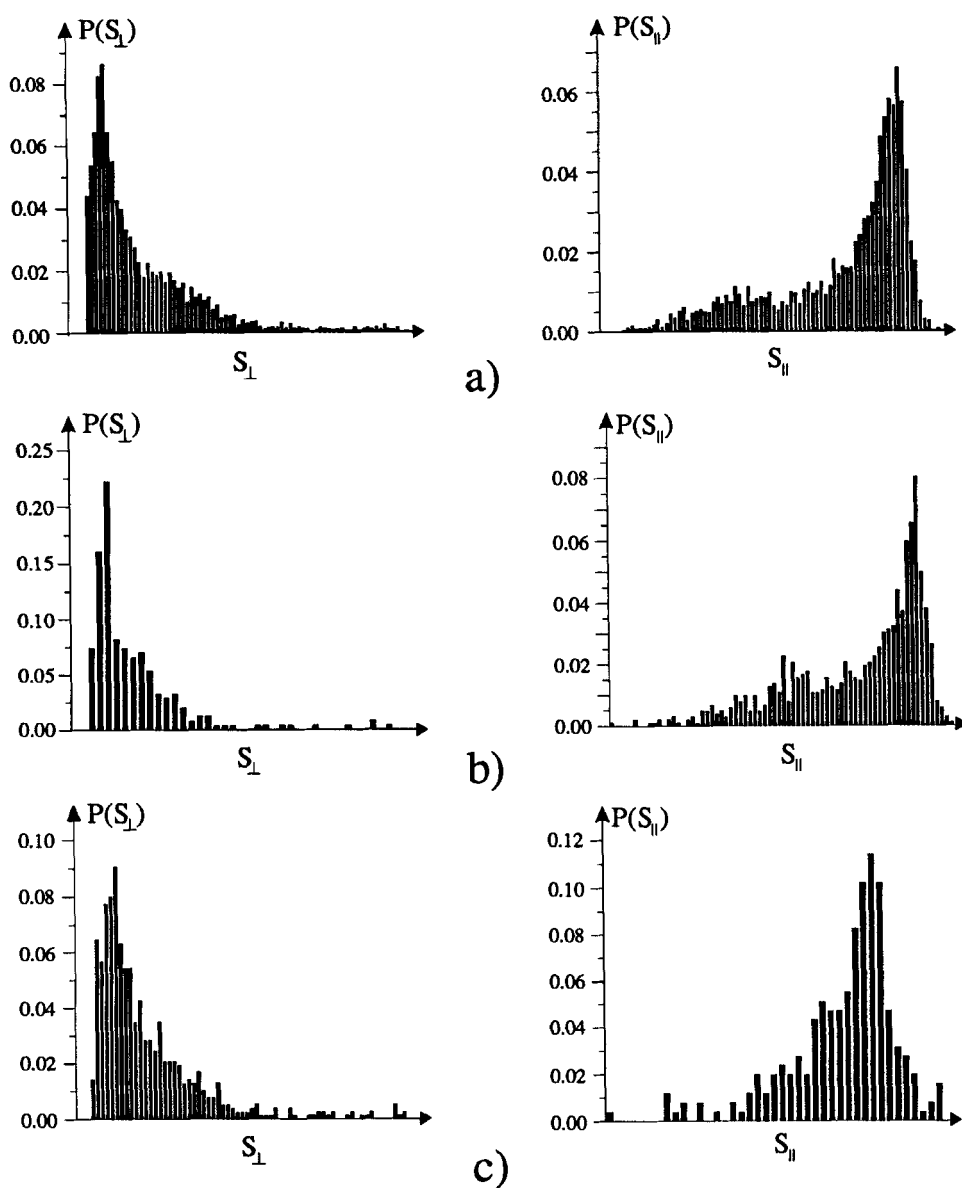


FIGURE 6 $P(S_{\perp})$ and $P(S_{\parallel})$ reported for three average intervals:

a): $\Delta t = 0.4$ sec.; b): $\Delta t = 1.6$ sec.; c): $\Delta t = 6.4$ sec.

In conclusion, in the phenomenon of molecular reorientation induced in NLC by an intense optical field (in a particular geometry) we have:

- a) Calculated the fractal dimension of the attractor for both the time series of experimental signals, getting the information that our phenomenon could be modeled by a low dimensional system of differential equations;
- b) Calculated the maximum Lyapunov exponent for the same signals, finding that, for $P_{inc} > 700$ mW, their behaviors become chaotic after time intervals which are respectively of the order of 10 sec for I_{\perp} and 33 sec for $I_{//}$;
- c) Shown intermittency, that is a non normal distribution of fluctuations of the transmitted light intensity (I_{\perp}). Since its the time evolution reflects (in a complicate and not clear way) the real motion of the director in the space, intermittency indicates the presence of places where the director "prefers" to stay, also in a chaotic motion.

Our analysis is useful to gain more insight into the observed phenomenon and the performed calculations open a new way to a theoretical understanding of the reorientation in terms of a better modeling of the interaction between the radiation and the matter.

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