PHYSICAL REVIEW E VOLUME 47, NUMBER 5 MAY 1993

Optically induced chaotic behavior in nematic liquid-crystal films

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(Received 7 July 1992)

In this paper we report the dynamical behavior occurring in a nematic liquid-crystal film during the optically induced molecular reorientation. The system at first undergoes an incomplete period-doubling cascade, then a biperiodic regime appears, and finally a transition to a chaotic regime occurs. This sequence is experimental evidence of a physical system that mixes two different routes for the transition to a chaotic regime.

PACS number(s): 05.45.+b, 42.65.−k, 61.30.−v

The optical Fréedericks transition occurring in nematic liquid crystals NLC's is due to the molecular director reorientation induced by the light field above a threshold, which is typical of the used material. Several authors [1] have observed and studied different features of this effect. A comprehensive description, however, which includes all the possible geometries, i.e., NLC orientation with respect to light polarization, and any kind of light polarization is not yet available. Among the different phenomena, the light self-pulsing which can be observed in the spatial pattern of the beam transmitted by the NLC film under particular geometries [2–4] is of great interest. Indeed, above the threshold, a precise (even if, generally, very complicated) dependence on the molecular director **n** exists for intensity and polarization in different points in this pattern [3]. Therefore, the time evolution of the transmitted intensity, detected for example in the center of the outcoming beam, is an evidence of the time behavior of the director reorientation. This self-pulsing effect has been studied in the case of circular and elliptical polarization of the light impinging normally on the NLC film [4]; although in these cases the authors did not report the transition of the system to a chaotic regime. A similar behavior is also observed when the linearly polarized light beam impinges on a homeotropically aligned NLC film and two conditions are fulfilled: the beam polarization is perpendicular to the incidence plane and the incidence angle is small but different from zero. Under these circumstances the typical ring pattern, due to self-phase modulation, appearing in the transmitted beam undergoes periodic oscillations giving rise to a limit cycle in the phase space [2]. The period of these oscillations depends on the impinging intensity and is in the range of seconds. Up to now no satisfactory theory has been worked out to describe this phenomenon even if it can be understood in the framework of the elliptically polarized radiation studied by Santamato *et al.* [4]. Under the above-mentioned geometry the linearly polarized beam will become elliptically polarized inside the NLC. For this reason we believe that a kind of rotation and nutation of the molecular director may occur in this case as well.

In this Brief Report we report a study of the pulsation effect showing that the above-mentioned limit cycle becomes unstable and the system takes a very peculiar route towards a chaotic regime. The peculiar characteristic of the transition to chaos which occurs in our system makes our experimental results very interesting in the framework of the ergodic theory of chaos. As the incident power is increased the system at first undergoes a period-doubling cascade which is stopped by the noise. Then through a phenomenon of "self-organization" a biperiodic regime appears and finally the universal transition from a quasiperiodic to a chaotic regime [5] occurs.

In our experiment, a light beam from an Ar^+ laser ($\lambda = 5145$ Å) is linearly polarized and focused (focal length $l_f = 150$ mm) on a homeotropically aligned NLC film (compound E7 from the British Drug House, thickness $d = 50$ μm). At normal incidence ($\mathbf{k} \| \mathbf{n}$, where $\mathbf{k}$ denotes the wave vector) and above a threshold value for the impinging light power, the formation of the well-known self-phase-modulation rings is observed on a screen placed after the sample. If the angle α between $\mathbf{k}$ and $\mathbf{n}$ is increased (in particular in the range $2° < \alpha < 15°$) and if the electric vector $\mathbf{E}$ of the light is normal to the plane of incidence, we observe that, above a power threshold that depends on α and on temperature T, the ring formation follows a pulsing behavior. Since the impinging linear polarization becomes elliptical in the medi-

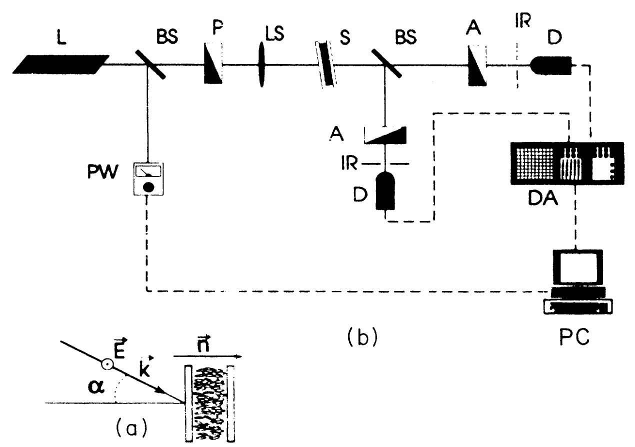

FIG. 1. (a) Geometry of the experiment for the case of oblique incidence of the light and vertical polarization. (b) Sketch of the experimental setup. *L*, Ar^+ laser; BS, beam splitter; PW, power meter; *P*, polarizer; LS, lens; *S*, sample; *A*, analyzer; IR, iris; *D*, detector; DA, data analyzer; PC, personal computer.

um, we detect, in the center of the outcoming beam, the intensity of the two components ($I_{\parallel}, I_{\perp}$) polarized respectively parallel and perpendicular to the incoming polarization. The experimental setup is shown in Fig. 1. A beam splitter (BS) placed after the sample separates the beam into two branches which are analyzed by two crossed polarizers. The signals from the detectors are sent to a personal computer which enables their storage for a further processing (fast Fourier transform). In Figs. 2 and 3 we report the measurement obtained for $\alpha=7°$ and at room temperature. The oscillating time behavior of $I_{\parallel}$ and $I_{\perp}$ for different increasing values of the impinging light power $P_{\rm in}$ are shown, along with their power spectra. When $P_{\rm in}$ increases, the detected signal $I_{\perp}$ undergoes a transition from a clear periodic regime to a chaotic one (while the time behavior of $I_{\parallel}$ does not exhibit the same strong dependence on the impinging power as $I_{\perp}$ does). However, looking at the power spectrum of $I_{\perp}$, we observe some peculiar features which can be only partially recognized in the standard views of such a transition. In the first case [Figs. 2(a) and 3(a)] the power spectrum shows a single fundamental frequency indicating the periodic pulsation of the rings (limit cycle). As $P_{\rm in}$ is increased at first the frequency decreases, thus indicating that the period of the oscillations increases; then the system becomes unstable and a period doubling appears (in some cases we have observed also an incomplete subharmonics cascade). In the spectrum relative to P_b [Fig. 3(b)], we can indeed recognize the frequencies belonging to the subharmonics of period 4. At higher $P_{\rm in}$ values,

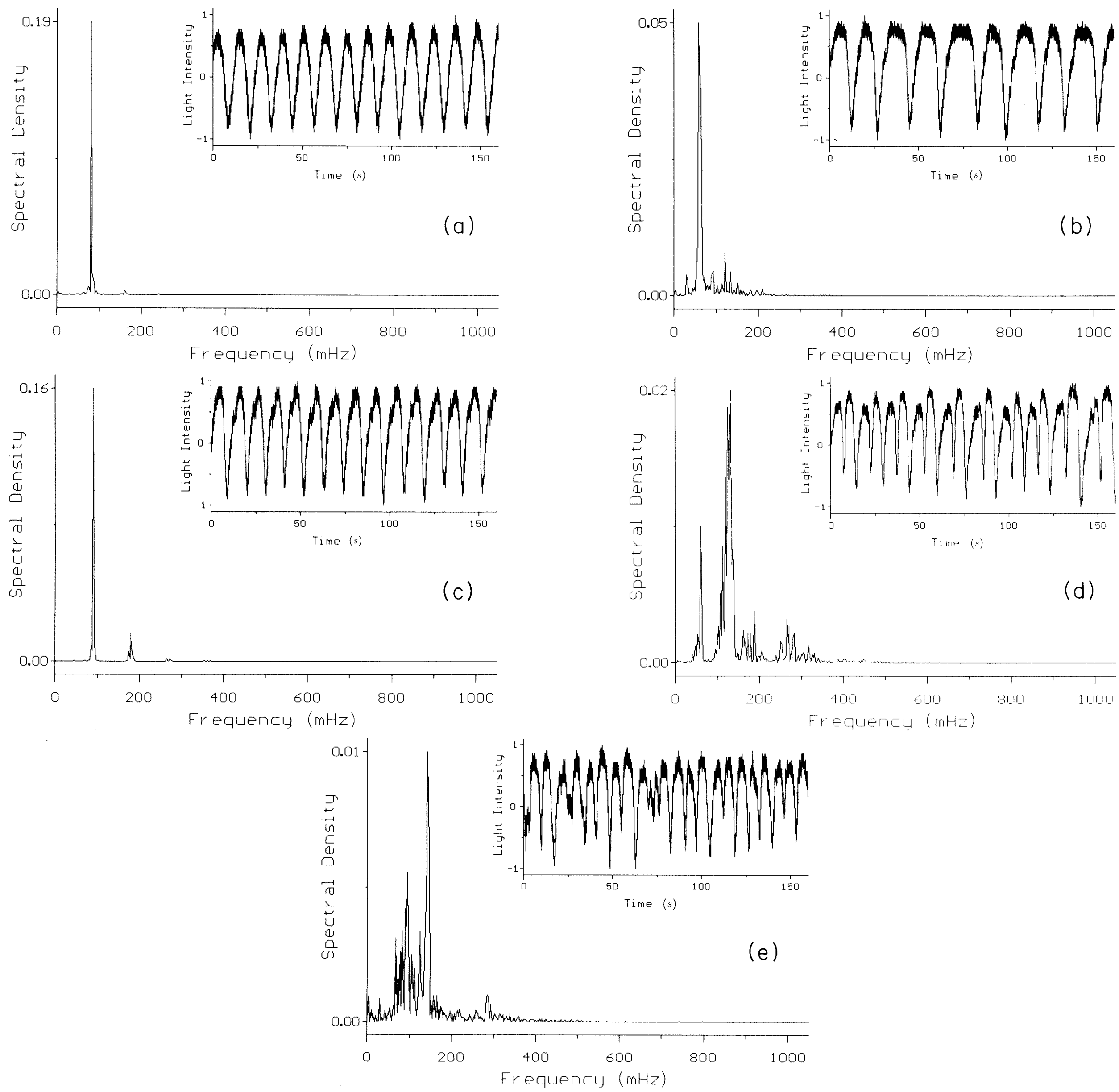

FIG. 2. Power spectra of the signal $I_{\parallel}$ for different increasing values of the impinging light power $P_{\rm in}$. (a) $P_a=350$ mW; (b) $P_b=420$ mW; (c) $P_c=480$ mW; (d) $P_d=650$ mW; (e) $P_e=680$ mW. In the little squares, the fluctuations of the light intensity are reported.

the noise destroys the occurrence of further subharmonics and one can find a noisy power spectrum. Nevertheless when the value P_c is reached, a particular feature appears: we can observe that two frequencies grow up from the noise, one being two times larger than the other [Fig. 3(c)]. We think that this behavior can be due to a kind of "self-organization" [6] of the system, which allows the system to give rise to a biperiodic motion again. Since the same effect is observed also in $I_{\parallel}$, obviously we argue that this "self-organization" can arise from a nonlinear coupling between $I_{\parallel}$ and $I_{\perp}$: in fact they oscillate at the same two frequencies f and $f/2$. When $P_{\rm in}$ is further increased, f increases both in $I_{\parallel}$ and $I_{\perp}$ until the value of f for $I_{\parallel}$ (say $f_{\parallel}$) becomes a little bit different from $f_{\perp}$ and we observe a phase locking of $f_{\parallel}$ and $f_{\perp}$. The locking ratio $f_{\parallel}/f_{\perp}$ is estimated (within the experimental accuracy) to be close to 26/27. At this stage, a number of sharp frequencies appear in the spectrum of $I_{\perp}$ [Fig. 3(d)], the value of each frequency being expressed as the ratio between two rational numbers. Finally, for higher $P_{\rm in}$ values, the peaks broaden and the gaps between them become shallower [Fig. 3(e)]. This last trend is typical of the universal transition from a quasiperiodic regime, characterized by two incommensurable frequencies, to a regime which exhibits, in the phase space, a chaotic motion on a strange attractor [5,7].

Therefore we observe the following sequence: (i) a period-doubling cascade stopped by the noise; (ii) the onset of new periodic regime; (iii) a transition from a periodic to quasiperiodic (mode-locked) regime; (iv) finally a transition from a quasiperiodic to chaotic regime. We emphasize that this sequence (i)–(iv) is experimental-

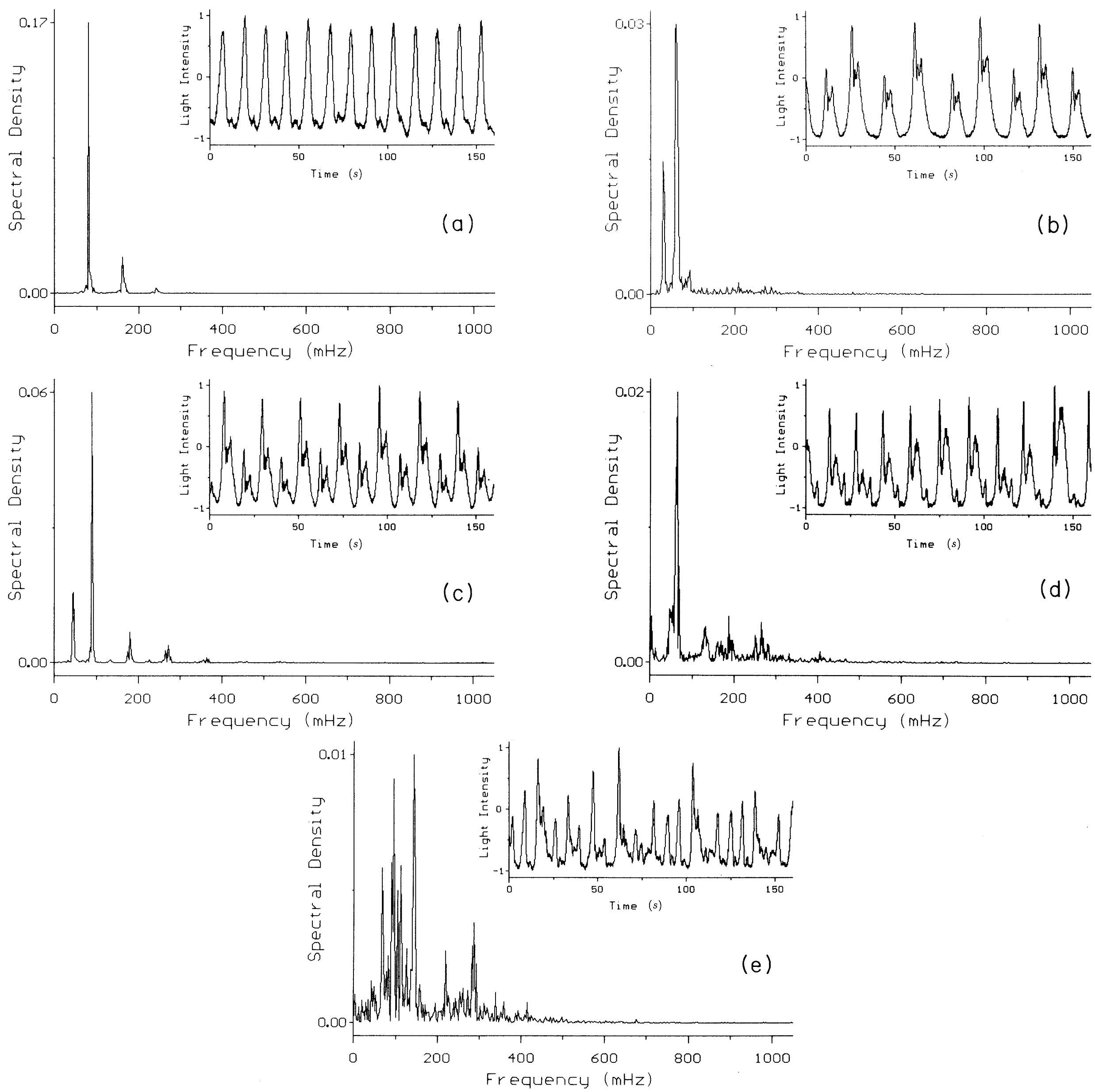

FIG. 3. Power spectra of the signal $I_{\perp}$ for the same power values as in Fig. 2.

ly observed in the same physical system, in which only the control parameter is varied while all the other physical parameters remain unchanged. In fact interesting experimental observations and detailed quantitative characterizations show that when only the control parameter is varied, the system can follow either of two different routes; (a) a transition periodic $\rightarrow$quasiperiodic (mode-locked)$\rightarrow$chaotic regime [8], or (b) a transition periodic$\rightarrow$period-doubling cascade$\rightarrow$chaotic regime [9] (in some papers [10,11] it is reported that this route is stopped at the period-doubling cascade stage by the noise which does not allow the process of the cascade towards the chaotic regime). The sequence (i)–(iv) that we observe shows the two routes (a) and (b) mixed together.

In order to study the dynamical properties of the signal in the chaotic regime, two main steps can be performed [7]: the determination of the fractal dimension of the attractor and the determination of, at least, the largest Lyapunov exponent of the trajectory embedded in the phase space. In order to get several data points, we carried out a new set of measurements in which we detected the total output intensity $I_{\text{out}}=I_{\parallel}+I_{\perp}$ whose power spectrum shows the same behavior as $I_{\perp}$. This procedure is chosen because of limitations of the data acquisition and storage capability of our equipment. In fact, in this way we can double the number of available data points. The storage time of each experiment is 5000 sec, with a sampling time constant $\Delta t=0.3052$ sec, so that we obtain a time series of $N=16\,384$ points. The orbit on the attractor in an embedding phase space of dimension d_E [7] is reconstructed by forming the d_E-tuples $\mathbf{X}(t)=\{I_{\text{out}}(t), I_{\text{out}}(t+\tau),\ldots,I_{\text{out}}[t+(d_E-1)\tau]\}$, where τ is a suitably chosen lag time (we have used $\tau=1.8$ sec which is the value of the first zero of the autocorrelation function of the data).

Using the N_p points of the vector $\mathbf{X}$, we have computed the correlation sum, $C(r)=(1/N_p^2)\sum_{i,j}\Theta(r-\|X_i-X_j\|)$, where Θ is the Heaviside function and $\|X_i-X_j\|$ is the usual norm in the embedding phase space R^{d_E}. Then the information dimension ν of the attractor can be found from the relation $C(r)=r^{\nu}$ [12].

If the embedding dimension d_E is large enough, the slope of the plots $\ln[C(r)]$ vs $\ln(r)$ settles down to a value independent of d_E. In our case, the slope, for $d_E\geq 9$, converges to the value $\nu=3.35$ which represents the information dimension of our attractor. As is well known, the chaoticity of the attractor is unambiguously proved if at least the largest Lyapunov exponent, say λ, is larger than zero. Since the value of λ is affected by the noise present in the experimental data [5,7], the system should be characterized by numbers that are the Lyapunov exponents for the noise-free system, averaged over the range of noise-induced states [13]. As a matter of fact, in the present analysis, the precise value of λ is not necessary: we need only know if $\lambda>0$. Therefore we have estimated λ, using an algorithm similar to that developed by Wolf *et al.* [14]. The estimate has been carried out from experimental data obtained at different values of P_{in} in the chaotic regime. In all cases, we found that λ increases with P_{in}, and converges towards positive values: $\lambda\geq 0.08$. This is well above the value $\lambda\approx 10^{-3}$ that we have calculated for the case of an experimental periodic signal and that represents our real zero value. The convergence of λ for different lag times and different embedding dimensions has also been tested.

A more detailed analysis of the phenomenon requires the evaluation of the entire spectrum of the Lyapunov exponents as well as the precise determination of the critical orbit above which the chaos grows up. Furthermore, a deeper insight could be given by performing measurements at different temperatures and at different α. These are in progress and will be reported in a forthcoming paper.

In conclusion we have reported an experiment carried out on a dissipative nonlinear NLC system which was not purposely set up to observe a particular route to chaos. In this system, above a power threshold, two (at present not clear) competitive mechanisms try to reorient the director, causing a periodic behavior in its dissipative reorientation motion and therefore a pulsation in the self-phase-modulation ring formation. When further increasing the power, an instability and then a transition to chaos arises; as it evolves towards the chaotic state, the system shows a very peculiar behavior. This behavior could be explained as a kind of intermittence effect in which the period-doubling cascade is stopped by the noise and the quasiperiodic-to-chaotic transition occurs.

This work was partially supported by Ministero dell'Università e della Ricerca Scientifica e Tecnologica, Italy. R. Bartolino, V. Carbone, G. Cipparrone, C. Umeton, and C. Versace are also with Consorzio INFM, unità di Cosenza, Italy; F. Simoni is also with Consorzio INFM, unità di Napoli, Italy. The authors thank L. Fronzoni for helpful discussions.

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