Homoclinic gluing bifurcations during the light induced reorientation in nematic-liquid-crystal films

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An *s*-polarized laser beam that impinges at small incidence angle on a homeotropically aligned nematic liquid crystal produces very interesting nonlinear phenomena. In this paper, we show that, due to the symmetry of the system, a cascade of successive homoclinic gluing bifurcations is responsible for the transition towards a stochastic regime in the experiment. We compare the experimental results with a model describing a sequence of gluing bifurcations.

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Light impinging on a nonlinear medium is one of the simplest tools to investigate nonlinear phenomena [1]. In particular, interesting optical effects due to the molecular reorientation induced by the laser beam in nematic-liquidcrystal (NLC) films have been widely studied in the past 20 years [2]. Very interesting dynamical behavior can be observed in the case of an ordinary wave that impinges with a small angle on an omeotropically aligned NLC sample [3-5]. These experiments showed the presence of persistent oscillations of the diffraction rings observed in the far-field pattern of the transmitted beam. As the incident power is increased, the dynamical behavior becomes more complex with a series of bifurcations towards a stochastic regime [4,5]. In a first series of experiments [4], we observed a kind of unexplained self-organization of the system, before the transition to chaos. In the last experiment [5,6], we improved the experimental apparatus and this allows us to better observe the phenomenology of the dynamical behavior. Even if experiments are very simple, equations describing the dynamics are very complicated, the phenomenon being described by the nematodynamics equations coupled with Maxwell's equations for the optical field [7].

Recently [8], it has been suggested that the observed behavior towards an irregular regime in the above reported experimental geometry could be described by a cascade of gluing bifurcations [9]. The phenomenology of the gluing bifurcation requires two attractors that lie at the opposite side of a saddle separatrix in the phase space. As the external parameter is increased, the unstable homoclinic orbit approaches the separatrix in a way that the old attractors are destroyed and a new attractor is created in the same place. At this point, two possible scenarios can take place [10] according to whether the structure arising from the destruction of the homoclinic orbit will be unstable or stable. In the first case, a Lorenz-like route to chaos is visible, while in the second case the destruction of the orbit is accompanied by the rising of a stable periodic regime. In this last case, as the control parameter is increased, successive homoclinic connections occur through which the original attractors disappear and new periodic orbits are continuously created. In general, at the end of the cascade, a chaotic attractor occurs. Of course, the homoclinic orbit exists only when a given symmetry is present in the system. In the simplest case in which the phase space is three dimensional, the symmetry $(x,y,z) \rightarrow (-x,-y,z)$ is required [9], which is a rotation of π around the *z* axis. This kind of phenomenology has been observed both in some numerical experiments [11] and in experiments with both nonlinear devices [12] or using a carefully chosen experimental geometry [13]. In the first experiment [12], a nonlinear optothermal device, made by an interferometric cavity with an absorbing input mirror and a multilayer spacer of alternatively opposite thermo-optical materials, is proposed. The absence of a proper symmetry requires the presence of two control parameters for the phenomenology to be present. In the second case [13], critical flows are built up using a cylinder containing water mounted on a loudspeaker. This device is oscillated in the vertical directions and excited surface waves appear. In this case, the required symmetry is produced with a proper choice of the experimental geometry.

Some time ago, Arneodo *et al.* [9] introduced a cascade of gluing bifurcations as a new route to chaos in a Lorenz-like model. The system we use is represented by three ordinary differential equations (ODE) where a cubic nonlinearity is present for the required symmetry,

$$x = \alpha x - \alpha y,$$

$$\dot{y} = -4\alpha y + xz + \mu x^{3},$$
 (1)

$$\dot{z} = -\delta\alpha z + xy + \beta z^{2}.$$

The parameters $1 < \delta < 4$, α and β are arbitrary constants, while $\mu \sim 1/\rho$ is the external control parameter. Here we report time series obtained with the same values, $\delta = 1.5$, $\alpha = 1.8$, and $\beta = -0.07$, as reported in the paper by Arneodo *et al.* [9]. The first gluing bifurcation happens at $\mu = 0.076071$ (see Ref. [9] for the detailed sequence of bifurcation points).

Liquid-crystal molecules are anisotropic and possess permanent dipoles [7]. In bulk form, these molecules tend to align in a way that their collective dipole moment vanishes. In other words, most phases of liquid crystals, such as the nematic phase, are characterized by a centrosymmetry, due to the equivalence between both the directions n and -n, of the molecular director orientation. The coupling between this nonlinear medium and the optical field could originate the nonlinear gluing process without *ad hoc* experimental conditions. Demeter and Kramer [8] derived a system of N ODE describing the Fourier coefficients of the time behavior of both polar and azimuthal angles of the molecular director. Then the authors investigate and discuss the case in which only three equations are retained. In our experiment (described later), we measure the time variations of a polarization state of a probe beam that goes through the region of the sample in which the nonlinear dynamics is induced. Then we get time series for both the ellipticity and the azimuthal angle of the major axis of a light probe beam. These quantities are related, even if not in an explicit way, to the dynamical variables used in Ref. [8]. For these reasons, rather than with the model described in Ref. [8], we prefer to compare our experimental results with the simpler model that describes the sequence of gluing bifurcations [9] from a generic point of view.

In our experiment, whose apparatus has been extensively described in [6], an s-polarized light beam impinges on a homeotropically aligned NLC film with a small incident angle of 5°. The sample was a 75- μ m NLC (E7 by Merck) cell. We use a pump-probe technique to reveal the dynamics of the molecular director induced in the sample by the light pump beam. The probe beam (He-Ne laser) is sent at normal incidence on the NLC film and focused on the sample by a focal length shorter than those of the pump beam (Ar^+ laser). The polarization of the transmitted probe beam is analyzed by a four-detector polarimeter [14], which allows us to measure the four Stokes parameters. From the measured time series of these parameters, we calculate the azimuthal angle of the major axis $\Theta(t)$ and the ellipticity e(t) [14,6]. The external control parameter is $\rho = I/I_{\text{th}}$, where I is the pumpbeam light intensity and Ith is the optical Freedericksz transition (OFT) threshold for normal incidence. As ρ is increased, by looking at both $\Theta(t)$ and e(t), we observe the birth of successive different dynamical regimes. The sequence of regimes for both $\Theta(t)$ and e(t) is exactly the same for a fixed value of ρ [6], and in the following we report results only for $\Theta(t)$.

In Fig. 1, we show the time evolution of both $\Theta(t)$ for four different values of ρ (left-hand panels) and of x(t) for four different values of the parameter μ (right-hand panels). The first three panels show the behavior of the gluing bifurcation as observed through time series. In Fig. 2, we show the phase space reconstructed from the observed time series, that is, $\Theta(t+\tau)$ versus $\Theta(t)$ for $\tau=5$ sec, and the twodimensional projection y(t) versus x(t) of the phase space from the model. Finally, in Fig. 3 we report the Fourier spectra for both experiments and the model. After a first look, we can see that very surprisingly the simple model (1) reproduces with good qualitative fidelity what is observed in the real experiment. More properly, from Fig. 1(a) we can see that, after a Hopf bifurcation that happens at $\rho = 1.6$, the trajectory lies in one side of the saddle separatrix in the phase space, that is, persistent oscillations of one definite sign are present. The sign of $\Theta(t)$ is constant, and can depend on the initial conditions of the sample.

As ρ is increased, the two attractors approach the separatrix at the origin and the trajectory in the phase space jumps randomly from one side to the other [Fig. 2(b)]. This is a very unstable regime, and we observe persistent oscillations with both signs in the time series [Fig. 1(b)]. The same behavior is visible in the time evolution of the simple model.



FIG. 1. We show the time behavior of both $\Theta(t)$ (left-hand panels) derived from our experiment, and x(t) derived from the model by Arneodo *et al.* [7] (right-hand panels), for four different values of the parameters ρ (from experiments) and μ (from the model). Namely (a) $\rho = 1.6$ and $\mu = 0.0761$; (b) $\rho = 1.9$ and $\mu = 0.076071$; (c) $\rho = 2.3$ and $\mu = 0.076$, and (d) $\rho = 4.2$ and $\mu = 0.02$.

The time periods during which the system visits one side of the attractor are random, even though we noted that they tend to reduce as the time goes on. Looking at the Fourier spectra (Fig. 3), we can see that this phase is accompanied by an increase of the low-frequency part of the spectrum, due to the stochastic jumps that introduce a kind of low-frequency oscillation. It is worthwhile to mention that a similar phenomenology is present in the Lorenz system when the control parameter is set to r = 13.96, that is, lower than the value $r \approx 28$ required for the chatic behavior due to the usual Lorenz strange attractor (see Ref. [15]). We observe this phenomenology in our experiments as well as in the model.

The chaos observed in the above regime is not attracting, and is named "transient chaos" in Ref. [15]. The meaning of the word "transient" can be recovered in experiments. In fact, as the control parameter is further increased and homoclinic trajectories glue at the origin, a new attractor is created and the trajectory visits both sides of the phase space in an alternate way [Figs. 1(c), 2(c), and 3(c)]. The system now lies on a new stable symmetric orbit and the gluing bifurcation happened. In this regime, the spectrum is characterized by a single fundamental frequency (Fig. 3). Then the



FIG. 2. We show the reconstructed phase space $\Theta(t+\tau)$ vs $\Theta(t)$ from experiments (left-hand panels) with $\tau=5$ sec, and the two-dimensional projection y(t) vs x(t) of the phase space from the model (right-hand panels). Different panels refer to different values of parameters as reported in Fig. 1.

broadband spectrum of Fig. 3(b) disappears, and the whole dynamics that characterizes the gluing bifurcations can be seen as a kind of "self-organization" of the system [4]. After the first gluing just described, as ρ is further increased, a new gluing bifurcation takes place in the experiment, and finally a stochastic regime is observed [Figs. 1(d), 2(d), and 3(d)]. As we can see from Figs. 1–3, this last regime is very noisy with regard to the experiment with respect to its numerical counterpart, and a broadband spectrum is present (see Fig. 3). At high values of ρ , from a technological point of view, the anchoring of molecules at glass surfaces is destroyed after a finite time, and the measurements are no longer possible [5]. This implies that for high values of ρ , experimental time series are relatively short, and unfortunately this rules out the possibility of a proper investigation of the stochastic regime.



FIG. 3. We show Fourier spectra calculated through both time series $\Theta(t)$ from experiments (left-hand panels), and x(t) from the model (right-hand panels). Different panels refer to different values of parameters as reported in Fig. 1.

As a conclusion, we found that a sequence of successive homoclinic gluing bifurcations is at the heart of the transition to chaos in light-induced reorientation in nematic-liquidcrystal films. This has been predicted by Demeter and Kramer [8]. Our system is not an *ad hoc* experiment to reproduce both the transient and permanent homoclinic chaos, but gluing bifurcations happens in a natural way due to the symmetry of the system. To our knowledge, this is the only system that behaves in this way. We find that the gluing bifurcations appear like a sequence of successive selforganized states of the system, and a kind of "transient chaos" is present. The time behavior of the measured quantities, namely the ellipticity and the azimuthal angle of the major axis of the light probe beam, is surprisingly reproduced by a very simple model of three ODE's introduced some time ago by Arneodo et al. [9] to reproduce the gluing bifurcations cascade.

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