On-off intermittency in chaotic rotation induced in liquid crystals by competition between spin and orbital angular momentum of light

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We observed on-off intermittency in the chaotic rotation induced by a cw laser beam in a thin liquid crystal film where the spin and the orbital angular momentum of light compete in reorienting the sample. We found that the azimuthal angle $\phi(t)$ of the molecular director increased linearly in time on large time scales but, occasionally, it exhibited large fluctuations about its average value $\omega_0 t$, so that its angular velocity $\dot{\phi}(t)$ undergoes an on-off intermittent motion. The intermittent signal $\omega(t)=\dot{\phi}(t)-\omega_0$ obeyed the scaling laws of on-off intermittency, including the symmetry between laminar and burst phases. The chaotic rotations were observed only when the spin and the angular momentum of light were transferred simultaneously to the sample.

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Chaotic rotations have a central role in the study of the dynamics of bounded chaotic flows [1] and in the phase synchronization of coupled chaotic oscillators [2]. After a suitable choice of the phase space variables, many chaotic flows, such as those arising in Rössler's and Lorenz's systems, can be reduced to a chaotic rotation [1], characterized by on-off intermittency of its instantaneous angular velocity [3]. These results imply that the on-off intermittency may be more pervasive than previously thought, as its existence in chaotic flows apparently does not rely on symmetry in the system equations [1]. Nevertheless, observing intermittent chaotic rotations in real physical systems may be difficult, because the accessible time signals correspond usually to generic variables reflecting the chaotic features of the underlying dynamics, as stated by Takens' theorem [4]. To observe an on-off intermittency, one must pick up time signals corresponding to "suitable" variables that cannot be discovered easily [3]. For this reason, on-off intermittency was mainly studied in electronic circuits [5], although an observation of on-off intermittency was reported in a spin wave experiment [6], in a gas discharge plasma system [7], and also in a convective liquid crystal (LC) cell driven by random external voltage [8].

Here, we report the observation of a laser-induced chaotic rotation of the molecular director of a nematic liquid crystal accompanied by on-off intermittency of its instantaneous angular velocity. Our results provide experimental support to the chaotic rotational dynamics recently studied by Lai *et al.* [1]. Laser-induced chaotic oscillations in nematic liquid crystals was observed long time ago using an *s*-polarized laser beam at a small incidence angle [9]. The transition to chaos of this system was studied extensively both experimentally [10] and theoretically [11,12] and it was ascribed to a cascade of successive homoclinic gluing bifurcations [10–12]. In the present work, we used a different experimental configuration, namely, a strongly astigmatic circularly polarized laser beam at normal incidence. Our experimental geometry is very interesting on its own grounds, because the spin and the orbital angular momentum of light may both interact with the sample, leading to complex dynamics [13,14]. The extensive study of the observed dynamical regimes when the external control parameters (laser intensity, polarization, profile, etc.) are changed will be the object of a forthcoming paper; in this work we studied only the regimes where chaotic rotation and on-off intermittency were observed. Unlike other experiments about on-off intermittency, no external noise was supplied to the system, which is entirely governed by its own autonomous dynamics. In other words, we observed chaotic, rather than stochastic on-off intermittency. Our sample was a 50-μm-thick E7 nematic liquid crystal film enclosed between plane glass walls coated with octadecyldimethyl(3-trimethoxy-silylpropyl)ammonium chloride for strong homeotropic anchoring condition. A frequency doubled circularly polarized continuous wave Nd:YVO_4 laser beam at $\lambda=532$ nm was sent at normal incidence onto the sample. Two cylindrical lenses of focal lengths $f_x=500$ mm and $f_y=30$ mm, having their astigmatic axes carefully aligned along the vertical *y* axis and the horizontal *x* axis, respectively, were used to make the beam profile elliptical at the sample position. The radii ($1/e^2$ of maximum intensity) of the beam at the sample position were measured to be $w_x=80$ μm and $w_y=8$ μm, respectively. Our detection apparatus was described elsewhere [15]. It allows for real-time monitoring of the outer ring diameter $D(t)$ and the average polarization direction $\Phi(t)$ of the far field self-diffraction pattern. As it is well known, the angular divergence of the ring pattern is roughly related to the instantaneous space average value of the polar angle θ of the molecular director $\boldsymbol{n}=(\sin\theta\cos\phi,\sin\theta\sin\phi,\cos\theta)$ $(D(t)\propto\sin^2\theta(t))$, while the ring polarization measures the space average value of the azimuthal angle ϕ $(\Phi(t)\simeq 2\phi(t))$. The quantities $D(t)$ and $\Phi(t)$ provide roughly independent degrees of freedom, from which the time evolution of the components $n_x(t)$ and $n_y(t)$ of $\boldsymbol{n}$ in the plane transverse to the beam propagation direction can be estimated. The overall

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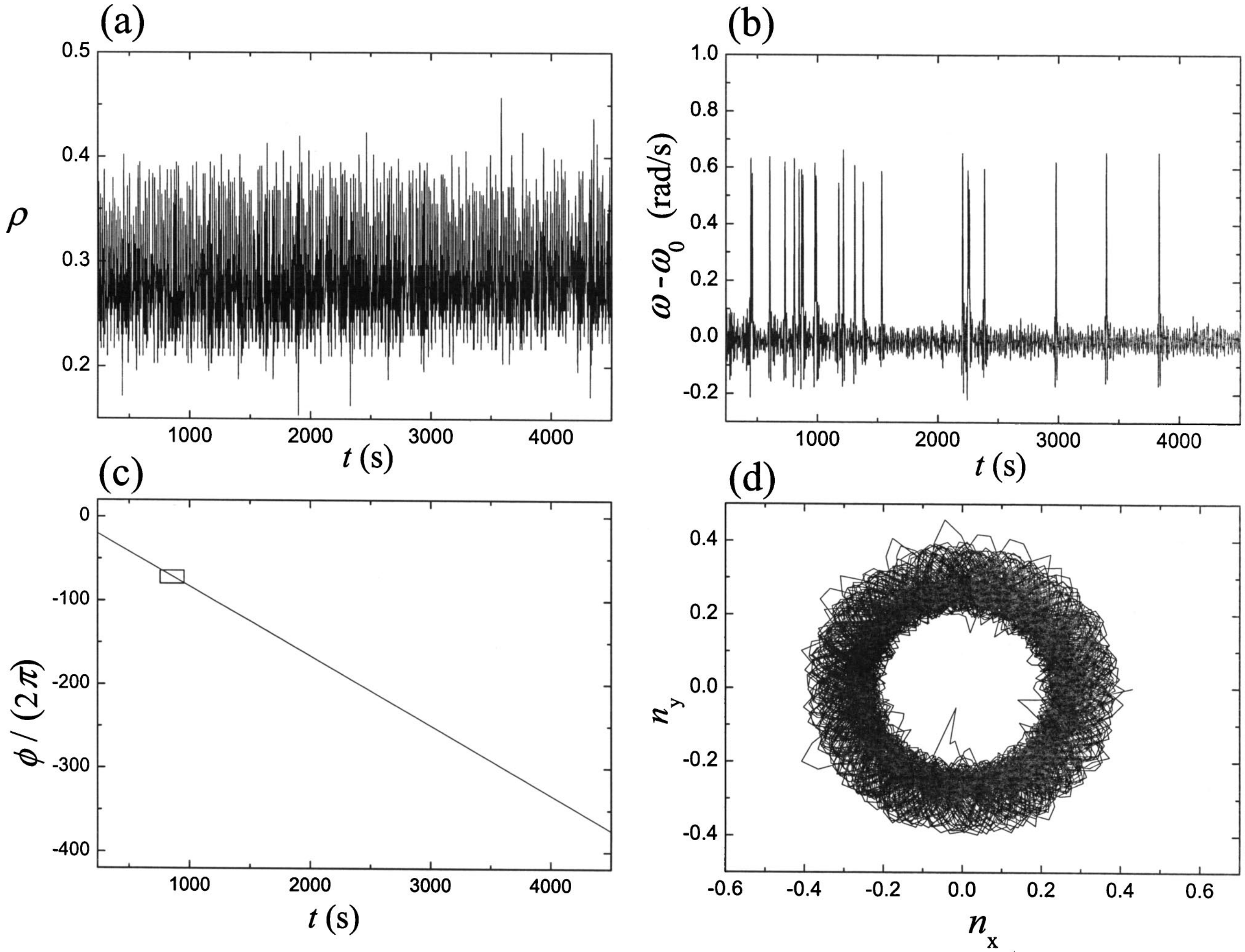

FIG. 1. (a) Radius $\rho(t)$ of the trajectory; (b) instantaneous angular velocity $\omega(t)$; (c) rotation angle $\phi(t)$, the rectangular region is shown enlarged in Fig. 2; (d) trajectory in the x,y plane of the chaotic rotation of the molecular director $\boldsymbol{n}$. All time traces were taken at an incident laser power $P=445$ mW, and in (a)–(c) the time scale is the same.

geometry of our experiment was the same as the one used to induce the collective rotation of the liquid crystal molecules with circularly polarized light [16], apart from the fact that the cross section of the laser beam was made elliptical. This last circumstance may change dramatically the dynamics of the system, because of the presence of the extra torque due to the orbital part of the angular momentum of light [13,14,17]. One of the most striking features observed in our experiment was that at a critical value of the incident power, the polar angle $\theta(t)$ of $\boldsymbol{n}$ was found to undergo apparent irregular oscillations, while the rotation of the azimuthal angle $\phi(t)$ was found to become chaotic and its angular velocity $\dot{\phi}(t)$ to become intermittent. At a very high incident power, both $\theta(t)$ and $\phi(t)$ were found to become chaotic. Typical time signals of $\rho(t)=\sin^2\theta(t)$ and $\phi(t)$ are shown in Figs. 1(a) and 1(b), respectively, and the trajectory of the chaotic rotation of $\boldsymbol{n}$ in the x,y plane is drawn in Fig. 1(d). Each time series acquisition lasted about 2.5 h after which the sample was damaged by its long exposure to the laser light. Our sampling rate was 0.34 s, which is one order of magnitude faster than the reorientational response time of the LC sample. We notice the irregular oscillations of $\rho(t)$ and the monotonic increase of $\phi(t)$ at large time scale, which are the main features of chaotic rotations [1]. In Fig. 1(c) we plotted the difference $\omega(t)=\dot{\phi}(t)-\omega_0$ of the instantaneous angular velocity $\dot{\phi}(t)$ and its time average value ω_0 as a function of time. The intermittent character of $\omega(t)$ is evident. The rectangular region in Fig. 1(b) is shown enlarged in Fig. 2. In the short time scale, the uniform rotation is interrupted by randomly distributed kinks, where $\phi(t)$ changes abruptly and $\dot{\phi}(t)$ undergoes a very large excursion. These kinks lead finally to the observed intermittency of $\omega(t)$. As shown in Fig. 2, a small noise is superimposed to the signal $\phi(t)$. This noise is not due to the experimental apparatus, but it reflects the random motion of the other degrees of freedom of the system, which are chaotic. Increasing the laser power, the chaotic noise becomes larger and larger, until its amplitude becomes comparable to the intermittent kinks. In these conditions, the intermittency is hard to see because the laminar phases of $\omega(t)$ become very short and they are randomly interrupted by the chaotic noise. The subsequent figures

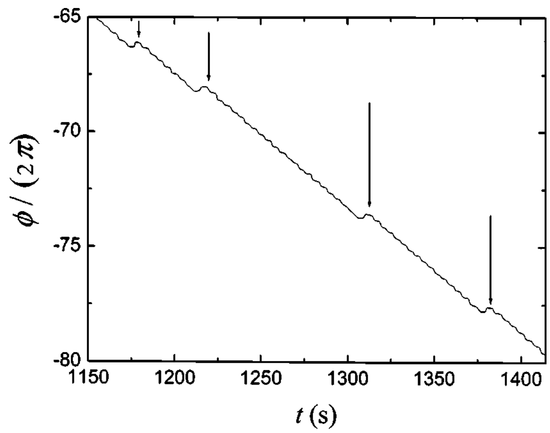

FIG. 2. Short time behavior of $\phi(t)$. The kinks pointed to by the arrows lead to the intermittency of $\omega(t)$.

show the tests we made to characterize the on-off intermittency of $\omega(t)$. The probability distribution Λ_τ of the duration τ of the laminar phase $\omega(t)=0$ is shown in Fig. 3(a) for a set of incident laser powers where on-off intermittency was observed. The distribution Λ_τ is characterized by the power law $\tau^{-3/2}$ at small τ. At large τ, the distribution decays exponentially, as expected [18] for on-off intermittency. In finding the slope of the curves, the points corresponding to just one or two events in the whole measurement time were discarded because they have no significant statistic. Nevertheless, we left these points in the figure to show that very long laminar phases where sometimes observed, which excludes type-I Pomeau-Manneville (PM) intermittency, where Λ_τ drops to zero at finite duration $\tau=\tau_c$ [19]. We checked also that the average duration $\overline{\tau}$ of the laminar phases was proportional to ϵ^{-1}, our best fit to the power law $\overline{\tau}\propto\epsilon^k$ yields $k=-0.98$

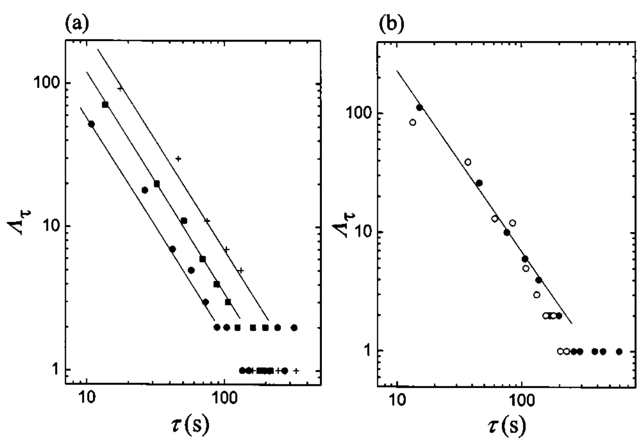

FIG. 3. (a) Log-log plot of Λ_τ for different values of the laser incident power. $(+)P=445$ mW, (□) $P=482$ mW, and (○) $P=520$ mW. The slope $-3/2$ at low τ is characteristic of on-off intermittency. The best fit results are $B(+)=-1.47\pm0.09$, $B(\square)=1.54\pm0.04$, $B(\bigcirc)=-1.52\pm0.06$, respectively. (b) Log-log plot of the distribution Λ_τ of the burst and laminar phases as a function of the duration τ. They have the same slope, which is the characteristic of on-off intermittency. The slopes of the best fit are $B=-1.5\pm0.2$ for the burst, and $B=1.52\pm0.06$ for the laminar phase.

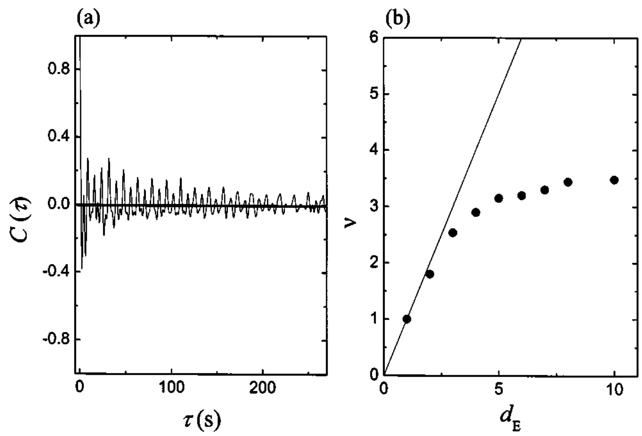

FIG. 4. (a) Autocorrelation function of $\rho(t)$; (b) correlation dimension ν as a function of the embedding dimension d_E. The full line is the bisectrix of the first quadrant.

±0.2. The bifurcation parameter $\epsilon=(P-P_{int})/P_{int}$, P being the incident laser power, was obtained by fitting the data to the cumulative probability $P(\tau_0)$ of having a laminar phase duration larger than τ_0. The threshold power P_{int} for intermittency was estimated by a linear fitting to $\epsilon(P)$, obtaining $P_{int}=445\pm8$ mW. All observed scaling laws of the lengths of the laminar phases are consistent with the type-III PM intermittency as well as with on-off intermittency. A peculiar feature of on-off intermittency, besides its scaling universality, is the symmetry between laminar and burst phases [20]. In the PM intermittency, in fact, the dynamics occurs in a neighborhood of an invariant fixed point or periodic orbit and, hence, the growing of the bursts is governed by the nonlinear dynamics far from this object, which is system dependent. In the on-off intermittency, the invariant object is a chaotic attractor, which is itself governed by universal scaling laws and, hence, the statistical properties of burst and laminar phases are independent of the details of the system and, actually, they are the same [20]. We measured the distribution function of the burst duration and compared it with the distribution of the laminar phases, as shown in Fig. 3(b). All curves have the same slope, as required by the symmetry of on-off intermittency (as before, the points corresponding to one and two events were discarded in doing the fit). Once the on-off intermittent character of $\omega(t)$ has been established, we proceed to study the chaotic behavior of $\rho(t)$. We conducted this study for a different power P of the incident beam, but we report here only our results at $P=445$ mW. The decay of the autocorrelation function $C(\tau)=\langle\rho(t)\rho(t+\tau)\rangle$ is shown in Fig. 4(a). From the figure we estimated a correlation time $\tau_c=100$ s. We used the standard TISEAN tools[1] to analyze the time signal $\rho(t)$. We looked first for the dimension ν of the chaotic attractor. The dimension ν estimated from the correlation sum is plotted in Fig. 4(b) as a function of the embedding dimension d_E. The asymptotic value of the curve provides the dimension of the attractor,

[1]The TISEAN software package is publicly available at http://www.mpimpks-dresden.mpg.de/~tisean

which in our case is $\nu = 3.50 \pm 0.05$. If $\rho(t)$ were governed by stochastic noise, the curve in Fig. 4(b) would be always increasing and for stochastic noise it would be along the bisectrix of the first quadrant. Thus, the figure demonstrates clearly that $\rho(t)$ is chaotic and not stochastic. With the same tools we calculated the maximum Lyapunov exponent λ and the entropy h of the attractor. We found $\lambda = (3.3 \pm 0.5) \times 10^{-3}\ \mathrm{s}^{-1}$ and $h = (9.5 \pm 0.7) \times 10^{-2}\ \mathrm{s}^{-1}$. The values of λ and h must be compared with the zero point values λ_0 and h_0 of the regular rotating regime. We found $\lambda_0 = (1.0 \pm 0.3) \times 10^{-3}\ \mathrm{s}^{-1}$ and $h_0 = (3.0 \pm 0.3) \times 10^{-2}\ \mathrm{s}^{-1}$. These values are lower than λ and h, as they should, but not so low to exclude a residual noise having stochastic origin. The origin of the residual noise is probably instrumental, because $\rho(t)$ was measured by the images recorded by the charge-coupled device that is sensitive to small fluctuations of the laser power. At high incident power P, in fact, λ and h become much larger than their zero point values, confirming the chaotic dynamics of $\rho(t)$. For example, at $P = 520$ mW we obtained $\lambda = (9.5 \pm 0.5) \times 10^{-3}\ \mathrm{s}^{-1}$ and $h = (10.8 \pm 0.7) \times 10^{-2}\ \mathrm{s}^{-1}$. We stress, however, that the character (chaotic or stochastic) of the noise does not affect the intermittency properties. As a final test, we tried to see if the intermittency of $\omega(t)$ was on-off or in-out [21,22]. The crucial difference between the two kind of intermittencies is that in the on-off case the system, after the burst, is reinjected into the same chaotic attractor, while in the in-out case the system passes, after the burst, to a different attractor [22]. Therefore, we collected all signals $\rho(t)$ before each burst of $\omega(t)$ in a time series and all signals $\rho(t)$ after each burst of $\omega(t)$ in a different time series, and then we analyzed the two series separately, calculating the dimension ν, the maximum Lyapounov exponent λ, and the entropy h of the attractor. We repeated this procedure by changing the fraction of the laminar phases of $\omega(t)$ where the data were taken from 0.5 to 0.25. In all cases we found, within the errors, the same values of ν, λ, and h in the two series; thus proving that the attractor around which the system evolves is only one. This result seems to exclude, in the present case, in-out intermittency.

The occurrence of chaotic rotations and on-off intermittency in our system can be understood as follows. Assume for a moment the laser beam having a circular profile. Then, after the reorientation, the liquid crystal behaves roughly as a birefringent plate having an optical axis at angle $\phi(t)$ and retardation $\delta(t)$ proportional to $\rho(t) = \sin^2\theta(t)$. When the circularly light beam passes through the sample, its polarization changes and the spin of each photon changes, on the average of $\hbar\Delta s_3 = \hbar(\cos\delta(t) - 1)$. Then, the molecular director of the liquid crystal starts to rotate with angular velocity $\dot{\phi}(t)$ proportional to the average angular momentum S_z transferred to the sample in unit time, given by $S_z = (P/\omega)(\cos\delta(t) - 1)$. In view of the cylindrical symmetry of the intensity profile, the polar angle θ stays constant during the rotation, which is uniform, because $\theta = \mathrm{const}$ implies that $\delta = \mathrm{const}$, $\rho = \mathrm{const}$, and $\dot{\phi} = \omega_0 = \mathrm{const}$, as confirmed by the experiments [16]. Assume now an elliptical profile of the laser beam intensity at the sample location. Then the polar angle θ will acquire an asymmetric profile in the x,y plane, and the incoming optical wave sees an astigmatic distribution of refractive index. The sample behaves now as a cylindrical lens having effective focal lengths $f_1(t)$ and $f_2(t)$, and cylindrical axis rotated at angle $\gamma(t)$. As it is well known, an astigmatic laser beam may transfer orbital angular momentum L_z to a cylindrical lens [13,23]. In general, $\gamma(t)$ is different from $\phi(t)$. It should be stressed, however, that the angles γ and ϕ are coupled because of the anisotropy of the elastic constants of the material: for nematic materials having the splay elastic constant larger than the twist elastic constant ($k_{11} > k_{22}$) the minimum elastic energy is reached when $\phi = \gamma$ [17]. In the experiment presented here, both S_z and L_z were transferred to the liquid crystal simultaneously, and all relevant degrees of freedom $\theta, \phi, \gamma, f_1, f_2$ are coupled together. The equation for the angle ϕ assumes the general form

$$\dot{\phi} = \omega_0 + F(\phi, \rho, \boldsymbol{x}), \tag{1}$$

where ω_0 is proportional to the time average $P/\omega\langle(\cos\delta(t) - 1)\rangle$, and $\boldsymbol{x} = (\gamma, f_1, f_2)$ is the set of variables related to the profile of the θ distribution in the x,y plane. A model based on these guidelines will be the object of a forthcoming work. Having a detailed model, however, is irrelevant here, because one of the most striking properties of on-off intermittency is its universality, i.e., its independence on the details of the nonlinear system [20]. In fact, taking the time derivative of Eq. (1) yields

$$\dot{\omega} = \alpha(t)(\omega + \omega_0) + \beta(t), \tag{2}$$

where $\omega = \dot{\phi} - \omega_0$. As shown in Fig. 1, ρ (and all other coordinates) undergoes a chaotic motion on time scales much faster than the average duration $\overline{\tau} \simeq 150$ s of the laminar phases, while ω changes slowly most of the time. The quantities $\alpha(t)$ and $\beta(t)$ may then be regarded as fast chaotic functions of time. We may take the long time average of Eq. (2) considering $\alpha(t)$ statistically independent of ω, thus obtaining $\overline{\alpha(t)}\omega_0 + \overline{\beta(t)} = 0$, which implies $\overline{\alpha(t)} = \overline{\beta(t)} = 0$. The two chaotic processes $\alpha(t)$ and $\beta(t)$ have therefore approximately zero mean. We may then pose [1] $\alpha(t) = \lambda_\perp + \alpha_0(t)$, where $\overline{\alpha_0(t)} = 0$ and $\lambda_\perp$ is a small positive transverse Lyapunov exponent, defined with respect to the "off" state. From our data on ω, we found $\lambda_\perp = 2 \times 10^{-3}\ \mathrm{s}^{-1}$, which is appreciably small, being about twice the zero reference level λ_0. In this way, Eq. (2) becomes identical with the model for on-off intermittency in the presence of noise treated, for example, in Ref. [24]. In the present case, however, the noise is related to the chaotic motion of the other coordinates of the system.

In conclusion, we reported the observation of a laser-induced chaotic rotation accompanied by on-off intermittency of the molecular director of a homeotropically nematic liquid crystal film. This process was recently modeled by Lai *et al.* [1]. Our experiment was carried out using an astigmatic circularly polarized laser beam at normal incidence with elliptical profile at the sample location. The observed chaotic rotation resulted from the competition between the optical torques originated from the spin and the orbital angular momentum of light.

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