

Liquid crystal reorientation induced by completely unpolarized light

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We studied the molecular reorientation in liquid crystal films induced by fully unpolarized laser beams. We found that, unlike ordinary materials, liquid crystals may be sensitive to higher order time correlations of the light polarization. A model accounting for the optical torque due to the second order time correlations of the optical polarization is also presented. Different kinds of fully unpolarized light may produce different torques. An example is shown in the experiment reported here. [S1063-651X(98)02803-7]

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I. INTRODUCTION

Fully unpolarized quasimonochromatic light is characterized by having coherence matrix $\mathbf{J} = \langle \mathbf{E}\mathbf{E}^* \rangle = \frac{1}{2} \mathbf{I}_2$ proportional to the two dimensional unit matrix \mathbf{I}_2 . This polarization state is the only one having complete rotational symmetry around the beam propagation direction z . The intensity I of the beam is defined here as $I = \text{Tr} \mathbf{J} = \langle |E_x|^2 \rangle + \langle |E_y|^2 \rangle$, where x and y denote the fixed laboratory axes. The brackets $\langle \rangle$ denote average over times much longer than the characteristic time τ_p of variation of the wave polarization state. We may also think of completely unpolarized light as being made of fully polarized light with a polarization state changing much faster than the response of the optical medium, so that all components of the reduced Stokes vector $\mathbf{s} = [|E_x|^2 - |E_y|^2, 2 \text{Re}(E_y^* E_x), 2 \text{Im}(E_y^* E_x)]/I$ average to zero: $\langle \mathbf{s}(t) \rangle = 0$. Among optical materials, liquid crystals proved to be extremely sensitive to light polarization, because they couple directly to the angular momentum carried by the radiation field [1-3]. Completely unpolarized light carries no average intrinsic angular momentum along its propagation direction z . Moreover, it has the property of remaining completely unpolarized even after passing through a birefringent film. One could conclude that no average angular momentum along z can be transferred by completely unpolarized light to a liquid crystal sample. This statement is not strictly correct, however, because as we will show in this work *liquid crystals may be sensitive to higher order correlations of the polarization state of completely unpolarized light*. In fact, due to the presence of higher order correlations in the Stokes vector $\mathbf{s}(t)$, completely unpolarized light may exchange a small, but not zero, angular momentum with matter. The resulting torque, unlike the usual optical torque, is proportional to the square (or higher order power) of the light intensity. In a sense, we may say that the liquid crystal film acts as a sort of Hanbury Brown and Twiss correlator for the light polarization, rather than for the light intensity. As in the Hanbury Brown and Twiss experiment, the response is quadratic in the light intensity I and proportional to the ratio $\varepsilon = \tau_p / \tau_r$ between the correlation time τ_p of the light polarization state and the response time τ_r of the detector that in our case is the liquid crystal.

II. OPTICAL TORQUE OF FULLY UNPOLARIZED LIGHT

The torque equations governing the optical reorientation in nematic liquid crystals have been derived by many authors [4] with special emphasis on the optical Fréedericksz transition (OFT) effect at normal incidence [5,6]. In the OFT geometry, one needs only the torque equation for the polar angle θ of the nematic molecular director $\mathbf{n}(\theta, \phi)$, because no angular momentum along the propagation direction z is transferred from the light to the sample. In the present work we need the torque equations for both polar angles θ and ϕ as well as the equation governing the evolution of light polarization in the propagation through the sample. All of these can be found, for example, in Ref. [7], in the case of normal incidence of the light beam (assimilated to a plane wave), as considered here. For strong homeotropic anchoring and in the small distortion approximation, a Galerkin approximation to these equations can be obtained by assuming a director profile given by $\theta(z, t) = \theta(t) \sin[(\pi z)/L]$ and $\phi(z, t) = \phi(t)$, where L is the sample thickness [9]. The functions of time $\theta(t)$ and $\phi(t)$ are the maximum tilt angle and the average azimuthal angle of the molecular director profile in the sample, respectively. We point out that $\theta(t)$ is assumed to be small, but $\phi(t)$ may be large. We then substitute these expressions into the torque equations and leave out the z dependence by projecting the resulting equations along the lowest spatial modes, after having retained the lowest order terms in the small quantity $\theta(t)$ [8]. The final equations are

$$\frac{d\phi}{d\tau} = \mathbf{s}(\tau) \cdot \mathbf{p}(\phi, \alpha),$$

$$\frac{d\alpha}{d\tau} = f(\alpha) + \mathbf{s}(\tau) \cdot \mathbf{q}(\phi, \alpha), \quad (1)$$

where $\mathbf{s}(\tau) = [s_1(\tau), s_2(\tau), s_3(\tau)]$ is the reduced Stokes vector of the incident wave and

$$\mathbf{p}(\phi, \alpha) = (\tilde{I}/2\alpha)(-\sin 2\phi \sin \alpha, \cos 2\phi \sin \alpha, \cos \alpha),$$

$$\mathbf{q}(\phi, \alpha) = \tilde{I}\alpha(1 - a\alpha)(\cos 2\phi, \sin 2\phi, 0),$$

$$f(\alpha) = (\tilde{I} - 2)\alpha - (a\tilde{I} - b)\alpha^2.$$

In the last equations, $\tilde{I}=I/I_{\text{th}}$ is light intensity normalized to the intensity threshold $I_{\text{th}}=\pi^2ck_{33}/(n_o\mu L^2)$ for OFT and $\tau=t/\tau_r$ is the reduced time, normalized to the sample response time $\tau_r=\gamma_1L^2/(\pi^2k_{33})$, where γ_1 is the orientational viscosity coefficient of the liquid crystal ($\tau_r=2.5$ s for the sample used in our experiment). Backflow effects are neglected. The constants a and b are given by $a=(2/\mu\tilde{L})(1-9\mu/4)$ and $b=(4/\mu\tilde{L})(1-k_{11}/k_{33})$, where k_{11} and k_{33} are the liquid crystal elastic constants for splay and bend, respectively. Moreover, we set $\tilde{L}=2\pi n_oL/\lambda$ and $\mu=1-n_e^2/n_o^2$, where n_e and n_o are the refractive indices for the extraordinary and ordinary wave, respectively, and λ and c are the optical wavelength and speed of light. The quantity α is the nonlinear phase shift accumulated by the optical wave across the sample, related to $\theta(\tau)$ by

$$\alpha=\frac{1}{4}\mu\tilde{L}\theta^2(\tau). \quad (2)$$

It is worth noting that the first of Eqs. (1) may be cast in the form

$$\frac{d\phi}{d\tau}=\left(\frac{\pi^2\tilde{I}}{2\mu\tilde{L}}\right)\Delta s_3, \quad (3)$$

where Δs_3 is the ellipticity change suffered by the beam polarization in passing through the sample. This equation says that the intrinsic angular momentum carried by the optical wave along z is transferred to the sample, producing a rotation of the director \mathbf{n} along the same axis (inertial terms are neglected) [1–3,9]. In the case of fully unpolarized light, the Stokes vector $\mathbf{s}(\tau)$ in Eqs. (1) is rapidly oscillating and averages to zero. In the limit of very fast oscillations, it can be effectively replaced by zero. Therefore, the equilibrium value of α is given by $f(\alpha)=0$, while ϕ is left indeterminate. The steady state solution $\alpha=0$ corresponds to the undistorted sample and it is stable for $\tilde{I}\leq 2$, while the other solution of $f(\alpha)=0$ corresponds to a distorted state that is stable for $\tilde{I}>2$. The intensity threshold needed to induce distortion is twice that for the linear polarization OFT because only one polarization component of the optical field is effective in reorienting the sample. The arbitrariness left in the azimuthal angle ϕ is a consequence of the cylindrical symmetry of the whole system around the z axis. For $\tilde{I}>2$, this symmetry is spontaneously broken and the angle ϕ , the ‘‘Goldstone mode’’ of the symmetry breaking process, is left indeterminate by light. Actually, ϕ is fixed by the small unavoidable residual anisotropy of the sample, due, for example, to imperfect parallelism of the sample walls, surface anchoring small pretilt, gravity effects, etc. Extremely weak external fields may affect the angle ϕ . We stress that perfect cylindrical symmetry $D_{\infty h}$ may occur only for fully unpolarized light. Even in the case of circular polarization, the helicity (left or right) of the incoming beam breaks the inversion symmetry of the system, producing a steady rotation of ϕ [1–3].

So far, the rapid oscillations of light polarization have been completely neglected. We know from mechanics, however, that rapidly oscillating zero average forces may pro-

duce long term nonzero average effects [10]. Similarly, the rapid oscillations of the Stokes vector \mathbf{s} in a fully unpolarized optical wave may also give rise to small terms in Eqs. (1) not averaging to zero. These terms are expected to be proportional to the small ratio $\varepsilon=\tau_p/\tau_r$ and can be calculated by averaging Eqs. (1) over times long with respect to τ_p , but still short with respect to τ_r . The averaging procedure can be carried out as follows. Equations (1) are rewritten in the compact form

$$\frac{dy_i}{d\tau}=f_i(y)+A_{im}(y)s_m(\tau/\varepsilon) \quad (i=1,2), \quad (4)$$

where the rapid rate of change of the Stokes vector components s_m is made explicit by the small constant ε in their argument. Sum over repeated indices is understood. In Eqs. (4), $y_1=\phi$, $y_2=\alpha$, A_{im} is a 2×3 matrix, and the index m runs over the range $m=1,2,3$. We stress that the last term on the right-hand side of Eqs. (4) is not a small one, so that ordinary perturbative methods cannot be exploited. On the other hand, this term is quickly changing in time and averages to zero, so we expect its effects to be small in some sense. We then introduce the new independent variables \bar{y}_i , defined by

$$\bar{y}_i=y_i-\varepsilon A_{im}(y)\xi_m(\tau/\varepsilon) \quad (i=1,2), \quad (5)$$

where $\xi_m(\tau/\varepsilon)$ are three zero-average rapidly varying functions of time to be chosen conveniently. We take ξ_m to be a primitive function of s_m , i.e.,

$$\xi_m(v)=\int^v s_m(u)du+C_m \quad (m=1,2,3), \quad (6)$$

where C_m are integration constants to be chosen so that $\langle \xi_m(t) \rangle=0$. Inserting Eqs. (5) into Eqs. (4) yields

$$\frac{d\bar{y}_i}{d\tau}=f_i(y)-\varepsilon\xi_m\frac{\partial A_{im}}{\partial y_j}(y)f_j(y)-\varepsilon\xi_m s_n\frac{\partial A_{im}}{\partial y_j}(y)A_{jn}(y) \quad (i=1,2), \quad (7)$$

where y in the argument of all functions on the right-hand side must be expressed as a function of \bar{y} , using Eqs. (5). Although fully equivalent to Eqs. (4), Eqs. (7) appear to be more complicated, but have the great advantage that they can be solved perturbatively with respect to the small parameter ε . At the zero order approximation, we get $d\bar{y}_i/d\tau=f_i(\bar{y})$, which is precisely what we expect when the quick terms in Eqs. (4) are replaced by their mean values. It is evident that the function $\bar{y}(\tau)$ varies in time on the slow time scale ($\sim\tau_r$). When only first order terms are retained, Eqs. (7) become

$$\frac{d\bar{y}_i}{d\tau}=f_i(\bar{y})-\varepsilon\xi_m s_n\frac{\partial A_{im}}{\partial y_j}(\bar{y})A_{jn}(\bar{y})+\varepsilon\xi_m\left[\frac{\partial f_i}{\partial y_j}(\bar{y})A_{jm}(\bar{y})-f_j(\bar{y})\frac{\partial A_{im}}{\partial y_j}(\bar{y})\right] \quad (i=1,2). \quad (8)$$

As a final step, we take the time average of Eqs. (8) over times long with respect to the quick time scale ($\sim \tau_p$), but still short with respect to the slow time scale ($\sim \tau_r$). The last term on the right-hand sides of Eqs. (8) averages to zero, because the terms in square brackets are slow functions of time, and the fast functions of time ξ_m average to zero. The surviving terms are

$$\frac{d\bar{y}_i}{d\tau} = f_i(\bar{y}) - \varepsilon \langle \xi_m s_n \rangle \frac{\partial A_{im}}{\partial y_j}(\bar{y}) A_{jn}(\bar{y}) \quad (i=1,2). \quad (9)$$

We see that the first order deviation from the average motion is proportional to the quantities $T_{mn} = \langle \xi_m s_n \rangle$, which, in view of Eqs. (6), are related to the second order correlations of the Stokes vector components. These terms are proportional to the square of the light intensity I . The quantities T_{mn} are independent of the integration constants C_m in Eqs. (6), so we may take $C_m = 0$ with no loss of generality. If the perturbation is carried on to the second order, further terms proportional to $\langle \xi_m \xi_n \rangle$ and to $\langle \xi_m \xi_n s_p \rangle$ appear on the right-hand sides of Eqs. (9). These terms account for the effects due to third order correlations in the light polarization state and are proportional to I^3 . Higher order correlation effects are obtained recursively. Returning to the notation of Eqs. (1), Eqs. (9) read

$$\begin{aligned} \frac{d\bar{\phi}}{d\tau} &= \mathbf{M} \cdot \mathbf{T}, \\ \frac{d\bar{\alpha}}{d\tau} &= f(\bar{\alpha}) + \mathbf{N} \cdot \mathbf{T}, \end{aligned} \quad (10)$$

where matrices \mathbf{M} and \mathbf{N} are given by

$$\mathbf{M} = (\partial \mathbf{p} / \partial \phi) \mathbf{p} + (\partial \mathbf{p} / \partial \alpha) \mathbf{q}, \quad (11)$$

$$\mathbf{N} = (\partial \mathbf{q} / \partial \phi) \mathbf{p} + (\partial \mathbf{q} / \partial \alpha) \mathbf{q}, \quad (12)$$

and we introduced the matrix of correlation times

$$\mathbf{T} = \langle \xi \mathbf{s} \rangle = \left\langle \left(\int_0^\tau \mathbf{s}(\tau') d\tau' \right) \mathbf{s}(\tau) \right\rangle. \quad (13)$$

The matrix \mathbf{T} contains all of the required information on the time behavior of the incoming polarization. As was said before, the average in Eq. (13) is to be made over times long with respect to the characteristic variation time τ_p of the polarization state, but short with respect to the time response τ_r of the liquid crystal. The matrix \mathbf{T} accounts for the time correlations of the light polarization state. It is worth noting that two fully unpolarized light beams, both having coherence matrix $\mathbf{J} = \frac{1}{2} \mathbf{I}_2$, may have different correlation matrices \mathbf{T} . If we assume that the polarization state of the incoming fully unpolarized light changes periodically with period $\tau_p \ll \tau_r$, we get

$$\mathbf{T} = \frac{\varepsilon}{4\pi} \sum_{n=1}^{\infty} [\mathbf{b}_n \mathbf{a}_n - \mathbf{a}_n \mathbf{b}_n], \quad (14)$$

where $\varepsilon = \tau_p / \tau_r$ and \mathbf{a}_n and \mathbf{b}_n are the coefficients of the sine and cosine terms in the Fourier expansion of $\mathbf{s}(\tau)$. A periodic



FIG. 1. Time sequence of polarization states used in the experiment. Arrows indicate the temporal order of the sequence, corresponding to the sign of T .

change of the polarization state of the fully unpolarized light yields a skew-symmetric matrix \mathbf{T} of correlation times [11].

III. EXPERIMENT

In our experiment we used the light beam of an argon laser made artificially unpolarized by means of a Pockels cell. To produce all polarization states two Pockels cells are needed. We used a single Pockels cell, however, yielding polarization periodically changing from linear to elliptical to circular and so on, as depicted in Fig. 1.

In a reference frame having the z axis along the beam propagation direction and the x axis along the fast axis of the Pockels cell, the Stokes vector of the beam exiting the Pockels cell is $\mathbf{s}(t) = [0, \pm \sin(2\pi t/\tau_p), \cos(2\pi t/\tau_p)]$, where t is time and τ_p the period. We have $\langle \mathbf{s}(t) \rangle = 0$, so that the light is fully unpolarized. Inserting the Stokes vector $\mathbf{s}(t)$ into Eq. (13) and performing the appropriate time average, we find that all elements of the matrix \mathbf{T} are zero, except $T_{23} = -T_{32} = \pm \varepsilon/4\pi$. The double sign refers to the two possible scanning directions over the polarization states (see Fig. 1). In this case, Eqs. (10) assume the explicit form (bars are omitted hereafter for simplicity)

$$\begin{aligned} \frac{d\phi}{d\tau} &= \pm \frac{\varepsilon \bar{I}^2}{8\pi\alpha^2} g(\alpha) \sin 2\phi, \\ \frac{d\alpha}{d\tau} &= f(\alpha) \pm \frac{\varepsilon \bar{I}^2}{4\pi} h(\alpha) \cos 2\phi, \end{aligned} \quad (15)$$

where $g(\alpha) = \alpha(1 - a\alpha)(1 - \cos\alpha - a\sin\alpha) - \sin\alpha(1 - \cos\alpha)$ and $h(\alpha) = (1 - a\alpha)(1 - \cos\alpha)$. For $\bar{I} > 2$ the trivial solution $\alpha = 0$ becomes unstable and other steady-state solutions appear at $\phi = 0, 180^\circ$ and $\phi = 90^\circ, 270^\circ$. Above the reorientation threshold, the solutions corresponding either to the first or to the second pair of ϕ values are stable, while the other two are unstable, depending on the sign of $\pm g(\alpha)$. For a given beam intensity \bar{I} , the value of α [and hence of $g(\alpha)$] in the stable steady states is the same and it is given by the equation $f(\alpha) \pm \varepsilon \bar{I}^2 h(\alpha)/4\pi = 0$, where the double sign corresponds to $\mp g(\alpha) > 0$. The stability of the states $\phi = 0, 180^\circ$ and $\phi = 90^\circ, 270^\circ$ is reversed by changing the sign in front of ε . We expect, therefore, that a sudden change in the sign of $s_2(\tau)$ in the input light polarization (i.e., in the time sequence of the polarization states) will produce a 90° azimuthal rotation of the molecular director equilibrium state. The orientation switching effect is well suited to detecting the optical correlation torque because it cannot be ascribed to other spurious effects such as laser induced thermal indexing. During switching, the phase angle α is expected to vary until, in the final steady state, it resumes its initial value [14].

In the limit $\varepsilon \rightarrow 0$ the effect of the polarization correlations becomes negligible and the angle ϕ is left indefinite, as expected for the Goldstone mode. Nevertheless, as discussed above, small sample imperfections break the cylindrical symmetry so that, in the limit $\varepsilon \rightarrow 0$, the molecular director is oriented to some angle ϕ_0 . The value of ϕ_0 is unpredictable and may even vary from point to point in the sample. In order to account for these effects, we added a phenomenological term of the form $-A \sin(\phi - \phi_0)$ to the right of the first of Eqs. (15), with the understanding that A is a very small quantity. Then, the final equations of the model read

$$\frac{d\phi}{d\tau} = \pm \frac{\varepsilon \bar{I}^2}{8\pi\alpha^2} g(\alpha) \sin 2\phi - A \sin(\phi - \phi_0),$$

$$\frac{d\alpha}{d\tau} = f(\alpha) \pm \frac{\varepsilon \bar{I}^2}{4\pi} h(\alpha) \cos 2\phi, \quad (16)$$

with constant A and ϕ_0 . Due to the presence of the last term in the first equation, the steady state values of ϕ are no longer multiples of $\pi/2$ and are obtained as solutions of the transcendental equation

$$\sin 2\phi = \pm X \sin(\phi - \phi_0) \quad (17)$$

with

$$X = \frac{8\pi A \alpha^2}{\varepsilon \bar{I}^2 g(\alpha)}. \quad (18)$$

For fixed intensity \bar{I} and phase angle α , the steady value of the angle ϕ depends on the time ratio ε and on the constants A and ϕ_0 characterizing the residual anisotropy torque. The switching effect is still present, but the rotation $\Delta\phi$ is lower than 90° and it depends on ε as well.

Experiments were carried out by focusing an argon laser beam onto a $50 \mu\text{m}$ thick, homeotropically aligned nematic liquid crystal cell, as in usual OFT measurements. The beam profile at the sample position was measured by recording the light passing through a $10 \mu\text{m}$ pinhole moved across the beam cross section. The profile was found Gaussian with a $63 \mu\text{m}$ beam waist. We also checked that the waist was the same in both the x and y directions, within the experimental errors. The material was the nematic liquid crystal mixture E7 from Merck. The laser beam passed through a Pockels cell driven by an appropriate sawtooth signal. The amplitude of the driving signal was adjusted to obtain full depolarization of the laser light. A proper calibration of the signal driving the Pockels cell is critical, because even very small residual elliptical polarization may lead to appreciable effects [12]. We measured the polarization of the beam beyond the Pockels cells by using a standard technique [13]. The average reduced Stokes parameters of the depolarized beam used in the experiment were measured to be $|\langle s_i \rangle| < 0.01$ ($i = 1, 2, 3$) in the full range of driving frequencies (from 0.1 to 100 Hz). In the power range used in the experiment (up to 400 mW), we estimate a laser induced heating below 1° .

The threshold intensity for reorientation was measured, as usual, by counting the number of rings in the far field self-diffraction pattern beyond the sample as a function of inten-

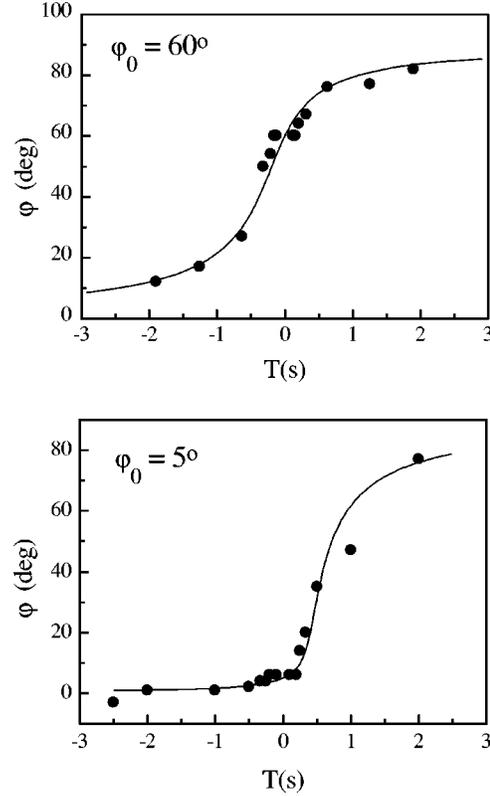


FIG. 2. Measured steady state azimuthal angle ϕ as a function of polarization correlation time T for two points in the sample. Laser intensity was fixed to $\bar{I} = 2.2$. The solid line is a best fit from theory, as described in the text.

sity, and then extrapolating to zero rings. The threshold for the ordinary OFT was found in the same way by using a linearly polarized input beam. A ratio very close to two was found, in agreement with theory. Above threshold, a steady state is reached through a second order transition. The steady-state reorientation plane ϕ with respect to the fast axis of the Pockels cell was measured by looking at the polarization of the outer ring pattern. Only the extraordinary component of the wave is scattered in the far field rings, so that they are approximately linearly polarized in the plane containing the liquid crystal molecules [15]. We measured the steady state angle ϕ as a function of the time $T = \pm \tau_p = \pm \varepsilon \tau_r = 4\pi T_{23} \tau_r$, for fixed intensity \bar{I} . Examples of results, obtained in two different points of a sample, are reported in Fig. 2. The sign of T corresponds to the direction of the time sequence of polarization states in the incident beam. The switching effect of ϕ is evident in both cases, even if the values of ϕ_0 are quite different. During the ϕ -switching process, the number of rings in the far field and the optical phase shift α changed dramatically, until, in the final state, they resumed the same value as before switching, as predicted by theory. The data were fitted using Eq. (17) with ϕ_0 and X as adjustable parameters. From the best fit value of X the strength A of the residual anisotropy torque can be deduced, once the other quantities in Eq. (18) are given.

Tabulated values for other material constants have been used. From our data, only the order of magnitude of A can be estimated, because the phase retardation α is known from the ring number with an uncertainty of about π and the function

$g(\alpha)$ changes appreciably over π . We found that $A \approx 10^{-3}$, corresponding to a residual volume torque of about 10^{-4} dyn/cm² (as for an external magnetic field of about 50 G) or to an azimuthal surface anchoring energy of about 10^{-7} erg/cm² (about five orders of magnitude smaller than typical azimuthal anchoring energy of a homogeneously aligned liquid crystal). Other samples, prepared with ITO conductive coating on the walls show, instead, a much stronger residual anisotropy, such that we were unable to see any rotation effect at all. This observation points to surface effects as the main source of residual anisotropy. We made our liquid crystal cells with standard surface coating techniques and we took no care in aligning the sample walls to minimize residual anisotropy. By exploiting suitable techniques, cells having higher azimuthal symmetry could be realized, lowering the residual torque and thus increasing the sensitivity of the Goldstone mode. A final point is worth noting. As shown in Fig. 2, the correlation times used in our experiments ranged from $-2 \text{ s} \leq T \leq 2 \text{ s}$. Although comparable to the intrinsic response time τ_r of the material, these correlation times are still short enough with respect to the effective response time of the θ and ϕ angles, due to the critical slowing down occurring near the OFT threshold. In fact, from Eqs. (1) we see that, for small distortions, the effective response times of ϕ and θ angles are $\tau_\phi \approx \tau_r \alpha / \tilde{I} \approx 30 \text{ s}$ and $\tau_\theta \approx \tau_r (\tilde{I} - 2) \approx 10 \text{ s}$, for $\tilde{I} \approx 2.2$ and $\alpha \approx 10\pi$, which are typical values of our experiment.

IV. CONCLUSIONS

We have shown that liquid crystals can be sensitive to higher order time correlations in the polarization of com-

pletely unpolarized light. Thus, at least in principle, liquid crystals can distinguish among different kinds of unpolarized optical fields. A convenient averaging technique was applied to the torque equations in the nematic and a set of coupled differential equations was derived [Eqs. (16)] where optical driving terms proportional to second order polarization correlations are present. An interesting consequence of these equations is that switching of the time sequence of polarization states leads to stability exchange between two equilibrium values of the molecular director azimuthal angle ϕ . The switching effect was indeed observed experimentally. In our experimental geometry, this angle is the rotational ‘‘Goldstone mode’’ of the system and it may be sensitive to very small external torques. Any small broken symmetry involved in the sample geometry would influence the ϕ angle, so that this sensitivity could be exploited, for example, in testing the quality of liquid crystal cells. Our observations seem to suggest that anchoring conditions can influence the switching effect dramatically, but further work in this direction is needed. We were able to estimate the broken-symmetry residual torque in our samples. As expected, it was found to be very weak, more than five orders of magnitude weaker than the usual anchoring surface torques in homogeneously aligned samples.

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- [1] E. Santamato, B. Daino, M. Romagnoli, M. Settembre, and Y. R. Shen, *Phys. Rev. Lett.* **57**, 2423 (1986).
 - [2] E. Santamato, M. Romagnoli, M. Settembre, B. Daino, and Y. R. Shen, *Phys. Rev. Lett.* **61**, 113 (1988).
 - [3] L. Marrucci, G. Abbate, S. Ferraiuolo, P. Maddalena, and E. Santamato, *Phys. Rev. A* **47**, 4859 (1992).
 - [4] For a recent review, see, e.g., E. Santamato and Y. R. Shen, in *A Guide to Liquid Crystal Research*, edited by P. J. Collings and J. S. Patel (Oxford University Press, New York, 1997).
 - [5] A. S. Zolot'ko, V. F. Kitaeva, N. Kroo, N. N. Sobolev, and L. Csillag, *Pis'ma Zh. Eksp. Teor. Fiz.* **32**, 170 (1980) [*JETP Lett.* **32**, 158 (1980)].
 - [6] S. D. Durbin, S. M. Arakelian, and Y. R. Shen, *Phys. Rev. Lett.* **47**, 1411 (1981).
 - [7] E. Santamato, G. Abbate, and P. Maddalena, *Phys. Rev. A* **38**, 4323 (1988).
 - [8] In the θ -equation the term proportional to θ_1^3 is left to allow for saturation at high light intensity I .
 - [9] G. Abbate, P. Maddalena, L. Marrucci, L. Saetta, and E. Santamato, *J. Phys. II* **1**, 543 (1991).
 - [10] L. D. Landau and E. M. Lifshitz, *Mechanics* (Pergamon, London, 1959), Sec. 30.
 - [11] It can easily be shown that for a random walk on the Poincaré sphere spanned by $\mathbf{s}(\tau)$, the matrix \mathbf{T} is diagonal and, hence, symmetric.
 - [12] E. Santamato, G. Abbate, P. Maddalena, L. Marrucci, and Y. R. Shen, *Phys. Rev. Lett.* **64**, 1377 (1990).
 - [13] R. M. A. Azzam and N. M. Bashara, *Ellipsometry and Polarized Light* (North-Holland, New York, 1977).
 - [14] This change of α produces a visible change in the number of diffraction rings in the far field during switching. Other steady states exist corresponding to $g(\alpha) = 0$ and ϕ obtained from the second of Eqs. (15). These states have never been observed in our experiment.
 - [15] B. Ya. Zel'dovich and N. V. Tabiryan, *Sov. Phys. JETP* **55**, 656 (1982).