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OPTICAL FRÉEDERICKSZ TRANSITION INDUCED BY DEPOLARIZED LIGHT

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Abstract The optical Fréedericksz effect induced by depolarized laser light in a nematic liquid crystal is investigated both theoretically and experimentally. By "depolarized" is here intended light having a rapidly varying polarization whose density matrix is isotropic. The main features of the phenomenon are determined only by the average properties of the light. However, when the light polarization dynamics is about one order of magnitude faster than the response time of the liquid crystal, the system becomes very sensitive to the temporal correlations of light polarization, i.e., to the fourth-order time correlations of the optical field. This prediction is confirmed by the experimental results.

INTRODUCTION

When a sufficiently intense laser beam crosses a nematic liquid crystal (NLC) a phenomenon of collective molecular reorientation is induced known as optical Fréedericksz transition (OFT).[1] In many respects, this phenomenon is similar to the reorientation induced by static electric fields, which is the basic working mechanism of all NLC displays. Namely, the liquid crystal molecular director **n**, initially oriented in a direction imposed by boundary conditions only, reorients to a new stationary state with a direction given by the competition between boundary conditions and the wave electric field. However, a closer look reveals large differences between the optical and electrostatic effects, mostly due to the role played by light propagation and polarization. Circularly or elliptically polarized light, for instance, carries angular momentum directed along the propagation direction, and this angular momentum can be efficiently exchanged with the liquid crystal, thus inducing a continuous molecular precession or other complex dynamic effects.[2–5]

Fully depolarized light has a larger rotational symmetry than polarized light and it carries no angular momentum associated with polarization. Therefore, it can be expected that interaction of a NLC with a depolarized laser beam give rise to a different behavior from that observed with any kind of polarized light.

Our study was restricted to a NLC film with strong homeotropic alignment (i.e., the boundary conditions constrain the director $\mathbf{n}$ to be perpendicular to the film walls), and to light impinging at normal incidence. In our analysis we approximate the wave to a plane wave. After the reorientation takes place, the two polar angles in the coordinate system having the z-axis parallel to the beam propagation direction, i.e., the tilt ϑ and the azimuthal angle φ, are used to describe the orientation of $\mathbf{n}$. These angles are, in general, functions of the spatial coordinate z and time t.

DEPOLARIZED LIGHT

Depolarized light is actually light with a polarization varying rapidly with respect to the NLC response time τ_{LC} and satisfying the following conditions on the Jones' matrix (which is the polarization density matrix):

$$J_{ij} = \langle E_i E_j^* \rangle = |E|^2 \delta_{ij}, \tag{1}$$

where $i, j = x, y$, and the angular brackets denote time average. In other words we require that the light Jones' matrix be isotropic for rotations around the z-axis. Equivalent to Jones' matrix is the description based on the three reduced Stokes' parameters $s_1 = (|E_x|^2 - |E_y|^2)/S_0$, $s_2 = 2\Re\mathrm{e}(E_x E_y^*)/S_0$, and $s_3 = 2\Im\mathrm{m}(E_x E_y^*)/S_0$, with $S_0 = |E_x|^2 + |E_y|^2$. Then Eq. (1) can be rewritten as $\langle s_1 \rangle = \langle s_2 \rangle = \langle s_3 \rangle = 0$.

Most incoherent sources generate "unpolarized" light, which has a random polarization dynamics fulfilling conditions (1). The coherence time of the polarization dynamics is in such cases the same as that of the phase and amplitude change. On the other hand, laser light is usually polarized, and it can be made artificially "depolarized" by means of one or more electro-optic or mechanical devices. The polarization dynamics, being electronically driven, is in these cases much slower than the coherence time of the light, and it can be either random or, more frequently, periodic. For instance, by means of a single electro-optic cell (e.g., a Pockels' cell) driven by a saw-tooth voltage function, the following polarization dynamics, satisfying Eq. (1), can be easily produced:

$$\begin{aligned} s_1(t) &= 0, \\ s_2(t) &= \sin(2\pi t/T), \\ s_3(t) &= \cos(2\pi t/T), \end{aligned} \tag{2}$$

where T is the oscillation period. Equally easy to generate with a single electro-optic cell is any other polarization dynamics obtained from this by exchanging the indices 1, 2, and 3, or by mixing directions 1 and 2 by means of a z-axis rotation.

NEGLECTING FLUCTUATIONS

If the effect of fast polarization fluctuations can be neglected, then the action of light is completely determined by its average properties, described for instance by Jones' matrix J_{ij} or equivalently by the average Stokes' parameters $\langle s_i \rangle$. Then, the following simple reasoning is sufficient to predict most of the essential features of the resulting OFT.

The Jones' matrix of any depolarized light, given by Eq. (1), is the same as that of an incoherent superposition of two waves linearly polarized along arbitrary orthogonal directions, each one having half of the total intensity. Suppose now that the director $\mathbf{n}$ fluctuates out of the initial z direction, in a plane specified by the angle φ. We loose no generality by taking one of the two linearly polarized waves as oscillating in the φ-plane, and the other one in the orthogonal plane. Notice that the birefringence associated with the director reorientation does not affect the Jones' matrix of the light propagating inside the sample. The wave polarized orthogonally to the director generates no optical torque. The wave polarized in the φ-plane, instead, will produce the same torque as a single linearly polarized wave having half of the total intensity. Therefore, the OFT reorientation is identical to that induced by a single linearly polarized wave with half of the intensity, polarized along an arbitrary direction (at least as long as the field $\mathbf{n}(z)$ is contained in a single plane). In particular, the OFT threshold intensity for depolarized light will be twice that of linearly polarized light.*

The arbitrariness of the reorientation direction φ is of course a consequence of the cylindrical rotational symmetry of the system, which is possessed both by light and by the homeotropic boundary conditions. By reorienting, the system spontaneously breaks such symmetry. The angle φ, i.e., the "Goldstone" mode of the symmetry breaking process, is therefore left undetermined by light. Ideally it should be chosen at random from the initial fluctuations. Actually, it is usually fixed by the small residual anisotropy of the sample. Extremely weak external fields, moreover, should be capable of controlling and changing the angle φ.

In the above reasoning, we assumed that light polarization fluctuations do not play any role. But is this assumption correct? Examples from other fields of physics indicate that sometimes fast fluctuations of the external force, even if very small in amplitude and with zero average, may produce a significant and rather unexpected effect. For instance, in classical mechanics it is known that a pendulum with a rigid wire subject to fast fluctuations of the suspension point feels, besides gravity, an additional "effective" potential due to the fluctuations.[6] This potential may even change the upside down vertical orientation of the pendulum from unstable to stable, when the fluctuations are fast enough and are along a vertical direction. It is therefore necessary to perform a deeper theoretical analysis to understand what is the effect of light polarization fluctuations in our case.

THEORY

The dynamical equations describing the reorientation of the director $\mathbf{n}$ in the plane wave and small birefringence approximations can be found for instance in

*This is like in the case of circularly polarized light. The latter actually differs in the fact that light polarization inside the sample is affected by the birefringence. However, the medium birefringence is, for small ϑ, proportional to ϑ^2, and therefore it does not enter the linearized equations which determine the threshold intensity.

Ref. 4. In terms of the polar coordinates ϑ and φ, they are

$$\begin{aligned} \dot{\vartheta} &= (1 - p_1 \sin^2 \vartheta)\, \vartheta'' - \frac{p_1}{2} \sin 2\vartheta\, \vartheta'^2 - \left(\frac{1}{2} - p_2 \sin^2 \vartheta\right) \sin 2\vartheta\, \varphi'^2 \\ &\quad + \frac{\tilde{I}}{2} \sin 2\vartheta\, (1 + s_1 \cos 2\varphi + s_2 \sin 2\varphi), \end{aligned} \tag{3}$$

$$\dot{\varphi} = (1 - p_2 \sin^2 \vartheta)\varphi'' + 2(\cot \vartheta - p_2 \sin 2\vartheta)\, \varphi'\vartheta' + \tilde{I}(s_2 \cos 2\varphi - s_1 \sin 2\varphi), \tag{4}$$

with the boundary conditions for strong homeotropic anchoring

$$\vartheta(z=0) = \vartheta(z=1) = \varphi'(z=0) = \varphi'(z=1) = 0. \tag{5}$$

With a prime we denote derivative with respect to the dimensionless space coordinate $\tilde{z} = z/L$, where L is the film thickness; a dot denotes derivative with respect to dimensionless time $\tilde{t} = t/\tau_{LC}$, where $\tau_{LC} = \gamma_1 L^2/\pi^2 K_{33}$ is the response time of the liquid crystal, γ_1 being an orientational viscosity and K_{33} the bend elastic constant; the constants p_1 and p_2 are given by $p_1 = 1 - K_{11}/K_{33}$ and $p_2 = 1 - K_{22}/K_{33}$, where K_{11} and K_{22} are the splay and twist elastic constants, respectively; $\tilde{I} = n_o(n_e^2 - n_o^2)L^2 I/2\pi^2 c n_e^2 K_{33}$ is light intensity I normalized to OFT threshold, where n_o and n_e are the ordinary and extraordinary refractive indices, respectively, and c is the speed of light. In the following, whenever it will not lead to confusion, we will omit tildes on the dimensionless coordinates z and t.

The equations for light propagation in the film, within the geometric optics approximation (GOA), can be written as

$$\boldsymbol{s}' = \frac{2\pi L}{\lambda}(\bar{n} - n_o)\, \boldsymbol{\omega} \times \boldsymbol{s}, \tag{6}$$

where $\boldsymbol{\omega} = (\cos 2\varphi, \sin 2\varphi, 0)$ and $\bar{n}$ is the effective extraordinary refractive index. We are assuming that the effective birefringence is small, i.e., $\bar{n} - n_o \ll n_o$. If this assumption is valid, then $\bar{n} \approx n_o[1 + (n_e^2 - n_o^2)\sin^2\vartheta/2n_e^2]$.

If we neglect fluctuations, we can substitute for the Stokes' parameters s_i their average values, which are all zero. In Eq. (4) for φ the optical torque term then vanishes. The function $\varphi(z,t) =$ constant is then a trivial solution, stable when ϑ is not too large. This means that the reoriented field $\mathbf{n}(z)$ will indeed be contained in a plane, at least for small deformations. Eq. (3) for ϑ, after the solution $\varphi =$ const.

is inserted in, becomes identical to the equation for the OFT induced by a single linearly polarized wave, except for a factor 2 missing in the optical torque. This confirms the threshold intensity doubling, as already anticipated above. All results obtained on the OFT for linearly polarized light apply also to the depolarized light case, except for the fact that the plane of reorientation is not determined by light.

Now we move on, to include the effect of fluctuations. We are only interested in the "average" effect of fluctuations, i.e., in the additional effective torque generated by fluctuations and acting on **n**. We followed Kapitsa's method,[6] whose basic idea is to split the time dependence of the system variables into a slow term and a fast zero-average oscillating term, which is supposed to be small:

$$\begin{aligned} \vartheta(z,t) &= \langle \vartheta(z,t) \rangle + \delta\vartheta(z,t), \\ \varphi(z,t) &= \langle \varphi(z,t) \rangle + \delta\varphi(z,t). \end{aligned} \tag{7}$$

Separate equations for the slow and fast terms can then be written down, the fast part can be solved in a first-order approximation, and the result back-substituted into the slow part. The outcome is the appearance of an effective torque arising from the oscillations.

Moreover, to reduce Eqs. (3)-(5) to a simpler set of integer differential equations, we used Galerkin's approach. The spatial dependence of **n** is written as a Fourier-mode expansion, and truncated to the first nonzero term:

$$\begin{aligned} \langle \vartheta(z,t) \rangle &\simeq \vartheta_1(t) \sin\left(\frac{\pi z}{L}\right), \\ \langle \varphi(z,t) \rangle &\simeq \varphi_1(t). \end{aligned} \tag{8}$$

After neglecting all powers of ϑ_1 larger than ϑ_1^3, partial differential equations (3)-(4) are projected onto such space-modes to obtain a set of integer differential equations for the time evolution of ϑ_1 and φ_1 only.

For brevity, we skip all intermediate steps of the outlined calculations and write down directly the final dynamical equations:[7]

$$\begin{aligned} \dot{\vartheta}_1 &= (\tilde{I}-1)\vartheta_1 - \frac{1}{2}(\tilde{I}-p_1)\vartheta_1^3 + \left[\tilde{I}^2\vartheta_1 A_{hk}\tau_{kh}\right], \\ \dot{\varphi}_1 &= \left[\tilde{I}^2 B_{hk}\tau_{kh}\right]. \end{aligned} \tag{9}$$

In these equations, the matrices A_{hk} and B_{hk} are functions of ϑ_1 and φ_1, and are given by

$$A_{hk} = \begin{pmatrix} 1+\sin^2 2\varphi \,\frac{\sin 2\alpha}{2\alpha} & -\sin 4\varphi \,\frac{\sin 2\alpha}{4\alpha} & \sin 2\varphi \,\frac{1-\cos 2\alpha}{2\alpha} \\ -\sin 4\varphi \,\frac{\sin 2\alpha}{4\alpha} & 1+\cos^2 2\varphi \,\frac{\sin 2\alpha}{2\alpha} & -\cos 2\varphi \,\frac{1-\cos 2\alpha}{2\alpha} \\ \sin 2\varphi \,\frac{1-\cos 2\alpha}{2\alpha} & -\cos 2\varphi \,\frac{1-\cos 2\alpha}{2\alpha} & 1-\frac{\sin 2\alpha}{2\alpha} \end{pmatrix}, \tag{10}$$

$$B_{hk} = \begin{pmatrix} \sin 4\varphi \,\frac{\sin\alpha}{\alpha} & -2\cos^2 2\varphi \,\frac{\sin\alpha}{\alpha} & 2\cos 2\varphi \,\frac{1-\cos\alpha}{\alpha} \\ 2\sin^2 2\varphi \,\frac{\sin\alpha}{\alpha} & -\sin 4\varphi \,\frac{\sin\alpha}{\alpha} & 2\sin 2\varphi \,\frac{1-\cos\alpha}{\alpha} \\ 0 & 0 & 0 \end{pmatrix}. \tag{11}$$

where $\alpha = (2\pi L/\lambda)\int_0^1(\bar{n} - n_o)dz$ is the optical phase-shift due to the sample birefringence ($\alpha \propto \vartheta_1^2$). The τ_{kh}'s, instead, are constants characterizing the polarization dynamics, and are defined as

$$\tau_{kh} = \langle s_h^0(t) \int^t s_k^0(t')dt' \rangle, \tag{12}$$

where the $s_i^0(t) = s_i(0,t)$ are the Stokes' parameters taken at the input face of the nematic film. In Eqs. (9), the terms between square brackets are the components of the effective torque arising from polarization fluctuations. It should be noticed that they are quadratic in the light intensity $\tilde{I}$ and are proportional to the polarization characteristic times τ_{kh}. They tend to vanish as the polarization change is made faster while the light intensity is kept constant. Therefore, if the polarization dynamics is fast enough, the system should behave as described previously under the assumption of no effect of light fluctuations. On the other hand, if the polarization dynamics is not too fast, but still much faster than the liquid crystal response time, the system could become sensitive to the polarization coherence times τ_{kh}, i.e., to the optical electric field fourth-order correlations. In particular, the Goldstone mode φ, if the residual anisotropy is neglected, is affected only by the fluctuation effective torque and should be especially sensitive to its effect.

Equations (9) describe the director dynamics under the action of depolarized light for every possible polarization dynamics. Different behaviors of the system may be expected depending on the specific depolarization procedure. Here we consider only one set of polarization dynamics, i.e., those given by Eqs. (2) at varying oscillation period T. There are two reasons for such choice: first, as we said, it is a polarization dynamics which can be easily generated by means of a single Pockels cell; second, the effect on the Goldstone mode was predicted to be especially strong and therefore easier to reveal. A more complete analysis of all possible cases will be presented elsewhere.[7]

Equations (12) show that for a polarization dynamics given by Eqs. (2) the only nonzero τ_{hk}'s are

$$\tau_{32} = -\tau_{23} = \frac{T}{4\pi\tau_{LC}}, \tag{13}$$

Equations (9) are then reduced to the following:

$$\dot{\vartheta}_1 = (\tilde{I} - 1)\vartheta_1 - \frac{1}{2}(\tilde{I} - p_1)\vartheta_1^3, \tag{14}$$

$$\dot{\varphi}_1 = \frac{\tilde{I}^2}{2\pi}\left(\frac{1 - \cos\alpha}{\alpha}\right)\left(\frac{T}{\tau_{LC}}\right)\sin 2\varphi_1. \tag{15}$$

Equation (14) for ϑ is now independent of the fluctuation effective torque. It describes the usual OFT splay-bend behavior, with a threshold intensity at $\tilde{I} = 1$, above which the solution $\vartheta_1 = 0$ becomes unstable, and a new intensity-dependent, stationary solution $\vartheta_1 = \sqrt{2(\tilde{I} - 1)/(\tilde{I} - p_1)}$ appears.

On the contrary, Eq. (15) shows that the angle φ_1 is, as anticipated, determined only by the fluctuation-induced torque. By substituting $\dot{\varphi}_1 = 0$ into Eq. (15), we find that four stationary solutions are possible, respectively two stable and two unstable: $\varphi_1 = 0°$ and $180°$ (stable if $T > 0$, unstable if $T < 0$), and $90°$ and $270°$ (unstable if $T < 0$, stable if $T > 0$). Depending on the sign of T, i.e., on the sole direction of the temporal sequence of polarizations which the light undergoes, the stability of these four states is interchanged. Therefore the angle φ_1 should switch by $90°$ when the polarization dynamics is temporally inverted. The sample birefringence slow-axis, which is determined by φ_1, also switches, producing large optical effects. Notice that the rotational symmetry is now broken by light, since the specific polarization dynamics chosen is not rotationally symmetric.

Ideally the switching should take place no matter how fast is the polarization dynamics. Actually, however, one must take into account residual anisotropy, which tries to fix φ_1 to some constant value φ_0. If the effect of light fluctuations is stronger than residual anisotropy the switching will occur, although somewhat reduced in its amplitude. But increasing the speed of polarization variation, the effective torque due to light fluctuations decreases, and the amplitude of the switching will be smaller and smaller and finally disappear. To model such competition between the fluctuation effect and the residual-anisotropy effect, we added to Eq. (15) the term $A\sin(\varphi_0 - \varphi_1)$, which can be considered a first term of a Fourier-mode expansion of a generic periodic function of φ.

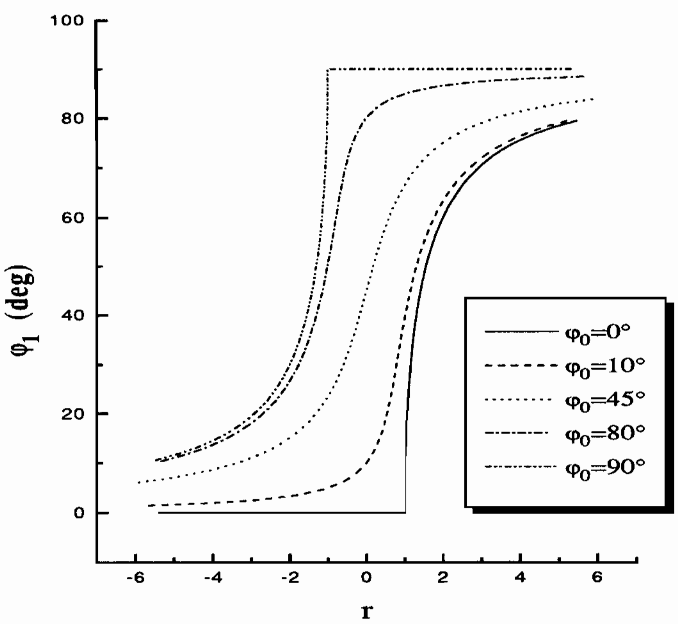

FIGURE 1 Stable equilibrium value of the azimuthal angle φ_1 of **n** for various values of the ratio r between the effective torque due to light fluctuations and the torque due to residual anisotropy.

Figure 1 describes the predicted value of stable equilibrium for φ_1 at varying light to residual anisotropy torque ratio, and for different φ_0's. These values are obtained by numerically solving the transcendent equation obtained from Eq. (15) by taking $\dot{\varphi}_1 = 0$.*

EXPERIMENT

The set-up of our experiment is extremely simple, and is illustrated in Fig. 2. Briefly, a function generator drives a Pockels' cell via an amplifier, to get de-

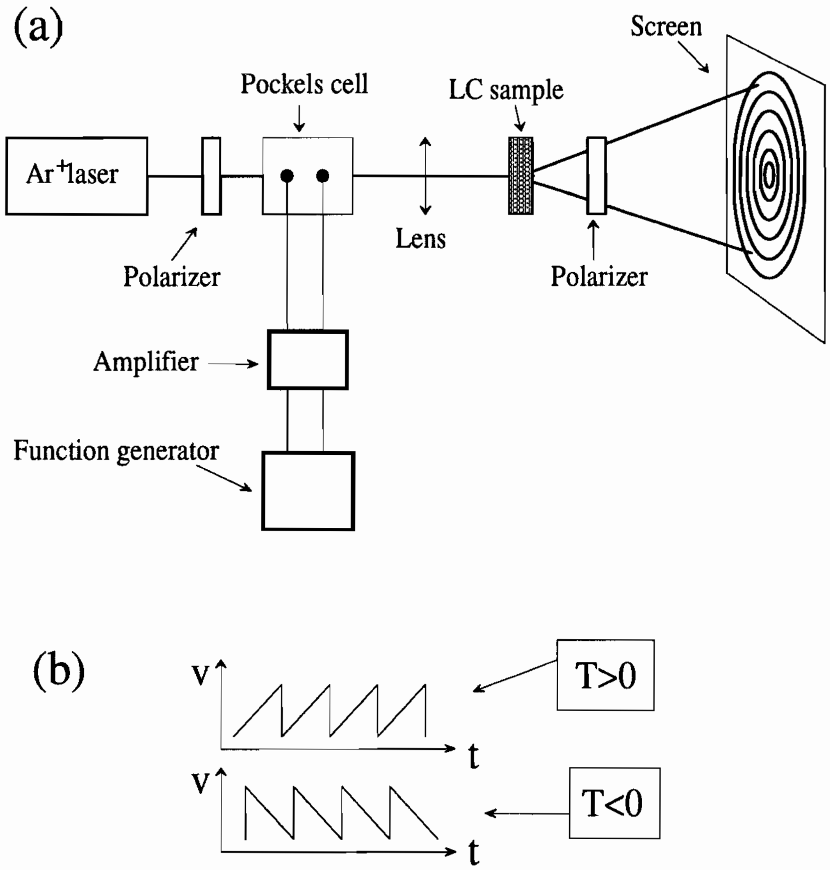

FIGURE 2 (a) - Experimental set-up. (b) - Saw-tooth signals utilized to control the Pockels cell for the two opposite values of the period T.

*Only in the special cases $\varphi_0 = 0, 90°, 180°$, or $270°$, an analytical solution can be also found.

polarized light out of a linearly polarized input beam generated by a cw Argon laser. The beam is then focused at normal incidence onto the sample. The OFT threshold intensity observed with depolarized light when the fluctuation period T was sufficiently short (less than 0.1 seconds) resulted to be twice larger than that observed with linearly polarized light, within $\pm$ 10%, in good agreement with the predictions. Above the threshold, the well known self-diffraction ring pattern is observed in the far-field domain. The diffraction rings are polarized almost uniformly (although with not a very large extinction ratio). Their polarization plane is just the same as the slow-axis birefringence plane, given by the angle φ_1. By analyzing the diffracted light with a polarizer the value of φ_1 was thus easily obtained.

The utilized NLC was the BDH mixture E7, at room temperature. The cell thickness was about 50 μm. The time constant τ_{LC} for such a sample is approximately 2.5 s. However, it must be considered that the response time for the two angles ϑ_1 and φ_1 is actually larger. A detailed analysis shows that two different response times can be defined for the two angles, which are $\tau_\vartheta = \tau_{LC}/2(\tilde{I}-1)$ (notice the "critical slowing-down" factor $(\tilde{I}-1)^{-1}$) and $\tau_\varphi = \tau_{LC}\alpha/\tilde{I}$. Our experiment was performed at a reduced intensity $\tilde{I} \simeq 1.1$, which gave rise to a birefringence phase-shift $\alpha \simeq 8\pi$ (i.e., about 4 rings in the diffraction pattern). Therefore we had $\tau_\vartheta \approx 10$ s and $\tau_\varphi \approx 60$ s.

Figure 3 shows the results of two experiments performed in two different spots of the same sample. Similar results were obtained with other samples. The solid line is the theoretical curve, after a best fit on the strength A and direction φ_0 of the residual anisotropy. As it can be seen, the agreement is good. The angle φ_1 shows a strong dependence on the specific polarization dynamics, although the "average" polarization state, i.e., Jones' matrix is always the same. What is the most impressive, two polarization dynamics characterized by the values T and $-T$, i.e., which differ for the polarization-sequence direction only, give rise to a large switching of the angle φ. The agreement is good for T as large as 2.5 seconds, which is only 5 times smaller than the response time of ϑ. For larger T's, we observed poor reproducibility of the experimental results. On the other

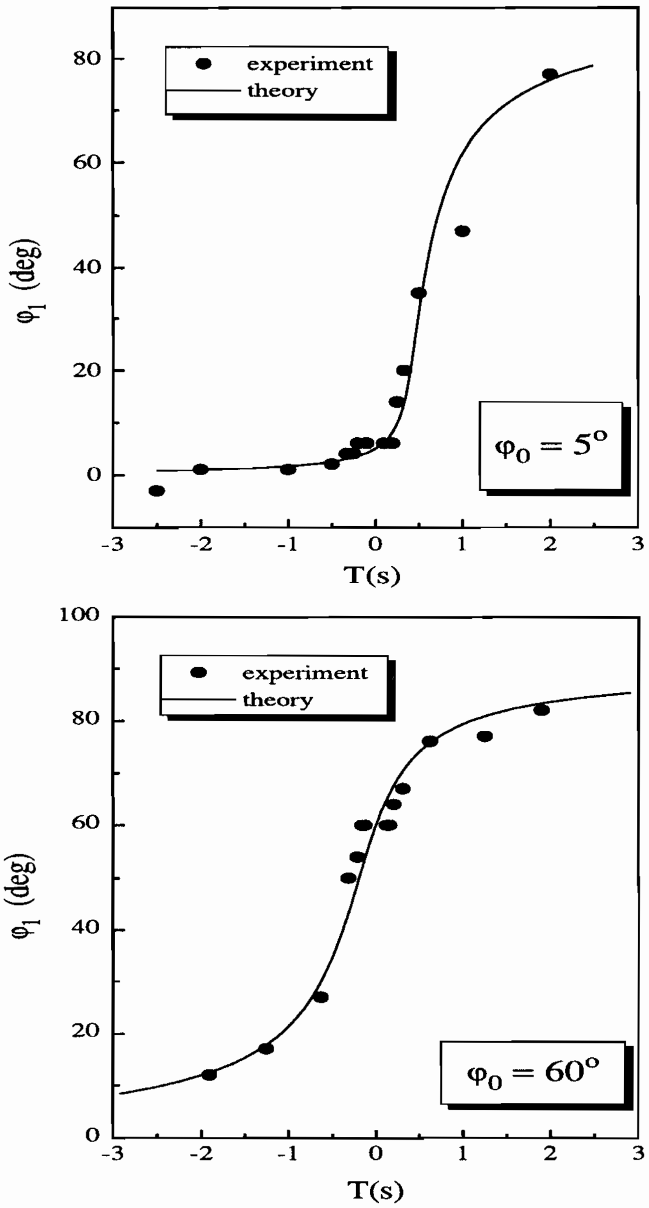

FIGURE 3 Two examples of measured (circles) and theoretical (solid line) values of φ_1 at fixed light intensity and varying the depolarization time T. The theoretical curve is after a best-fit on the strength A and direction φ_0 of the residual anisotropy. Different values of A and φ_0 were obtained even for different spots of the same sample.

limit, fluctuation effects were found significant only if T is larger than about 0.2 s, below which residual anisotropy dominated. From our fits we found that the order of magnitude of A is 10^{-3}. We do not know for sure if the residual anisotropy is a volume effect or a surface effect, although we favour the second hypothesis. If it is a volume effect, than A corresponds to a specific torque of order of 10^{-4} dyn/cm^2; if it is a surface effect, it corresponds to a surface azimuthal anchoring energy of order of 10^{-7} erg/cm^2, i.e., 5 orders of magnitude smaller than a typical azimuthal anchoring energy of a homogenously aligned NLC. Perhaps, the residual anisotropy might be further decreased by using different surface-treatements, since this is certainly a critical factor.*

CONCLUSIONS

For the first time, the phenomenon of OFT induced in a nematic liquid crystal by depolarized light is studied, both theoretically and experimentally. We found that the threshold for OFT is twice that of linearly polarized light. The plane containing the director reorientation and giving the slow-axis of the induced birefringence is left undetermined by light, and is ideally completely "free" from any internal constraint. It is somewhat similar to a tiny levitating particle. Therefore this degree of freedom is extremely sensitive to small perturbations and weak external fields. When the polarization coherence time of the depolarized light is not much shorter than the liquid crystal response time, the reorientation plane becomes sensitive even to the small average effect of polarization correlations, or equivalently to the fourth order correlations of the optical field amplitude.

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*For instance, samples prepared with ITO conductive coating showed much stronger residual anisotropy, so that we were unable to see any effect of light fluctuations.

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