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Optical reorientation in nematic liquid crystals controlled by the laser beam shape

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Abstract

We demonstrate experimentally that the transverse shape of the laser beam can strongly affect the laser-induced molecular reorientation in nematic liquid crystals. In this paper we studied only the case of elliptical beam cross-section. In this case, a relatively strong extra-torque was observed tending to reorient the liquid crystal director parallel to the ellipse major axis. The extra-torque is zero for circular beam cross-section and increases as the beam cross-section becomes more and more elliptical. © 1999 Published by Elsevier Science B.V. All rights reserved.

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1. Introduction

The giant optical nonlinearity due to optical-field-induced molecular director reorientation in the mesophases makes liquid crystals (LC) unique as nonlinear optical materials, and it gives rise to some very unusual optical effects that do not exist in other media. The optical Fréedericksz transition (OFT) was the first example reported in the literature [1,2], but many other unusual optical effects were discovered in the last decade, such as self-induced stimulated light scattering (SISLS) [3–5], intrinsic optical bistability [6], laser-induced nonlinear optical oscillations [7–9], deterministic chaos [10], spontaneous pattern formation, etc. All these experiments confirm the possibility of optically controlling the LC molecular reorientation by changing either the beam intensity or the beam polarization.

In this paper we show that also the shape of the beam cross section can be used as an effective parameter to control the LC reorientation. In particular, we found that an elliptically shaped laser beam can induce a relatively strong torque along the beam axis, tending to rotate the LC molecules towards the major axis of the beam profile. This beam shape-induced torque is zero for circular beam cross-section and increases with the beam shape ellipticity. Although circularly shaped laser beams are commonly used in the experiments, this shape-induced extra-torque may be of some importance in experiments made at very large oblique incidence angle or in strongly asymmetric geometries as in planar optical waveguides.

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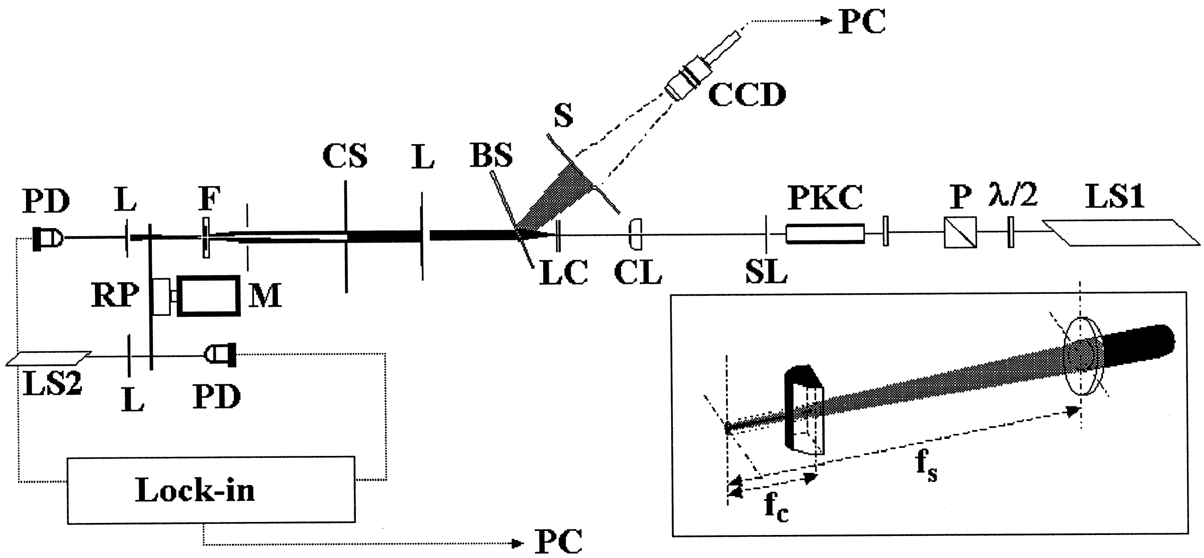

Fig. 1. Our experimental apparatus. LS1, LS2 laser sources, P polarizer, PKC Pockels cell, SL spherical lens, CL cylindrical lens, LC liquid crystal sample, S screen, BS beam splitter, CS circular stop, F neutral filter, L1, L2, L3 lenses, PD1, PD2 photodiodes, RP rotating polarizer, M motor, PC personal computer. Inset: lens geometry for producing elliptical beam cross-sections.

In order to distinguish the reorientation effects due to the beam shape from those due to the beam polarization, we made our measurements both with polarized and unpolarized light at normal incidence. The combination of a spherical and a cylindrical lens was used to produce an elliptical spot at the sample position (see inset in Fig. 1). In the case of unpolarized light, only the beam shape-induced optical torque survives and we observed that, when the laser intensity is raised above the threshold for the optical Fréedericksz transition (OFT), the LC director has a pronounced tendency to become aligned along the major axis of the elliptical spot at the cell. We made also measurements by using a laser beam linearly polarized at 45° with respect to the major axis of the spot ellipse. A competition between the beam polarization-induced and shape-induced torques was observed in this case.

2. The origin of the beam shape-induced optical torque

Any model for the optical reorientation in liquid crystals accounting for the finite beam cross-section is of formidable mathematical complexity, because the equations governing the elastic distortion in the material and the propagation of light in the medium must be solved retaining all spatial dimensions. Besides the plane-wave, that was extensively discussed in the literature, only the so-called ribbon problem, where the laser beam is assumed to be of infinite extension in one dimension, can be handled analytically, at least in the small distortion linear approximation and for particular beam profiles [11]. In this section we provide only a general energy argument to show why a reorientation along the major axis of the beam spot at the sample is preferred. Let us assume a completely unpolarized laser beam normally incident onto the sample. Let us assume also strong homeotropic boundary conditions at the sample walls. Then, if the beam spot has a circular shape, the whole system, radiation + LC, is cylindrically symmetric around the beam propagation axis, that we take as z-axis. In these conditions it can be shown that the optical reorientation can nevertheless occur provided the laser intensity is above a threshold value, which is twice the threshold intensity for the usual OFT in the case of linear polarization of the beam [12]. Because of the overall cylindrical symmetry, however, the initial plane where the LC director $\boldsymbol{n}$ moves remains completely unpredictable.

When the laser spot at the sample is made elliptical, the cylindrical symmetry is broken and a well defined preferential orientation plane can be expected. By symmetry consideration it is clear that such a plane should contain the z-axis and one of the two axes of the elliptical beam shape. Let a and b denote the length of the ellipse major and minor axis, respectively, and L the sample thickness. Assuming small elastic distortion, when the prevailing orientation direction is along the major axis the elastic energy density of the LC is given by

$$F_e \simeq \frac{k_{11}}{a^2} + \frac{k_{22}}{b^2} + \frac{k_{33}}{L^2} \,. \tag{1}$$

When the prevailing orientation distortion is along the ellipse minor axis, we have, instead

$$F_e \simeq \frac{k_{11}}{b^2} + \frac{k_{22}}{a^2} + \frac{k_{33}}{L^2} \,. \tag{2}$$

Because $a > b$ and, for usual nematic materials $k_{11} > k_{22}$, we see that in the first case (dominant twist distortion) the elastic energy is lower than in the second (dominant splay distortion). Therefore the orientation along the major axis is energetically preferred. It should be noticed that the two energies become equal in the one constant approximation, which implies that the elastic anisotropy of the nematic material is a crucial feature of this phenomenon. The simple argument presented here cannot evidently account for the details of the beam shape-induced torque, and a more complete theory is desirable. Nevertheless, this analysis shows that a sample initially oriented along a direction out of the beam spot ellipse major axis will experience a torque, tending to force the director $\boldsymbol{n}$ back along this axis.

3. The experiment

Our experimental apparatus is shown in Fig. 1. The sample was a 50 μm thick cell filled by the commercial nematic mixture E7 from Merck. The cell walls were coated with DMOAP for homeotropic alignment. Due to the very large temperature range where the E7 mixture is nematic (10°C–60°C), no temperature control of the cell was necessary. The main laser source (LS1) was a 5 W maximum-power frequency-doubled Nd:Yag cw laser ($\lambda = 532$ nm). The laser beam was sent through a variable attenuator and then through a Pockels cell (PKC) to obtain depolarized light. The Pockels cell fast axis was oriented at 45° with respect to the laser polarization plane and it was driven by a saw-tooth signal having λ-amplitude and frequency of 200 Hz. In this way, the polarization of the beam was made to pass very quickly through the states outlined in Fig. 2, yielding an effectively depolarized beam, as seen by the slow-responding liquid crystal sample. We used a polarimeter to be sure that all Stokes parameters of the beam emerging from the Pockels cell had zero mean value on the LC response time scale (~ 1 s). In the experiments with polarized light the Pockels cell was removed. The spherical lens SL ($f_s = 500$ mm) and the cylindrical lens CL ($f_c = 15$ mm) were used to obtain an elliptical spot at the sample (LC). The two lenses were assembled in the nearly confocal configuration, as shown in Fig. 1 inset. The major-to-minor axis ratio e of the beam spot size at the sample was changed by moving the LC film across the focal zone of the two lenses. In this way, e could be changed from 1 to 5. In practice, the vertical dimension of the laser spot was kept fixed at about 75 μm (half-width at $1/e^2$ intensity), while the other dimension was reduced, by the cylindrical lens, down to 15 μm. The light emerging from the LC sample was divided by a beam splitter (BS2). One beam was sent onto a screen (S), where the characteristic far-field self-diffraction ring pattern [1,2] produced by our LC film was observed by a CCD camera. From the acquired pattern, we extracted the angular aperture Θ of the outermost ring. The other beam was focused by the large aperture (15 cm diameter) lens L1 ($f_1 = 50$ cm) and by the lens L2 onto the photodiode PD1. The central zone of the beam was intercepted by a circular stop (CS). Before arriving in the photodiode, the light was first attenuated by a neutral filter (F) and then made to pass through a polarizer (RP), continuously rotating at about 50 Hz. The output of the photodiode was sent into the signal channel of a lock-in amplifier. A linearly polarized red diode laser (LS2) passed through the same rotating polarizer and was col-

Fig. 2. Polarization state sequence of our depolarized beam.

lected by the lens (L3) into a second photodiode (PD2) to provide the reference signal to the lock-in. The phase difference $\Delta\psi$ between signal and reference was determined by the lock-in and sent to the PC for storage and real time visualization, together with Θ.

The working principle of our equipment is based on the idea that only the extraordinary component of the optical wave can suffer the self-diffraction process and, hence, can be scattered into the outer region of the far-field pattern. The ordinary component, instead, propagates very close to the beam axis and it is almost completely eliminated by the circular stop CS. The result is that the light passing through the rotating polarizer is almost linearly polarized in the extraordinary direction. The phase difference $\Delta\psi$ between the signals of the photodiodes PD1 and PD2 is then twice the angle between the average polarization direction of the outer region of the far-field ring pattern and the fixed polarization direction of the reference diode laser LS2. The advantage of measuring $\Theta(t)$ and $\Delta\psi(t)$ as functions of time is that these quantities are simply related to the average values of the polar angles $\theta(t)$ and $\phi(t)$ of the molecular director $\boldsymbol{n}$ of the LC. We have, in fact,

$$\Theta \propto \langle \theta \rangle^2, \quad \Delta\psi \simeq \langle 2\phi \rangle, \tag{3}$$

where $\langle \cdot \rangle$ denotes the spatial average across the LC film. The first of relations (Eq. (3)) is well known [13] and the second follows from application of Mauguin's theorem [14] to the extraordinary wave. Although these relationships are only approximate and hold true for weak elastic distortion in the LC, their main feature is that Θ and $\Delta\psi$ are related to independent degrees of freedom of $\boldsymbol{n}$. With this apparatus we could get a real time track of the average motion of the molecular director $\boldsymbol{n}$ in the LC film, but, in the present paper, we reported only the steady state value of the azimuthal angle ϕ as a function of the beam intensity I and the spot ellipticity e. The study of the reorientation dynamics of $\boldsymbol{n}$ is deferred to future work.

We made two sets of measurements with elliptically shaped beam at normal incidence: in the first we used unpolarized light and in the second we used light that was linearly polarized at 45° with respect the major axis of the beam spot at the sample.

3.1. Measurements with unpolarized light

Some of our experimental results are reported in Fig. 3. These data were taken by orienting the cylindrical lens so as to have the major axis of the elliptical spot at the sample forming an angle $\alpha = 30°$ with respect to the horizontal plane. We repeated the measurements at $\alpha = 0°$, 60°, 90° with similar results. In all cases, for large enough ellipticity ($e \geq 2$) and large enough laser intensity ($I \geq 1.3\, I_{th}$, where I_{th} is the OFT threshold), the average azimuthal angle ϕ of $\boldsymbol{n}$ tends to be aligned with the major axis of the spot (30° in Fig. 3), confirming the presence of the beam shape-induced extra-torque. As the beam shape becomes more and more circular ($e \to 0$), the steady state value of ϕ tends towards a well defined but unpredictable value, changing from point to point in the sample. The same effect occurs when the laser intensity is decreased towards the threshold for fixed spot ellipticity e, although, in this limit, the accuracy of our measurements is reduced because the far-field ring pattern is almost completely intercepted by the circular stop. A similar repeatable, but unpredictable, azimuthal reorientation has been observed in previous experiments with unpolarized light and circular beam shape [12]. We ascribe this unpredictable reorientation to small uncontrolled and unavoidable factors breaking the perfect overall cylindrical symme-

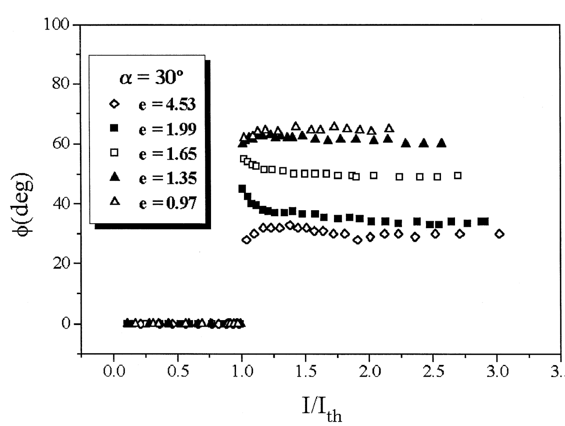

Fig. 3. Steady state value of the director average azimuthal angle ϕ as a function of the reduced laser intensity I/I_{th} for different values of the ellipticity e of the unpolarized beam spot at the sample. The spot ellipse major axis was oriented at $\alpha = 30°$. Below threshold ($I/I_{th} < 1$), the angle ϕ has no physical meaning.

try of the system. These may be, for instance, a non-perfect parallelism of the sample walls, a residual polarization degree in the incident light, a small deviation from perfect normal incidence, a small pre-tilt at the sample surface (this could explain the observation that the final value of ϕ may change from point to point in the sample), etc.

We measured also the threshold intensity for the OFT as a function of the beam spot ellipticity e. We found that the threshold intensity is almost independent of e, as expected, except for a small increase observed at very high values of e. This increase can be accounted for by noting that for large values of e the minor axis of the beam spot was smaller than cell thickness, so that well-known finite beam effects become appreciable [15,16]. The measured threshold intensity for a circular spot was $I_{th} = 7.16\ \mathrm{kW/cm^2}$.

3.2. Measurements with polarized light

Some of our experimental results are reported in Fig. 4. These data were taken by orienting the cylindrical lens so as to have the major axis of the elliptical spot at the sample forming an angle $\alpha = 45°$ with respect to the horizontal plane. The Pockels cell was removed so that the incident laser beam was linearly polarized in the horizontal plane. Therefore the z-component of the optical torque associated with light polarization is expected to attract the director $\boldsymbol{n}$ toward the plane $\phi = 0$, while the torque associated with the beam shape should attract $\boldsymbol{n}$ toward the plane $\phi = 45°$. If these two torques have a comparable magnitude, the equilibrium director should settle on intermediate values of ϕ, as determined by the balance of the two competing torques. In fact, in Fig. 4 we may see that the high-intensity value of ϕ ranges from about 0°, achieved for ellipticity e close to 1 where the polarization torque dominates, to about 45°, achieved for large values of e where the beam-shape torque dominates. Moreover we see that the equilibrium value of ϕ is approximately independent of intensity, except for a small range of intensities close to the threshold. An intensity-independent equilibrium is possible if the two torques have the same intensity dependence. Instead, we cannot readily explain the low-intensity behaviour of ϕ, where we would rather expect a simple proportionality of both torques to $I - I_{th}$ and therefore a constant ϕ. Actually, this behaviour might be an artefact of our detection approach, since, as noted above, our near-threshold measurements are poorly accurate. Alternatively, it could be the effect of the residual torques due to the imperfect cylindrical symmetry of the system. This effect would emerge only for $I - I_{th}$ small enough that both polarization and beam-shape optical torques are reduced to a magnitude comparable to the residual torques. The intensity threshold for the OFT in the case of circular beam shape was found to be $I_{th} = 4.18\ \mathrm{kW/cm^2}$, about half of the threshold for unpolarized light, confirming previous results and theory [12].

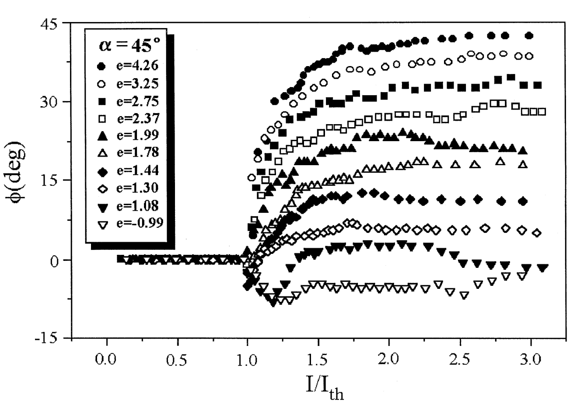

Fig. 4. Steady state value of the director average azimuthal angle ϕ as a function of the reduced laser intensity I/I_{th} for different values of the ellipticity e of the beam spot at the sample. The laser beam was linearly polarized in the horizontal plane $\phi = 0$ and the spot major axis was at $\alpha = 45°$ from the same plane.

4. Conclusions

We presented a set of measurements on the optical reorientation in nematic liquid crystals proving the existence of an optical torque related to the shape of the beam at the sample position. This torque acts along the beam propagation direction and may be competitive with the usual optical torque due to the beam polarization. The torque due to the beam shape vanishes for beams having circular cross section, so that it can be observed only when elliptically shaped beams are used or in experimental geometries where the overall cylindrical symmetry of the system is broken as, for instance, in planar optical waveguides. In order to single out the beam-shape-induced torque, in our experiments we used unpolarized light. We observed that for large enough beam ellipticity e the laser induced molecular reorientation in the nematic film always occurs along the major axis of the elliptical beam profile. For low ellipticity values, $e < 2$, the reorientation plane was governed by minor unavoidable symmetry-breaking imperfections of the system and, hence, it was unpredictable. We repeated our measurements using a laser beam linearly polarized at 45° with respect to the ellipse major axis. In this case, the steady state reorientation occurs at an intermediate angle between the ellipse major axis and the beam polarization direction, evidencing a competition between the beam-shape and polarization optical torques. The beam shape-induced torque should originate ultimately from the lack of cylindrical symmetry in the incident beam, although simple elastic energy considerations suggest that the phenomenon is also related to the anisotropy of the LC elastic constants. Our measurements are not accurate enough to provide detailed quantitative information about the shape-induced torque as, for example, on how this torque depends on the laser intensity and beam ellipticity. Future work is planned in this direction.

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