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## Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/gmcl20>

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Version of record first published: 18 Oct 2010

To cite this article: Enrico Santamato, Bruno Piccirillo & Angela Vella (2002): The Role of the Orbital Angular Momentum of Light in the Optical Reorientation of Liquid Crystals, *Molecular Crystals and Liquid Crystals*, 375:1, 607-616

To link to this article: <http://dx.doi.org/10.1080/10587250210575>

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## **The Role of the Orbital Angular Momentum of Light in the Optical Reorientation of Liquid Crystals**

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In this work we studied experimentally the optical effects produced by the orbital angular momentum of light on the Optical Fréedericksz Transition (OFT) in a nematic liquid crystal film. The measurements were carried out using a linearly polarized astigmatic laser beam with elliptical cross-section. The laser-induced reorientation was found to be strongly dependent on the angle between the polarization and the major axis of the beam profile. A threshold increase, out-of-polarization-plane reorientation and persistent oscillations were found to be the newest features observed in our experimental conditions. We ascribed all these features to the presence of an additional torque due to the orbital angular momentum of light.

Keywords: Angular Momentum, Liquid Crystals, Nonlinear Optics

### **INTRODUCTION**

The giant optical nonlinearity due to the optical-field-induced molecular 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 literature <sup>[1, 2]</sup>, but many other optical effects were discovered in the last two decades, such as Self-Induced Stimulated Light Scattering (SISLS) <sup>[3, 4, 5]</sup>, intrinsic optical

bistability [6], laser-induced nonlinear optical oscillations [9, 10, 11], deterministic chaos [12], spontaneous pattern formation [13], light-driven molecular motors [7, 8], etc. All these effects originated from the optical torque  $\tau_o$  acting on the unit volume in the liquid crystal, given by

$$\tau_o = \frac{\epsilon_a}{4\pi} \langle \mathbf{D} \times \mathbf{E} \rangle, \quad (1)$$

where  $\epsilon_a = n_e^2 - n_o^2$ ,  $n_e$  and  $n_o$  are the extraordinary and ordinary refractive indexes of the material, respectively, and the brackets mean the time average. The optical torque  $\tau_o$  acts also in crystals, as demonstrated long time ago by Beth in a clever experiment where the torque exerted by a circularly polarized light beam on a quartz plate was measured using a torsion balance [14]. In this experiment the optical torque on the quartz plate originated from the transfer of the photon spin from the light beam to the body. The photon spin transfer process was used also to put into rotation liquid crystals [3] and, more recently, microscopic calcite fragments trapped in optical tweezers [15].

In isotropic materials the torque (1) vanishes, since  $\mathbf{D}$  and  $\mathbf{E}$  are parallel to each other. Nevertheless, both theory [17, 18] and experiment [16] show that also isotropic transparent bodies may be rotated by a light beam, provided they are optically inhomogeneous. The optical torque acting on isotropic bodies cannot be ascribed to the torque given by Eq. (1) and it was soon recognized as due to the transfer to the body of the photon orbital angular momentum, rather than to the spin one. We then conclude that the torque associated to the photon orbital angular momentum transfer is a new source of optical torque, whose action on liquid crystals has never been investigated before. We expect, in fact, that in proper conditions liquid crystals could be sensitive to the orbital angular momentum of light too, since, besides anisotropy, these materials may exhibit a spatially refractive index changing from point to point. This was indeed demonstrated in a recent experiment made in liquid crystals using unpolarized light (which is another way to make  $\tau_o = 0$ ) [19].

The aim of the present work is presenting a preliminary experimental

study on the effects produced by the transfer of the orbital angular momentum of light on the OFT in nematic liquid crystals.

## THE TRANSFER OF THE ORBITAL ANGULAR MOMENTUM OF LIGHT TO LIQUID CRYSTALS

In the case of liquid crystals, the optical torque given by Eq. (1) is nonzero, in general, so we expect to observe a combined effect of the photon spin and the photon orbital angular momentum when liquid crystals are optically reoriented. In order to make the effects due to the orbital angular momentum of light apparent, we must first find out a situation where the orbital angular momentum of light is transferred to liquid crystals and then try to separate these effects from the ones due to the usual optical torque  $\tau_o$ . It is worth noting that laser beams which are eigenstates of the orbital angular momentum, as for example Laguerre-Gauss beams, are not useful because they cannot transfer their angular momentum content to transparent media <sup>[17]</sup>. The torque exerted on a transparent body by the orbital angular momentum of light is, in fact, <sup>[17]</sup>

$$M_z = \frac{i}{\omega} \int dx dy I(x, y) \hat{L}_z \Delta\psi(x, y) = -\frac{i}{\omega} \int dx dy \Delta\psi(x, y) \hat{L}_z I(x, y) \quad (2)$$

where

$$\hat{L}_z = -i \left( x \frac{\partial}{\partial y} - y \frac{\partial}{\partial x} \right) \quad (3)$$

is the usual operator for the  $z$ -component of the orbital angular momentum in units of  $\hbar$ ,  $\omega$  is the optical frequency,  $I(x, y)$  is the intensity profile of the light beam across the transverse plane, and  $\Delta\psi(x, y)$  is the phase change suffered by the optical field in traversing the medium. The last equality in Eq. (2) was obtained upon a partial integration, which is correct in the present case, as  $I(x, y)$  and  $\Delta\psi(x, y)$  are both single-valued

functions\*. From Eq. (2) we see that  $M_z$  vanishes if either  $I(x, y)$  or  $\Delta\psi(x, y)$  are cylindrically symmetric around the  $z$ -axis. All the eigenstates of  $\hat{L}_z$  have a cylindrically symmetric intensity profile, so that their orbital angular momentum produces a zero torque on transparent bodies. We must use, therefore, light beams which are in a superposition of eigenstates of  $\hat{L}_z$  and regard to  $M_z$  as the average torque exerted on the body. Moreover, we stress that we could even use an incident light beam carrying zero average angular momentum. In fact, Eq. (2) shows that  $M_z$  is given by the *change* in the average orbital angular momentum of the beam produced by the medium. Our experiment, in fact, have been carried out with a laser beam having no average angular momentum. The simplest way to produce a light beam able to transfer its orbital angular momentum to a transparent body is making its transverse intensity profile elliptical by using cylindrical lenses. To see this, let us assume, for example, that the beam intensity profile at the sample position is given by

$$I(x, y) = \left( \frac{P}{\pi w_1 w_2} \right) e^{-\frac{x^2}{w_1^2} - \frac{y^2}{w_2^2}} \quad (4)$$

where  $P$  is the power carried by the beam and the two waists have been chosen so that  $w_1 > w_2$ . Then, let us assume that the phase change produced by the body has an astigmatic parabolic profile elongated in a direction forming an angle  $\alpha$  with the  $x$ -axis:

$$\Delta\psi(x, y) = -\frac{\pi}{\lambda} \left[ \frac{(x \cos \alpha + y \sin \alpha)^2}{f_1} + \frac{(x \sin \alpha - y \cos \alpha)^2}{f_2} \right]. \quad (5)$$

with effective focal lengths  $f_1 > f_2$ . We may then calculate the integral in Eq. (2) explicitly, obtaining

$$M_z = -\left( \frac{P}{4c} \right) \left[ \frac{(w_1^2 - w_2^2)(f_1 - f_2)}{f_1 f_2} \right] \sin 2\alpha. \quad (6)$$

Equating  $M_z$  to (minus) the viscous torque  $\gamma\dot{\alpha}$ , where  $\gamma$  is an effective friction coefficient, we find that for a focusing medium the only stable

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\*We are assuming that the body refractive index has a continuous distribution and that the body surface is disclination-free.

solution is  $\alpha = 0$ . In this case, the body suffers a torque which tends to align the major axis of  $\Delta\psi(x, y)$  with the major axis of  $I(x, y)$ . In the case of a defocusing medium, the two axes tend to be orthogonal. We notice that  $M_z \neq 0$  even if the incident beam carries no angular momentum. The amount of orbital angular momentum subtracted to the beam is determined by the process itself and it changes in time as long as the body is rotating. The whole process is intrinsically nonlinear, irrespective of the value of the beam intensity  $I$ , which is a characteristic of the Self-Induced Stimulated Light Scattering [3]. We observe that both the photon spin and the photon orbital angular momentum are transferred by the Self-Induced Stimulated Light Scattering process.

Having found how to transfer the orbital angular momentum of light to a body, the next problem, when liquid crystal are envisaged, is finding out an experimental situation where the spin and the orbital angular momentum contributions to the optical reorientation can be separated. We decided to perform a series of measurements in a geometry which is the same as in the standard OFT, apart from the fact that the beam cross-section at the sample position was taken elliptical. In this way, deviations from the well-known behaviour of the OFT could be ascribed to transverse effects connected to the elliptical shape and to the orbital angular momentum of light. We tried also to obtain a model to describe the reorientational effects due to the photon orbital angular momentum, but this is a very difficult task, because the plane-wave approximation is inadequate and one is faced, since the very beginning, with a full 3-D elastic and optical problem.

## THE EXPERIMENT

Our sample was a film 50  $\mu\text{m}$  thick of E7 nematic liquid crystal enclosed between glass walls coated with DMOAP for homeotropic alignment. A Nd:YVO<sub>4</sub> laser beam ( $\lambda = 532 \text{ nm}$ ) was focused onto the sample at normal

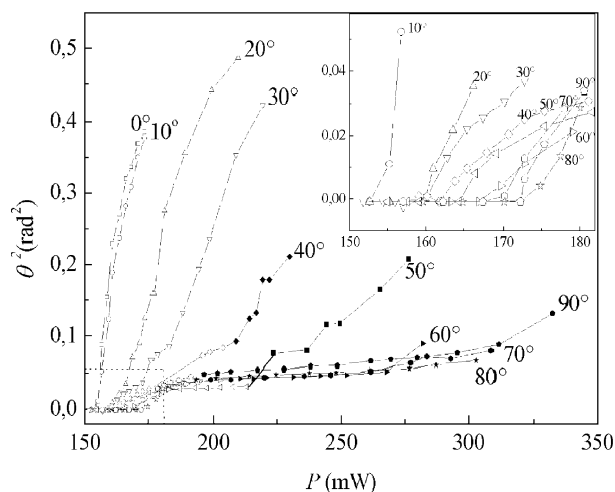


FIGURE 1 Square of the zenithal angle  $\theta$  of the molecular director as a function of the incident power  $P$ . The parameter over the curves is the beam polarization angle  $\beta$ . The inset shows the details in the near threshold region.

incidence by a set of cylindrical lenses so to produce at the sample position an elliptical intensity profile with waists  $w_1 = 100 \mu\text{m}$  and  $w_2 = 10 \mu\text{m}$ . The laser beam was linearly polarized and the polarization direction was manually rotated by a  $\lambda/2$  plate. When the laser power exceeded the threshold for the OFT, diffraction rings were formed in the far field beyond the sample. We used a CCD camera and a rotating polarizer to measure the angular diameter  $\Theta$  and the average polarization direction  $\Phi$  of the outermost ring. These two quantities provide roughly independent parameters to describe the average optical reorientation in the sample, because  $\Theta$  is roughly proportional to the square  $\theta^2$  of the zenithal angle  $\theta$  of the molecular director  $\mathbf{n}$ , and  $\Phi$  provides a rough measure of its average azimuthal angle  $\varphi$ . With this apparatus, we made several measurements of  $\Theta$  and  $\Phi$  by changing the power  $P$  and the angle  $\beta$  formed between the beam polarization direction and the major axis of its intensity profile. From these data and from the geometry of our experimental setup we

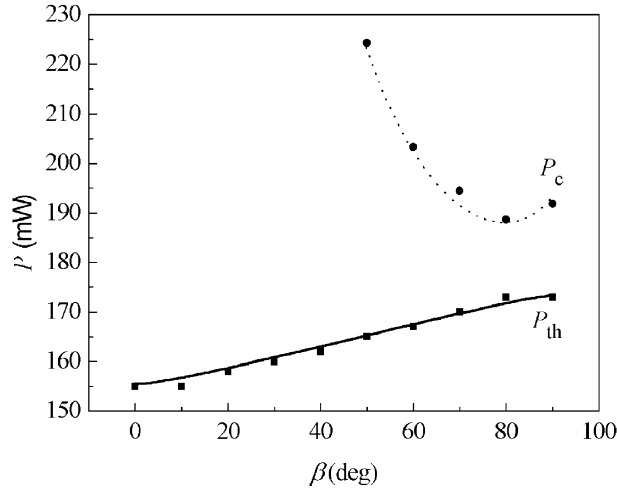


FIGURE 2 The threshold power  $P_{th}$  of the OFT and the critical power  $P_c$  where the oscillating states start up are reported as functions of the angle  $\beta$  between the beam polarization direction and the major axis of the intensity profile at the sample position.

deduced  $\theta^2$  and  $\varphi$ .

In Fig. (1)  $\theta^2$  at steady state is reported as a function of the power  $P$ , using  $\beta$  as parameter. We observe that the transition to the reoriented state is second order in the same way as the standard OFT [see the inset in Fig. (1)]. However, at a second critical power  $P_c$  the steady state becomes unstable and persistent nonlinear oscillations of  $\theta$  start up. The oscillating states are indicated in the figure by black symbols corresponding to the time averaged value of  $\theta$ . A similar behaviour has never been reported in the standard OFT configuration, so that we are led to ascribe the occurrence of the oscillating states to the competition between the photon spin and the photon orbital angular momentum in the optical reorientation process. As the polarization direction angle  $\beta$  was increased from 0 to  $90^\circ$  the threshold power for the OFT increased while the critical power  $P_c$  for instability decreased until, at  $\beta = 90^\circ$ ,  $P_{th}$  and  $P_c$  turn to be about



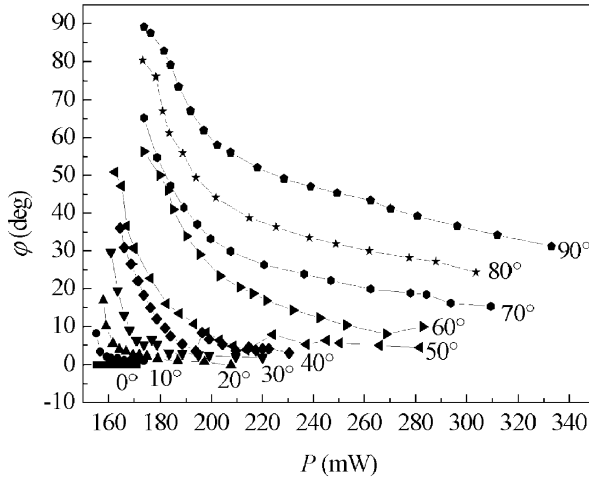


FIGURE 3 The azimuthal angle  $\varphi$  of the molecular director as a function of the incident power  $P$ . The parameter is the beam polarization angle  $\beta$ .

equal. This behaviour is shown in Fig. (2). Finally, in Fig. (3) we report the measured azimuthal angle  $\varphi$  as a function of the incident power  $P$ , using the polarization angle  $\beta$  as a parameter. We notice that very close to threshold, the molecular distortion starts up in the beam polarization plane ( $\varphi \simeq \beta$ ) as in the standard OFT, but as the power is increased, the reorientation plane moves towards the major axis of the intensity profile  $I(x, y)$ . This is also a clear manifestation of the competition between the transfer of the orbital and spin angular momentum of light. At steady state, in fact, the torques due to the transfer of the spin and of the orbital angular momentum of light must exactly balance.

## CONCLUSIONS

We presented a set of preliminary measurements on the laser-induced re-orientation in nematic liquid crystals in a geometry where both the spin

and the orbital angular momentum of light produce a torque on the sample. This was achieved by using a set of cylindrical lenses to render the cross-section of the laser beam elliptical at the sample position. We observed significant differences between the optical reorientation with the elliptically shaped beam and the standard OFT. In particular, the overall features of the molecular reorientation turned to be strongly dependent on the angle  $\beta$  between the beam polarization direction and the major axis of the intensity profile. Moreover, above a second critical power of the incident beam, we observed the onset of persistent nonlinear oscillations of the molecular director, an effect never observed in the standard OFT. Finally, our experiments indicate that the reorientation occurs in a plane different from the light polarization plane, thus demonstrating that both the spin and the orbital angular momentum of light are involved in the reorientation process. Modelling these experimental results seems a very difficult task because the plane wave approximation is inapplicable and the usual formula for the optical torque given in Eq. (1) is insufficient. Further experimental observation may also be of great help. The new effects observed in the present work may be of some relevance in geometries where the optical field is strongly confined in one dimension, as it is, for example, in optical waveguides, where an extra-torque could arise due to the strongly astigmatic shape of the propagating beam.

### Acknowledgments

We acknowledge the Istituto di Fisica della Materia for financial support.

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