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Undamped Nonlinear Oscillations of the Molecular Director Induced in a Liquid Crystal Film by the Competition between the Orbital and Spin Angular Momentum Transfer from Light

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Undamped nonlinear oscillations of the molecular director in a homeotropically aligned nematic liquid crystal film have been observed in the field of a normally incident laser beam with elliptical transverse cross-section and linear polarization along the direction orthogonal to the ellipse major axis. This phenomenon may be brought back to the competition between the longitudinal torques originating from the spin and orbital angular momenta the incident beam transfer to the sample.

Keywords: nonlinear optics; spin angular momentum; orbital angular momentum

INTRODUCTION

It is well-known that liquid crystals are able to couple to the angular momentum of light. In 1986, in fact, the collective rotation of liquid crystal molecules in the field of a normally incident circularly polarized laser beam was observed [1]. This phenomenon was explained in terms of a Self-Induced Stimulated Light Scattering (SISLS) process where a transfer of spin angular momentum from the beam to the sample was involved [2].

The total angular momentum $\boldsymbol{J}$ carried by the electromagnetic field is expressed in classical form by

$$\boldsymbol{J} = \frac{1}{4\pi c} \int \boldsymbol{r} \times (\boldsymbol{E} \times \boldsymbol{H})\, dV, \tag{1}$$

where $\boldsymbol{E}$ and $\boldsymbol{H}$ are the electric and magnetic field respectively, c the speed of light in the vacuum and the integral is performed all over the space. For monochromatic field, in the paraxial approximation, the angular momentum $\boldsymbol{J}$ may be univocally expressed as a sum $\boldsymbol{J} = \boldsymbol{L} + \boldsymbol{S}$ of an orbital part, $\boldsymbol{L}$, associated with spatial distribution and an intrinsic (or spin one), $\boldsymbol{S}$, associated with polarization.

In most cases, the angular momentum of light produces such small effects that experimental observation is a hard matter. In fact, the spin angular momentum flux carried by a circularly polarized 10 mW He-Ne laser-beam is of the order of 10^{-18} Nm. All the methods adopted to gather a direct observation of angular momentum transfer are based on the fact that it results into a torque along the propagation direction making the irradiated body rotate. In 1936, R. A. Beth, as first, measured the torque that a circularly polarized light beam exerted on a half-wave plate suspended by a fine quartz fiber [3]. Many other experiments have been made involving a spin angular momentum transfer to matter, such as the above mentioned SISLS process in liquid crystals [1, 2] or similar experiments performed on microscopic transparent calcite particles held by optical tweezers and made to rotate by circularly polarized light [4].

Five years ago, He *et al.* [5] – inspired by Allen *et al.* [6], who proposed a *gedanken experiment* analogous to the one of Beth's but aimed at detecting the orbital angular momentum – experimentally observed a transfer of orbital momentum to matter trapping absorbing particles in the dark central minimum of a linearly polarized hologram generated Laguerre-Gauss beam. Later, Friese *et al.* [7] reported on the transfer of both orbital and spin angular momentum, noticing that the two momenta added together.

In this paper a new experiment is presented involving a transfer of

orbital angular momentum from radiation to liquid crystals. The guiding idea the experiment is based on comes from the above cited theoretical work by Allen *et al.* according to which the measurement of the mechanical torque arising from the orbital angular momentum is to be performed from Beth's experiment mould. In such an experiment, the astigmatism of the optical system stands for the birefringence of the retardation plate in Beth's experiment.

TRANSFER OF ORBITAL ANGULAR MOMENTUM FROM RADIATION TO LIQUID CRYSTALS

An astigmatic optical element, adding a local phase $\Delta\psi(x, y)$ to an arbitrary input field, changes its orbital angular momentum and suffers in its turn a mechanical torque along the propagation direction z given by [8]

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

where ω is the optical frequency, $\hat{L}_z = i\,(x\partial_y - y\partial_x)$ is the angular momentum operator along z, $I(x, y)$ is the intensity profile and $\Delta\psi(x, y)$ is the phase change distribution produced by the astigmatic component, such as for instance a cylindrical lens. The last equality holds because the operator $\hat{L}_z$ is hermitian (we assume no singularity in the phase field $\Delta\psi(x, y)$). From Eq. (2) one argues it is impossible changing the orbital angular momentum of the incoming field by means of a single thin cylindrical lens, provided that the transverse intensity distribution breaks the ordinary laser beam cylindrical symmetry or the phase field exhibits a singularity (against the previous assumption).

A homeotropically aligned NLC film of thickness L, irradiated at normal incidence by means of a laser beam with a non-cylindrically symmetric transverse intensity profile $I(x, y)$, above the threshold for the Optical Fréedericksz Transition (OFT), behaves in a sense as a thin astigmatic lens. The reoriented sample, in fact, in these circumstances, will modify the incoming field distribution by introducing a non-symmetrical local

phase $\Delta\psi(x, y)$ of its own, just as a cylindrical lens. The sample may consequently suffer a mechanical torque along the propagation direction, expressed by Eq. (2), and the molecular director $\boldsymbol{n}$ may start to rotate around the beam axis.

Modelling molecular reorientation in the field of a non-cylindrically symmetric laser beam requires to release the commonly adopted plane wave approximation. Though the full complexity of the problem is still beyond our grasp, it is possible exploiting Noether's theorems to find out a conservation law for the total angular momentum of the system *liquid crystal + optical field*. For small elastic distortion ($n_x, n_y \ll 1, n_z \simeq 1$), the LC free energy density reads as [9]

$$F = \frac{1}{2}A(\partial_x n_x + \partial_y n_y)^2 + \frac{1}{2}B(\partial_y n_x - \partial_x n_y)^2 + \frac{\pi^2}{2}(\tilde{I} - 1)(n_x^2 + n_y^2), \quad (3)$$

where $A = k_{11}/k_{33}$, $B = k_{22}/k_{33}$, $\tilde{I} = I/I_{th}$, $I_{th} = ck_{33}n_e^2\pi^2/n_o(n_e^2 - n_o^2)L^2$ being the OFT threshold intensity for normal incidence and linear polarization (n_e, n_o are the refractive indices and k_{ii} the elastic constants of the material). Assuming that $n_x, n_y \propto \sin(\pi z/L)$ and integrating both sides of Eq. (3) over the sample thickness, a description of the problem is attained in terms of the transverse coordinates x and y only, and the sample may be regarded as an optical transducer. At first, supposing an unpolarized laser beam impinges on the sample, no transfer of spin angular momentum takes place. Then $\tilde{I}$ is assumed to depend on the transverse coordinates x and y, i.e. $\tilde{I} = \tilde{I}(x, y)$ – the z-dependence is obviously missing, as the medium is non-absorbing. The equations for the motion of n_x and n_y are obtained by adding to the Euler-Lagrange equations derived from the free energy $\mathcal{F} = \int F\, dxdy$ a viscous term, taken as $(-\dot{n}_x, -\dot{n}_y)$, where the dot means derivative with respect to the reduced time $\tau = (k_{33}/\gamma L^2)t$, and γ is the effective viscosity coefficient. Using Noether's theorems, the conservation law for the z-component of the total angular momentum flux is obtained in the form

$$\nabla\cdot\boldsymbol{\theta}_d = \frac{i\pi^2}{2}\tilde{I}(x, y)\hat{L}_z(n_x^2 + n_y^2) + i\sum_{k=x,y}\dot{n}_k\hat{L}_z n_k + (\dot{n}_y n_x - \dot{n}_x n_y), \quad (4)$$

where $\nabla = (\partial_x, \partial_y)$ and the term on the left contains the divergence of a two-component vector purely elastic in origin. From Eq. (4) several conclusions may be drawn. First of all, at steady state, $\dot{n}_x = \dot{n}_y = 0$ and, in the absence of the optical field, Eq. (4) reduces to

$$\nabla \cdot \boldsymbol{\theta}_d = 0. \tag{5}$$

This equation may be interpreted as expressing the balance, at the equilibrium, between the body torques, which develop in the bulk of LC, and the surface ones at the boundaries in the x, y-plane [10]. Besides, even in non-stationary conditions and in presence of the optical field, the left term in Eq. (4) vanishes upon integration over the x, y plane. Far from the illuminated region, in fact, the sample, due to homeotropic strong anchoring, remains perpendicular to the walls ($n_x = n_y = 0$) so that there is no net torque transferred to boundaries. Integrating Eq. (4) over the x, y plane, one obtains

$$\frac{\gamma L}{2}\left(\int dxdy\,(n_y\partial_t n_x - n_x\partial_t n_y) + i\int dxdy \sum_{k=x,y}\partial_t n_k \hat{L}_z n_k\right) = M_z \tag{6}$$

where the derivatives with respect to the ordinary time t have been restored, and M_z is the same as in Eq. (2). In the small distortion approximation the phase change accumulated by the optical field in traversing the sample, is given by

$$\Delta\psi(x,y) = \psi(x,y,L) - \psi(x,y,0) = \tilde{L}(n_x^2 + n_y^2), \tag{7}$$

where $\tilde{L} = \pi n_o \delta(L/2\lambda)$, $\delta = 1 - n_o^2/n_e^2$, and λ is the optical wavelength. According to equation (6), the angular momentum the light beam transfers to the homeotropically aligned LC sample may be balanced by bulk viscous forces only. Consequently, at steady state, since the viscous forces vanish, $M_z = 0$, and no angular momentum is transferred.

If the light beam impinging on the sample were polarized, an additional term should have been taken into account, namely $\int dxdy I(x,y)\Delta s_3$ due to the spin angular momentum transfer, where $\Delta s_3 = s_3(x,y,L) -$

$s_3(x, y, 0)$ is the polarization ellipticity change suffered by the beam in passing through the sample.

As a matter of fact, an NLC sample is an astigamtic and, at the same time, birefringent element, so capable to couple with both orbital and spin angular momenta. A closer inspection into Eq. (2) shows that M_z vanishes if either $\Delta\psi(x, y)$ or $I(x, y)$ depend on the radial coordinate $r = \sqrt{x^2 + y^2}$ only. For this reason, in our experiment we used two cylindrical lenses, one focusing in the x-direction and the other in the y-direction, to obtain an elliptical beam cross section at the sample position, so that both $\Delta\psi(x, y)$ and $I(x, y)$ were non-cylindrically symmetric. Moreover, M_z vanishes if $I(x, y)$ and $\Delta\psi(x, y)$ share the same symmetry axis in the x, y plane. This implies that the laser induced molecular distortion in the LC may evolve in time until the C-director $\boldsymbol{n}_C = (n_x, n_y)$ becomes either parallel or orthogonal to the major axis of the beam profile. Of the two equilibrium positions, the one with $\boldsymbol{n}_C$ parallel to the major axis of the intensity profile is stable, because it involves mainly twist deformation and, hence, it requires less elastic energy (the twist elastic constant is usually the lowest) [11].

THE EXPERIMENT

In our experiments the sample was a 50 μm thick E7 nematic film sandwiched between glass covers coated with DMOAP for homeotropic alignment. It was pumped by a frequency-doubled c.w. Nd:YAG laser source, working at $\lambda = 532$ nm. Two cylindrical lenses, assembled in the inner confocal configuration with their axes orthogonal to each other, were used to obtain an elliptical beam waist at the sample position. The focal lengths of the lenses were $f_x = 500$ mm and $f_y = 30$ mm in the x and y directions respectively. The beam radii in the common focal plane were found to be $w_x = 130$ μm and $w_y = 10$ μm, corresponding to a profile ellipticity $e \equiv w_x/w_y = 10$. All experiments were carried out using the

same lenses configuration. When required, a Pockels cell (PKC) was used to unpolarize the laser light [12]. The PKC fast axis was oriented at 45° with respect to the laser polarization plane and the cell was driven by a saw-tooth signal at λ-amplitude and a frequency of 200 Hz. This produced a periodic modulation of the light polarization trough the sequence of states shown in fig. 1. The time-averaged Jones matrix $J_{ij} = \langle E_i E_j^* \rangle$ of this varying polarization state is equal to the unit matrix (or, equivalently, all its average Stokes' parameters vanish) as for fully unpolarized light. The NLC orientational dynamics is much slower ($\simeq$ 1 s) than the modulation period, so that it will actually respond only to the time averaged Jones matrix (or Stokes' parameters) of the polarization. Therefore the laser beam emerging from the PKC is, for our purposes, completely equivalent to unpolarized light.

Our detection apparatus provides simultaneous and real time measurements of the angular aperture Θ and of the polarization direction angle Φ of the far-field self-diffraction pattern, which are formed beyond the LC sample when reorientation takes place [13]. For small LC distortion one approximately has $\Theta \propto \theta^2$ and $\Phi \simeq \phi$, where θ and ϕ are the polar coordinates of the molecular director $\boldsymbol{n}$, averaged over the sample [13]. The angles Θ and Φ provide roughly independent degrees of freedom for $\boldsymbol{n}$.

The present work is basically aimed at investigating the effect of the simultaneous transfer of both orbital and spin angular momentum in the reorientational processes in NLCs. Measurements with unpolarized light were accordingly performed just to check out that the orbital part of the light angular momentum tends to align the LC molecules along the major axis of the intensity profile. This alignment effect is shown in fig. 2, curve (a): when the unpolarized elliptically shaped laser beam is turned on, the director azimuthal angle ϕ reaches a steady-state having $\boldsymbol{n}_C$ along the intensity profile major axis (90° in the figure). Further details about the optical reorientation induced by unpolarized light beam with elliptical intensity profile were reported in Ref. [11], where it was also shown that the threshold intensity for the OFT with unpolarized light is roughly in-

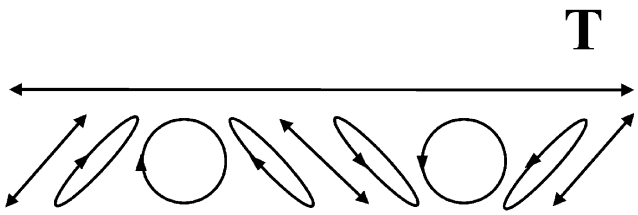

FIGURE 1 Polarization tate sequence in our artificially depolarized light.

dependent of the intensity profile ellipticity (for fixed spot area) and it is almost twice the threshold I_{th} for the OFT with linear polarization and circular beam cross-section ($I_{th} = 4.18$ kW/cm^2 for our sample). When the elliptically shaped laser beam is linearly polarized along the major axis of the beam intensity profile ($\alpha = 0°$), the spin and orbital parts of the light angular momentum cooperate and a steady state is reached with ϕ aligned along the ellipse major axis, as shown in fig. 2, curve (b). When the beam is polarized along a different direction ($\alpha \neq 0°$) a competition between orbital and spin angular momentum transfer is observed. For instance, if the beam is polarized at $\alpha = 45°$ with respect to the major axis of its intensity profile, the system reaches a steady state at an intermediate angle ϕ (fig. 2, curve (c)). We notice that, in spite of the higher incident power, the unpolarized light (fig. 2, curve (a)) is less effective in aligning $\boldsymbol{n}_C$ along the major axis than in curve (b), because in the former case the cooperating spin angular momentum is absent. The steady state in curve (a) is not exactly at $\phi = 90°$, because of the presence of residual small torques due to the unavoidable asymmetries of the system (small pretilt, not perfectly parallel sample walls, etc.) or to not perfect unpolarization of the incoming light [12].

When the beam polarization direction is along the the intensity profile minor axis ($\alpha = 90°$), we observed, above some intensity threshold, the onset of persistent oscillations of the molecular director. An example of this dynamical regime is shown in fig. 3. At higher laser power, the oscillations become less regular and apparently chaotic. The number and diameter of the far field diffraction rings were found changing in time as well. We tried also switching the laser polarization on and off: as expected, when the light became unpolarized, the oscillations were damped out and the system relaxed to the undistorted state if $I < 2I_{th}$ or to a steady distorted state if $I > 2I_{th}$. When the light polarization was restored, the previous oscillations were recovered (fig. 4) or, sometimes, the system was found to jump to a different regime (fig. 5). To our knowledge, the

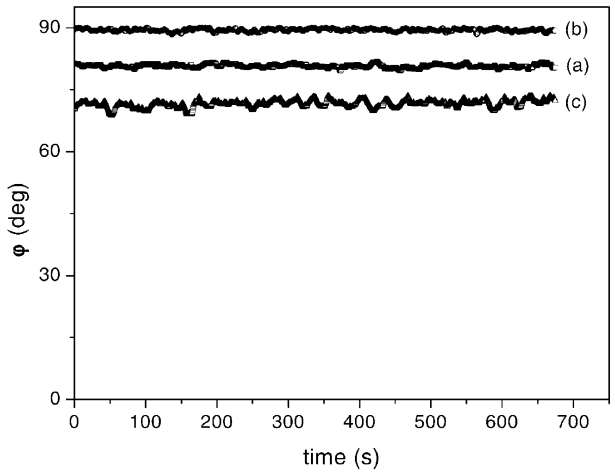

FIGURE 2 Steady states of he director azimuthal angle ϕ. (a) Unpolarized laser beam, $P = 480$ mW. (b) Linear polarization along the major axis of the intensity profile, $P = 300$ mW. (c) Linear polarization at $\alpha = 45°$ with respect to the major axis of the intensity profile, $P = 300$ mW.

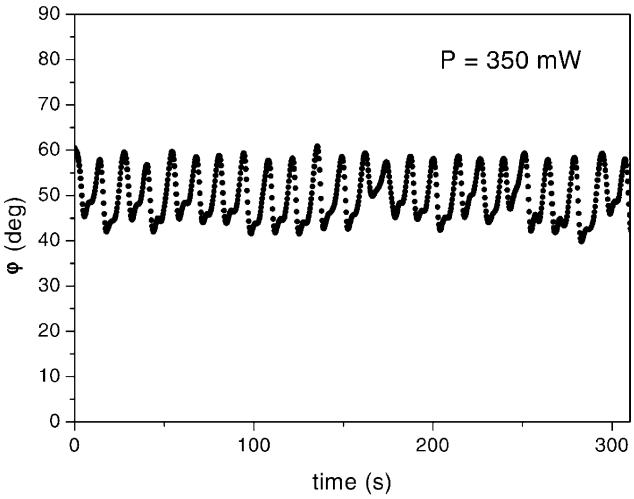

FIGURE 3 Example of persistent oscillations of the azimuthal angle ϕ of the molecular director.

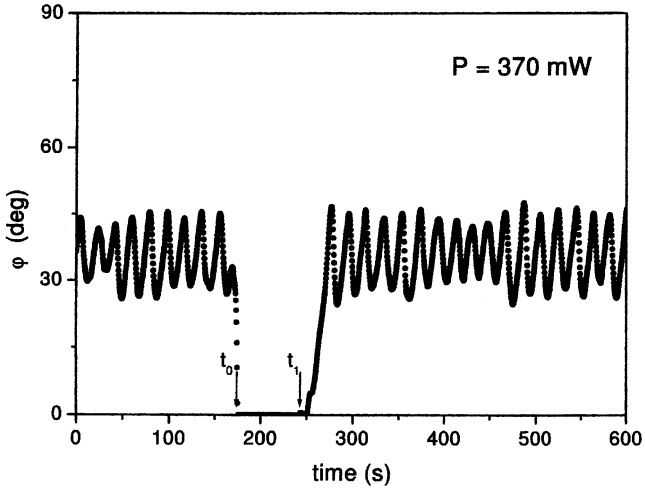

FIGURE 4 At time t_0 the laser polarization was switched to unpolarized and at time t_1 the polarization was restored. The system recovers the previous oscillating state.

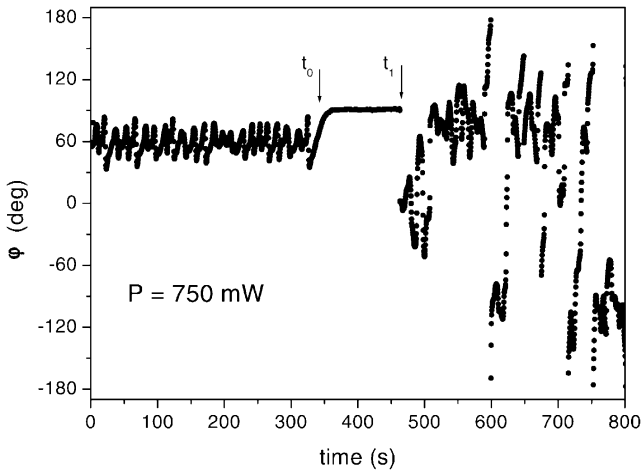

FIGURE 5 At time t_0 the laser polarization was switched to unpolarized and at time t_1 the polarization was restored. The system jumps to a different apparently chaotic regime.

only other case where laser-induced chaotic oscillations were observed in LC was the case of s-polarization at small incidence angle [14]. Recently, it was suggested that the route to chaos of the optical reorientation at oblique incidence may be governed by a new mechanism, namely the gluing of bifurcations [15]. We have presently not enough data to say if the apparent transition to chaos induced by competition of spin and orbital angular momentum of light follows the same route. We stress, however, that the model in Ref. [15] was based on the plane wave approximation, that certainly cannot be applied when the orbital angular momentum along z is transferred to the medium.

CONCLUSIONS

We have demonstrated that the z-component of both spin and orbital angular momentum can be transferred from a light beam to a LC sample. We derived Eq. (6) governing the total angular momentum conservation for the system *liquid crystal+optical field*, from which we proved that an elliptically shaped laser beam can be used to transfer orbital angular momentum to the medium. Using a laser beam linearly polarized at an angle α with respect to the major axis of its intensity profile, we studied in some detail the competition between the spin and the orbital parts of the light angular momentum transfer. A steady state is usually reached except in the case $\alpha \simeq 90^\circ$, where the onset of nonlinear irregular oscillations have been noticed. Our preliminary experiments suggest that the oscillation regimes can be obtained only for high enough intensity and ellipticity in the beam profile. A detailed study of all dynamical regimes as functions of the experimental parameters (I, e, α) will be the object of a forthcoming paper. Finally, it is worth observing that the transfer of orbital angular momentum is essentially due to the fact that the beam profile is non-cylindrically symmetric. This circumstance may happen in other strongly asymmetric configurations, such as, for instance, in the propagation of

the optical field trough an LC-filled waveguide. In this case, the optical torque arising from the orbital angular momentum transfer could be not negligible and eventually induce dynamical instabilities.

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