Orbital and Spin Photon Angular Momentum Transfer in Liquid Crystals

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All-optical angular control of the molecular alignment in liquid-crystal films is demonstrated using a laser beam having an elliptically shaped intensity profile. The material birefringence is unimportant, as proven by the fact that good alignment is obtained with unpolarized light. This raises the possibility of achieving optical angular control of transparent isotropic bodies. A general theoretical approach, based on light and matter angular momentum conservation, shows that the optical alignment is due to the internal compensation between the transfer of the orbital and the spin part of angular momentum of the incident photons to the material.

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Transfer of angular momentum from light to matter was proposed a long time ago by Beth [1] as an effective tool to achieve the angular manipulation of macroscopic objects. In Beth's experiment the photon spin angular momentum was transferred to a quartz wave plate producing a tiny torque that was measured. More recently, the same mechanism was used to induce rotation of liquid-crystal molecules [2,3] and laser-trapped microscopic calcite particles [4], raising the possibility of light-driven molecular motors [5]. Since only the spin part of the photon angular momentum is involved, Beth's mechanism requires birefringent materials and polarized light. Allen *et al.* [6] pointed out the possibility of an experiment to observe the torque produced by the transfer of the orbital part of photon angular momentum. This new mechanism is more interesting for applications because it raises the possibility of optically aligning and manipulating both isotropic and anisotropic transparent bodies using, eventually, unpolarized light. Transfer of the orbital angular momentum of light to opaque particles was recently achieved [7,8], using a laser beam oscillating in a high-order Laguerre-Gauss mode. Laguerre-Gauss laser beams, however, cannot transfer their orbital angular momentum to transparent materials [9]. Indeed, at present, no experiment is reported in the literature, in which the orbital angular momentum of photons is transferred to transparent bodies.

We demonstrate in this paper the possibility of achieving optical orientational control on transparent nematic liquid-crystal (NLC) film by transfer of the orbital angular momentum of photons. Liquid crystals were chosen because their strong birefringence provides a simple way to observe their rotation around the laser beam axis, just by looking at the polarization of the transmitted far field. Moreover, NLCs are affected by both the spin and the orbital angular momentum of light, providing good material to check the total angular momentum conservation law. We stress, however, that the material birefringence is not really important in our experiment, and that the same apparatus could be used, in principle, to optically manipulate isotropic transparent bodies also.

As shown in Ref. [9], in the paraxial optics approximation, the photon linear and orbital angular momentum components can be represented by the usual quantum mechanics operators $\hat{p}_i = -i\hbar\partial_i$ $(i = x, y, z)$ and $\hat{L}_z = -i\hbar(x\partial_y - y\partial_x)$ acting on Jones' ket $|J\rangle = (E_x, E_y)$. The rapidly varying factor $\exp(-i\omega t + ikz)$ in the optical field was suppressed. In the same approximation, the photon spin component operators are $\hat{S}_{x,y,z} = \hbar\hat{\sigma}_{1,2,3}$, where $\hat{\sigma}_i$ are the Pauli matrices [10], and the total photon angular momentum operator along the beam axis is $\hat{J}_z = \hat{L}_z + \hat{S}_z$. For a given operator $\hat{O}$, besides the local average $\langle\hat{O}\rangle = \langle J|\hat{O}|J\rangle$, it is useful to consider averages $\mathcal{O}(z) = \int dxdy\,\langle J|\hat{O}|J\rangle / \int dxdy\,\langle J\,|\,J\rangle$ over the x, y plane. With these definitions, the average spin z-component $S_z(z)$ of the photons crossing the plane $z =$ const is given by $S_z(z) = \hbar\bar{s}_3(z)$, where $s_3(x, y, z)$ is the reduced Stokes parameter of the local polarization of the beam, and the bar means the average over the transverse intensity profile $I(x, y, z) = (c/8\pi)\langle J\,|\,J\rangle$. Assuming that all media are transparent, the power $P = \int dxdy\, I(x, y, z)$ carried by the beam is independent of z. Transparent optical components along the beam path are represented by unitary operators $\hat{U}$ acting on $|J\rangle$. In particular, a NLC film can be considered as a thin (thickness $L \ll \pi w_0^2/\lambda$, the diffraction length associated with the beam waist w_0) inhomogeneous birefringent plate. The operator of such a plate is $\hat{U} = \exp[i(\psi_0 + \alpha/2)]\exp[i\alpha/2(\hat{\sigma}_1\cos 2\phi + \hat{\sigma}_2\sin 2\phi)]$, where ψ_0 is the phase change suffered by the ordinary wave in traversing the plate, α is the phase difference between the extraordinary and ordinary waves, and ϕ is the azimuthal angle of the optical axis with respect to the laboratory x axis. In NLC, the optical axis is along the molecular director $\boldsymbol{n} = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$. In principle, ψ_0, α, and ϕ may all depend on the x and y coordinates. For our purposes, it is enough to assume that $\alpha = \alpha(x, y)$ and constant ψ_0 and ϕ. Near the center of the beam, the optical phase difference α induced in the NLC is maximum, so we approximate $\alpha(x, y)$ with the parabolic profile $\alpha(x, y) = \alpha_0 - \frac{1}{2}B_{ij}r_ir_j$ $(r_1 = x, r_2 = y)$, the

2×2 matrix B_{ij} being defined positive. As shown in Ref. [9], the average total angular momentum change $\Delta J_z = \Delta S_z + \Delta L_z$, suffered by each photon traversing the plate, is obtained by averaging over the input ket $|J^{\rm in}\rangle$, and over the x, y plane the operator $\delta\hat{J}_z = \hat{U}^\dagger[\hat{J}_z, \hat{U}]$. The total angular momentum transferred to the medium per unit time (i.e., the torque acting on it) is $-F\Delta J_z$, where $F = P/\hbar\omega$ is the photon flux. Evaluating the relevant commutator, we calculated the average photon spin change as

$$\Delta S_z = \hbar[(1 - s_{30}^2)^{1/2}\overline{\sin\alpha}\sin 2(\psi - \phi) - s_{30}(1 - \overline{\cos\alpha})], \tag{1}$$

where the bar means the average with respect to the beam intensity profile $I(x, y)$ at the sample plane, and s_{30} and ψ are the ellipticity and angle of the incident beam polarization, respectively. In a similar way, we found the change in the average orbital photon angular momentum,

$$\Delta L_z = \hbar b g \sin 2(\gamma - \beta)\cos^2(\psi - \phi), \tag{2}$$

where β and γ are the angles between the fixed x axis and the major axis of the ellipse associated with the $\alpha(x, y)$ and the $I(x, y)$ profiles, respectively, b and g being the respective anisotropy constants. These two ellipses are defined by the equations $B_{ij}^{-1}\xi_i\xi_j = 1$ and $G_{ij}^{-1}r_i r_j = 1$, where $G_{ij} = \overline{r_i r_j}$ is the matrix of the second moments of the beam intensity profile $I(x, y)$ at the sample position. The positive constants b and g are defined as half the absolute difference between the eigenvalues of B_{ij} and G_{ij}, respectively. In the case of unpolarized light, Eq. (1) yields $\Delta S_z = 0$ and Eq. (2) reduces to

$$\Delta L_z = \tfrac{1}{2}\hbar b g \sin 2(\gamma - \beta). \tag{3}$$

Equation (3) also holds for isotropic materials, provided $\alpha(x, y)/2$ is replaced by the phase profile $\psi_0(x, y)$. In Eqs. (1)–(3), ψ, s_{30}, and γ are the control parameters. If the plate is rigid, the difference $\beta - \phi$ is constant, and ΔJ_z depends on one angular coordinate, say, $\Delta J_z = \Delta J_z(\phi)$. In the case of NLC, the minimum elastic energy is reached when $\beta = \phi$, because, in this configuration, the prevailing molecular distortion is the twist distortion, having the lowest elastic constant [11]. Therefore, we assume, throughout this paper, $\beta = \phi$. For a beam having a circular cross section (which applies, for example, to Hermite-Gauss and Laguerre-Gauss laser modes), $g = 0$ and, hence, $\Delta L_z = 0$, as mentioned above. If the plate is immersed in a high viscosity fluid, its equation of motion is $\eta\dot{\phi} = \Delta J_z(\phi)$, where $\dot{\phi}$ is the angular velocity and η is the viscosity coefficient. The body equilibrium condition is $\Delta J_z(\phi) = 0$ and thus ΔL_z and ΔS_z must exactly compensate (or both be zero). The NLC samples used in our experiments are not rigid bodies and their interaction with light is nonlinear: nevertheless, it can be shown that, for homeotropic strong anchoring at the sample walls, ΔJ_z is still balanced by viscous torque only (although the mathematical expression of this torque is more complicated) and the equilibrium condition is still $\Delta J_z = 0$. The equilibrium condition cannot be met in general. For example, in the case of circular cross section and circular polarization of the incoming beam, Eqs. (1) and (2) yield $\Delta L_z = 0$ and $\Delta S_z = \pm\hbar(1 - \overline{\cos\alpha}) \neq 0$ per photon, and the body is forced to continuously rotate, as reported in Refs. [2,4]. To achieve the angular control of the NLC alignment with circularly polarized light, two beams must be used having opposite handedness [5]. One-beam angular control can be obtained by making the beam cross section elliptical, so that $g \neq 0$. This should also work with isotropic bodies (or with unpolarized light). Anisotropic materials, such as NLC, are affected by both the spin and the orbital angular momentum of light, so we may control the reorientation plane of the sample by changing ΔS_z and ΔL_z separately, as shown in this paper.

In our experiments, we used a frequency doubled cw Nd:YAG laser source, working at $\lambda = 532$ nm. The sample was a 50-μm-thick E7 nematic film sandwiched between glass covers coated with octadecyldimethyl(3-trimethoxy-silylpropyl)ammonium chloride (DMOAP) for strong homeotropic alignment. The laser intensity profile at the sample position was made elliptical by using two confocal cylindrical lenses, having focal lengths $f_x = 500$ mm and $f_y = 30$ mm in the x and y directions, respectively. The beam radii ($1/e^2$ intensity) at the lens common focal plane were found to be $w_x = 130$ μm and $w_y = 10$ μm, corresponding to a profile ellipticity $\mu = w_x/w_y = 13$. The ellipticity of the intensity profile could be reduced to $\mu \simeq 1$ by moving the sample away from the confocal plane. When required, we used a Pockels cell to unpolarize the laser light, as described elsewhere [12].

Our detection apparatus was already described in detail [11], and we sketch here its main features only. A rotating polarizer was used to provide simultaneous and real time measurements of the angular aperture Θ and of the polarization direction angle Φ of the far-field self-diffraction ring pattern, which is formed beyond the NLC sample when reorientation takes place. For small LC distortion we have approximately $\Theta \propto \theta^2 \propto \alpha$ and $\Phi \simeq \phi$, where θ and ϕ are the polar coordinates of the molecular director $\boldsymbol{n}$, averaged over the sample thickness [11]. The angles Θ and Φ provide roughly independent degrees of freedom of $\boldsymbol{n}$. We made measurements using unpolarized as well as linearly polarized light. In the case of the unpolarized light, when the laser power P exceeded the threshold P_{th} for the optical Fréedericksz transition (OFT), the steady-state reorientation plane was found almost independent of P and parallel to the major axis of the beam intensity profile, as expected from Eq. (3). (This is shown in Fig. 1.)

The data were obtained by rotating the cylindrical lens gauge (and hence the intensity profile) to $\gamma = 30°$ with respect to the horizontal plane. Similar results were obtained at different angles γ, thus proving the possibility of achieving angular control by transfer of the orbital photon angular momentum only. Liquid crystals are birefringent

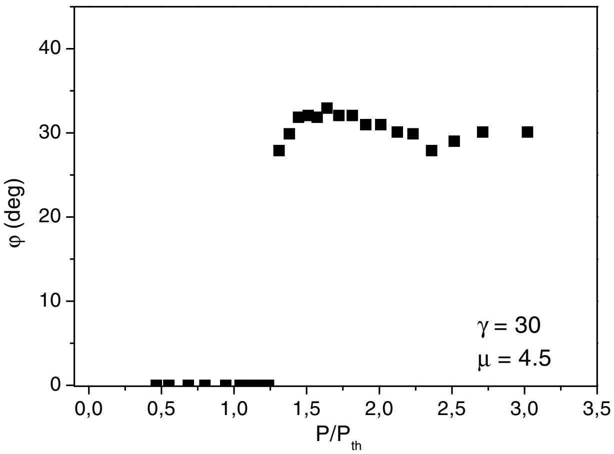

FIG. 1. The steady-state azimuthal angle ϕ of the NLC reorientation plane as a function of the laser power. The laser was unpolarized and its intensity profile was elliptical with ellipticity $\mu = 4.5$ and with the major axis set at $\gamma = 30°$ with respect to the horizontal plane.

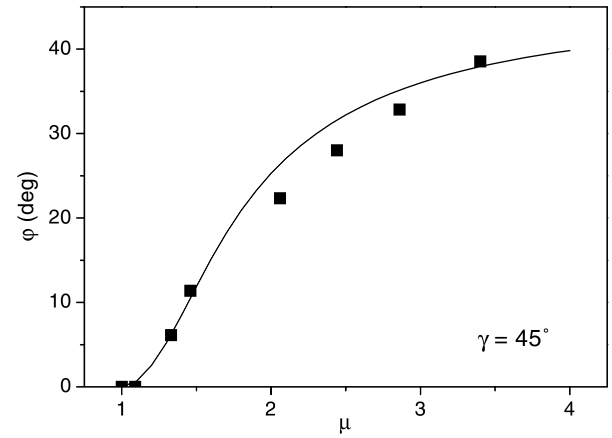

FIG. 2. The steady-state azimuthal angle ϕ of the NLC reorientation plane as a function of the beam profile ellipticity. The laser beam was linearly polarized and the major axis of the intensity profile was set at $\gamma = 45°$ with respect to the beam polarization direction. The solid curve is the best fit obtained from Eqs. (1) and (2).

and, hence, they are sensitive to photon spin as well. In order to study the competition between the two types of photon angular momentum, we used a linearly polarized laser beam having its elliptical profile at angle γ with respect to the polarization direction $\psi = 0$. In this case, the relative ratio between ΔS_z and ΔL_z is governed by the dimensionless parameter $B = \overline{\sin\alpha}/bg$. Changing B provides a way to achieve one-beam angular control by exploiting the internal compensation between the spin and the orbital angular momentum of photons.

In Fig. 2 we reported the angle ϕ of the NLC reorientation plane at steady state as a function of the beam profile ellipticity. The figure refers to a beam intensity profile at $\gamma = 45°$ with respect to the laser polarization direction. In obtaining these data, the sample was moved along the beam to change the intensity profile ellipticity $\mu = w_x/w_y$ at the NLC position. The laser power P was simultaneously adjusted to keep the peak intensity $I = 2P/\pi w_x w_y$ constant. The constant intensity condition was checked by looking at the far-field pattern beyond the sample: all points in Fig. 2 correspond to about one diffraction ring in the far field. The solid curve is the best fit to the data, obtained by solving $\Delta J_z(\phi) = 0$ as given by Eqs. (1) and (2). In the calculations we assumed that, at steady state, the elastic energy was minimum (i.e., $\beta = \phi$), and that the phase profile $\alpha(x,y)$ had approximately the same ellipticity of the intensity profile $I(x,y)$, so that $bg \simeq (\mu^2 - 1)^2/4\mu^2$. As a fit parameter we used $\overline{\sin\alpha}$, obtaining $\overline{\sin\alpha} = 0.378$. We notice that, for high enough profile ellipticity μ, the transfer of the orbital angular momentum of photons always prevails and the angle ϕ tends to be aligned along the intensity profile major axis. This is due to the fact that the spin transfer ΔS_z is bounded to $\pm 2\hbar$ per photon, while ΔL_z can be made large at will by increasing g. Similar results were obtained at other values of γ. In all cases, we checked that, for linear polarization, the power threshold to induce the OFT was about one-half the threshold measured in the case of unpolarized light, confirming previous results [12]. At $\gamma \simeq 90°$, and for large enough intensity profile ellipticity ($\mu > 5$), when the laser power exceeded a second critical threshold, we observed the onset of persistent time-dependent oscillations of the reorientation plane. (An example is shown in Fig. 3.)

As the laser intensity is increased, the oscillations became more and more irregular until an apparently chaotic dynamical regime is observed. The phenomenon is interesting in itself and was never observed before.

The only reported cases of optically induced nonlinear oscillations in NLC, in fact, are the OFT induced by elliptically polarized light [3] and the OFT induced by an s-polarized laser beam at small incidence angle [13].

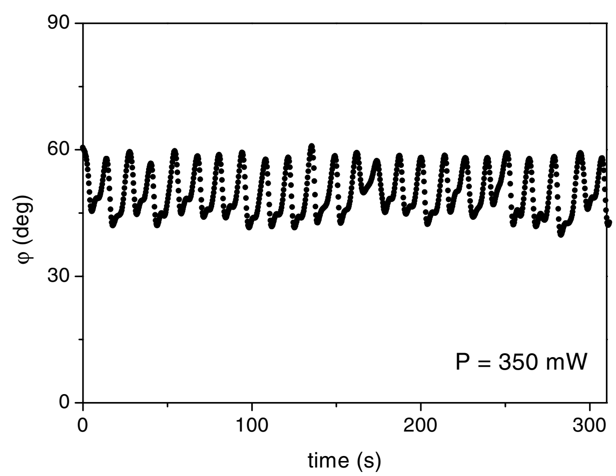

FIG. 3. Example of persistent time oscillations of the azimuthal angle ϕ of the molecular director. The laser beam was linearly polarized and the major axis of the intensity profile was set at $\gamma = 90°$ with respect to the beam polarization direction. The laser incident power was $P = 350$ mW.

At present, the mechanism underlying the onset of the observed oscillations is not clear, and the phenomenon will be the object of further study. We think, however, that Eqs. (1) and (2) may still retain some validity in the time-dependent case also. However, because the elastic energy must not be minimum out of equilibrium, the condition $\beta \simeq \phi$ may now be violated, thus adding a further degree of freedom. Developing a model to explain the occurrence of nonlinear oscillations and, eventually, deterministic chaos in our experimental configuration is certainly a difficult task, because the plane-wave approximation cannot be exploited. In particular, the recent model based on gluing of bifurcations [14] cannot be extended to the present case.

In conclusion, we proved that the alignment plane of NLC films above the threshold for the OFT can be controlled by using a laser beam having an elliptical cross section, so that the orbital angular momentum of photons could be transferred to the medium. Good alignment was obtained by using unpolarized light, showing that the sample birefringence is unimportant, thus raising the possibility of achieving angular control of isotropic bodies. The laser incident power is not critical in the alignment process. We also made measurements using linearly polarized light. In this case, the internal competition between the orbital and the spin part of photon angular momentum drives the system along a plane that can be controlled by acting on the ellipticity of the beam intensity profile. The experimental results are in good agreement with a simple model, where the NLC film is considered as an optically thin inhomogeneous birefringent plate. When the intensity profile major axis and the light polarization direction are perpendicular to each other, the alignment plane of the NLC was found to oscillate continuously. At very high laser power, the oscillations became apparently chaotic. This unexpected and intriguing phenomenon will be the object of future work.

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$$\hat{\sigma}_1 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}; \; \hat{\sigma}_2 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}; \; \hat{\sigma}_3 = \begin{pmatrix} 0 & i \\ -i & 0 \end{pmatrix}.$$

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