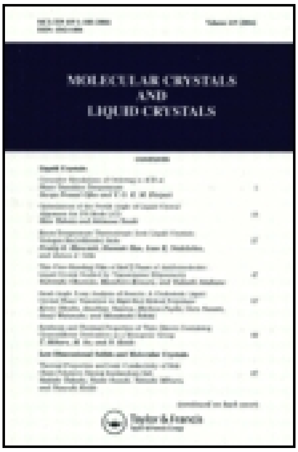

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Optical Manipulation in Liquid Crystals: Effects of Tight Focusing on Nonlinear Optical Reorientation

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We report a detailed analysis of the problem of nonlinear optical reorientation induced in a nematic liquid crystal by a tightly focused Gaussian beam under experimental conditions that prevent the effect of conventional trapping originated by optical gradient forces. Novel features arise under these conditions: no threshold effect; reduced reorientation if compared the one induced by a paraxial Gaussian beam, orientational singularity at the focal waist.

Keywords Nonlinear optics, optical trapping, nonparaxial beams

Introduction

The long range molecular interaction typical of liquid crystals gives rise to interesting phenomena when microsize silica particles are dispersed in these materials. In particular forces acting on the beads have been investigated studying the dependence of the interaction on geometrical parameters and on the topological defects around the particles [1]. The interaction is originated by the director distortion around the particles that propagates due to the elasticity of liquid crystal. In this way forces act on the particles in order to reduce the induced distortion and to minimize the elastic free energy of the liquid crystal. As a consequence self-assembled structures of microparticles can be formed. A peculiar effect arises when one particle interacts with the director reorientation induced by light. In this case even if the required conditions for optical trapping are not fulfilled the microbead moves towards the optical beam and get trapped on the edge of it. This unusual behaviour has been observed for the first time by Musevic and coworker [2]. We have pointed out that the nonlinear optical reorientation of liquid crystal plays a crucial role in determining the properties of this phenomenon [3,4].

As a matter of fact the experimental conditions necessary to observe this unconventional trapping require strong focusing on the liquid crystal sample, on the other hand nonlinear optical reorientation has never been investigated under this frame before the appearance of few recent papers [5–7]. In fact even when Gaussian beams have been considered instead of plane waves, the paraxial approximation has been always taken into account in order to keep negligible any depolarization effect in the focal waist. On the contrary strong focusing

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of a linearly polarized beam used to get nonlinear orientation of liquid crystals gives rise to complex features of the optical field in the focal area and unexpected features of the reorientation may be foreseen.

In this paper starting from the observation of optical trapping of microsize particles we address the problem of nonlinear optical reorientation induced in a nematic liquid crystal by a tightly focused Gaussian beam underlining the novel features arising under these conditions and comparing them to the behaviour expected for a Gaussian beam under paraxial approximation.

The Experiment

Experiments were performed using 50-μm thick cells filled by a mixture of spherical silica particles with average radius $r_b = 2.5$ μm and refractive index $n_s = 1.37$, dispersed in the nematic liquid crystal pentyl-cyanobiphenil (5CB). The refractive indices of 5CB at $\lambda = 532$ nm are $n_o = 1.54$ and $n_e = 1.71$. The surfactant N,Ndimethyl- noctadecyl-3-aminopropyl-trimethoxysilyl chloride (DMOAP) was used to coat the particles surface [3], in order to get strong homeotropic anchoring at the particle surface. Cell substrates were coated by DMOAP to induces homeotropic alignment in the samples. An inverted microscope configuration conventionally used in optical tweezers was used with a cw laser beam (frequency-doubled Nd:YVO_4 at $\lambda = 532$ nm) focused on the sample.

According to the theory of conventional optical trapping when the particle refractive index is lower than the one of the surrounding material the overall force acting one the particle is repulsive pushing it away from the focal spot, therefore in our case we should not expect any trapping phenomenon. However the liquid crystal molecules around the bead might change its effective refractive index with the result of having a particle "dressed" that would appear with a higher refractive index than the one of the homeotropic surrounding material thus fulfilling the conditions of the conventional optical trapping. In order to get rid of this problem we have used an objective with a limited numerical aperture NA = 0.45, that is high enough to get strong depolarization effect in the focal waist and, at the same time, low enough in order to avoid trapping by gradient forces. At the same time we have used underfilling conditions not favourable for conventional trapping. The theory developed by Ashkin [8] allows us calculating the gradient and the scattering forces under the geometrical optical approximation that is satisfactory for our case since we have $r_b \sim 5\lambda$. By considering a dressed particle such that m = 1.1, being m the ratio between its refractive index and the ordinary one of the liquid crystal, with the experimental filling factor $b = 0.5$ we found the balance between scattering force and gradient force always positive in z direction, that is the net effect of the light on the particle should be a push away from the focal waist [5]. Nevertheless trapping of silica particles was observed for all values of the used incident optical power, pointing out that a different physical effect must play a fundamental role. In Fig. 1 typical trajectories of the particle towards the area reoriented by the focused beam are shown. From this plot it is possible to get the force acting on the particle vs the distance r, that is shown in the inset.

Force is calculated following the usual approach [3,9], assuming that it is nearly balanced by the drag force $F_D = 6\pi r_b \eta \partial r/\partial t$, being η the medium viscosity. In fact, the measured inertial force is negligible with respect to the viscous drag due to the small value of the particle mass ($M \approx 10^{-13}$ Kg). Moreover, the particle motion is highly viscously damped, allowing a direct determination of the attractive force from the displacement data by differentiating them to obtain the particle speed. For the viscosity, the expression $\eta = 1/2\alpha_4$ has been used, where α_4 is the Leslie coefficients to be considered for flow orthogonal to

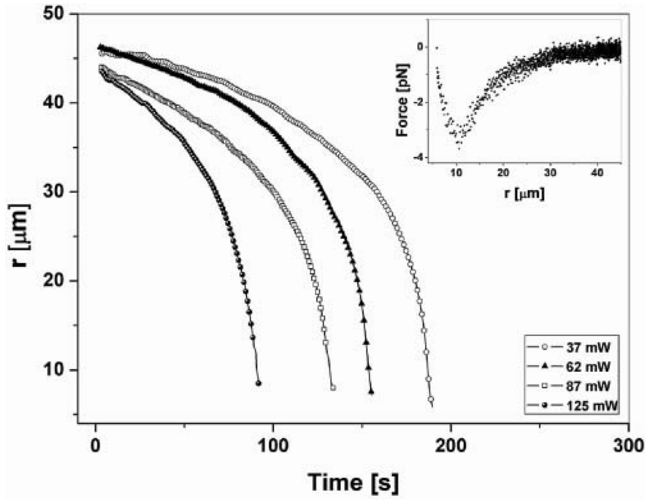

Figure 1. Positions of the particle vs time during trapping experiments. The final value of *r* is the equilibrium particle distance r_{eq} from the centre of the trap.

the director, giving rise to a relevant tilt of the molecular orientation [10]. In case of 5CB we get $\eta = 0.028$Pas.

A satisfactory fit of the experimental data is given by a function:

$$F = -\frac{A}{r^2} + \frac{B}{r^3}$$

The dependence of this force on the optical reorientation occurring in the focal area has been discussed in a previous paper [5].

It is quite interesting to compare the force due to the interaction between a particle and the distorted area to the one that has been observed between two colloids. It is amazing that in our case we have a Coulomb like behaviour scaling as $1/r^2$ while in case of two colloids a $1/r^4$ dependence is observed as expected from the interaction of two dipolar defects. On the other hand the repulsive part scales in our case as $1/r^3$ while the expected behaviour for two colloids scales as $1/r^6$ [11,12]. Therefore it is an experimental evidence that the interaction between an optically reoriented area and one colloid is much stronger than the one between two colloids and it leads to a longer range of interaction. Since the colloids interaction is explained in terms of topological defects and depends on their mutual symmetry, one basic question to be answered about this behaviour concerns the description of the light induced defect in the focal waist, originated by the optical reorientation of the molecular director.

Optical Reorientation Induced by a Focused Beam

These observations point out the necessity of a novel approach to the old problem of optical reorientation (namely the Giant Optical Nonlinearity discovered in 1980 [13]) which has been treated most of time under the plane wave approximation and only few times under the Gaussian beam paraxial approximation, needed to explain nonlocality effects [14,15] and wavefront curvature effects [16,17]. Therefore the depolarization effect occurring in a tightly focused beam was never considered, while it certainly strongly affects the director reorientation in case of tight focusing.

The very first step to be done is to follow the usual method of minimization of the free energy of the liquid crystal including the interaction with the field. As usual, the free energy density of the system includes contributions due to the elastic and the optical torques [18]:

$$F_{tot} = \frac{K_1}{2}(\vec{\nabla}\cdot\hat{n})^2 + \frac{K_2}{2}(\hat{n}\cdot\vec{\nabla}\times\hat{n})^2 + \frac{K_3}{2}(\hat{n}\times\vec{\nabla}\times\hat{n})^2 - \varepsilon_\perp\frac{|E|^2}{4} - \frac{\Delta\varepsilon}{4}(\hat{n}\cdot\vec{E})(\hat{n}\cdot\vec{E}^*) \tag{1}$$

where K_i are the elastic constants, **n** is the director, $\varepsilon_\perp$ is the ordinary dielectric constant, $\vec{E}$ is the optical field and $\Delta\varepsilon$ is the dielectric anisotropy at optical frequencies.

When a Gaussian beam is considered in the paraxial approximation, the electric field polarized along the **x** axis is given by:

$$\vec{\mathrm{E}} = E_0\frac{\omega_0}{\omega(z)}e^{\frac{-r^2}{\omega^2(z)}}e^{-i[kz+\frac{kr^2}{2R(z)}+tg^{-1}(\frac{z}{z_0})]}\cdot\vec{x} \tag{2}$$

and keeps is linear polarization for any value of z that corresponds to the axial direction of propagation. In Eq. (2) the parameters have the usual meaning.

Keeping the cylindrical symmetry, the dependence on the azimuthal angle is usually neglected and minimization is performed with respect to the tilt angle θ:

$$\frac{\partial}{\partial r}\frac{\partial F_{tot}}{\partial\vartheta_r} + \frac{\partial}{\partial z}\frac{\partial F_{tot}}{\partial\vartheta_z} - \frac{\partial F_{tot}}{\partial\vartheta} = 0 \tag{3}$$

where subscripts stand as derivatives with respect to the correspondent variable. It results in the following differential equation for the tilt angle θ:

$$\vec{\mathrm{E}} = K\left[\vartheta_r + r\vartheta_{rr} + r\vartheta_{zz}\right] + \frac{\Delta\varepsilon}{4}\mathrm{E}_0^2 r\frac{\omega_0}{\tilde{\omega}^2(z)}e^{\frac{-2r^2}{\tilde{\omega}^2(z)}}\sin(2\vartheta) = 0 \tag{4}$$

where $\tilde{\omega}(z) = \omega(z - \frac{d}{2})$ is the beam waist variation in the propagation bearing in mind that the beam is focused in the sample centre.

This differential equation is usually solved in case of strong anchoring ($\vartheta(r,0) = \vartheta(r,d) = 0$), taking into account that a maximum of the reorientation is expected at r = 0 where the field is maximum ($\vartheta_r(0,z) = \vartheta(\infty,z) = 0$). This problem has been previously solved [16] by choosing the following function for field above the Optical Freedericksz Transition (OFT):

$$\vartheta(r,z) = R(r)\sin\left(\frac{\pi z}{d}\right) \tag{5}$$

The simulation that we have carried out, later compared to the tight focusing beam condition, drops this approximation and allows for a full numerical integration of the above equation. Anyway the main peculiarities of the optical reorientation due to the paraxial Gaussian beams has been shown to be a strong nonlocalization of the director reorientation [14], a change in the OFT threshold [15], a double ring structure of the Self-Phase Modulation pattern [16,17].

Nevertheless this approach appears no more satisfactory when dealing with tightly focused beams due to the strong depolarization effects in the focal waist.

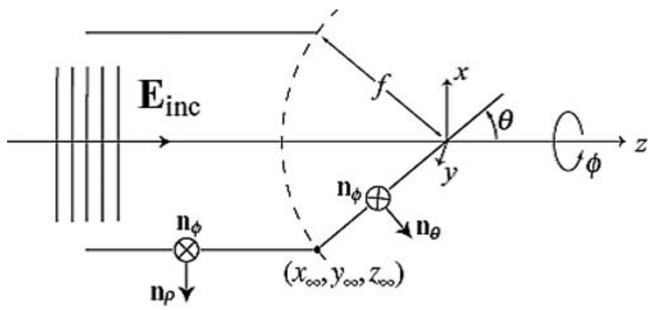

Figure 2. Geometrical representation of the aplanatic system and definition of coordinates.

In fact it has been shown that a linearly polarized optical field is transformed in the focal waist of a focusing lens in the following way [19]:

$$E(\rho, \varphi, z) = \frac{ikf}{2}\sqrt{\frac{n_1}{n_{eff}}}E_0 e^{-ikf}\begin{bmatrix} I_{00} + I_{02}\cos 2\varphi \\ I_{02}\sin 2\varphi \\ -2i I_{01}\cos\varphi \end{bmatrix} \tag{6}$$

where the used cylindrical coordinates are shown in Fig. 2 [19] and I_{00}, I_{01}, I_{02} are integral function defined in the following way:

$$\begin{aligned} I_{00} &= \int_0^{\theta_{\max}} f_w(\theta)\sqrt{\cos\theta}\,\sin\theta(1+\cos\theta)J_0(k\rho\sin\theta)e^{(ikz\cos\theta)}d\theta \\ I_{01} &= \int_0^{\theta_{\max}} f_w(\theta)\sqrt{\cos\theta}\,\sin^2\theta J_1(k\rho\sin\theta)e^{(ikz\cos\theta)}d\theta \\ I_{02} &= \int_0^{\theta_{\max}} f_w(\theta)\sqrt{\cos\theta}\,\sin\theta(1-\cos\theta)J_2(k\rho\sin\theta)e^{(ikz\cos\theta)}d\theta \end{aligned} \tag{7}$$

being J_n the nth-order Bessel function and $f_w(\theta) = \exp(-f^2\sin^2\theta/w_0^2)$.

The first step to be done to improve the model of the optical reorientation induced by a Gaussian beam is the use of this field in the minimization Equation (3).

The first approximation approach to solve the problem is to keep the cylindrical symmetry, namely to neglect the azimuthal angle dependence of the director reorientation. This is actually questionable when using high Numerical Aperture objectives, however in our case (N.A. = 0.45) the calculation of the field pattern in the focal area shows that the field distribution is weakly dependent on ϕ [5]. This make reasonable our approximation, therefore the differential equation coming from the free energy minimization reduces to:

$$\begin{aligned} &K(\theta_r + r\theta_{rr} + r\theta_{zz}) + \frac{\Delta\varepsilon}{16}k^2 f^2 \frac{n_1}{n_{eff}} r E_0^2[(D-H)\sin 2\theta + 2F\cos^2\theta] \\ &= \frac{\Delta\varepsilon}{16}k^2 f^2 \frac{\phi_1}{n_{eff}} r E_0^2 F \end{aligned} \tag{8}$$

keeping the one elastic constant approximation. Here D,H,F express constant parameters including the integral function I_{ii}:

$$\begin{aligned} D &= |I_{00}|^2 + |I_{02}|^2 + (I_{00}I_{02}^* + I_{02}I_{00}^*) \\ H &= 4|I_{01}|^2 \\ F &= 2i(I_{02}I_{01}^* - I_{02}^*I_{01} + I_{00}I_{01}^* - I_{00}^*I_{01}) \end{aligned} \tag{9}$$

By comparing this equation to the one obtained in the paraxial case we see that it is not homegeneous and the nonlinear part includes different terms, which make the related physics more complex. However a few remarks can be done to underline what can be expected in this case. We know that the optical torque due the field on the director can be expressed by:

$$\Gamma_{opt} = \frac{\Delta\varepsilon}{2}\mathrm{Re}(\hat{n}\cdot\vec{E}^*)(\hat{n}\times\vec{E}) \tag{10}$$

that for a homeotropic cell ($\hat{n}//\hat{z}$) becomes:

$$\Gamma_{opt} = \frac{\Delta\varepsilon}{2}\mathrm{Re}(-E_z^* E_y \hat{x} + E_z^* E_x \hat{y}) \tag{11}$$

In case of a plane wave with linear polarization in the x direction the initial torque is zero and this gives rise to the threshold effect known as Optical Freedericksz Transition, since orientation becomes effective when an extraordinary wave appears in the medium.

On the contrary looking at the expression (6) of the field, with strong focusing we do not expect any more to have a reorientation threshold since all the three components of the field are present from the beginning. When a linearly polarized plane wave or a Gaussian paraxial beam impinges on a standard homeotropic cell the longitudinal component of the field appears over the OFT threshold: in other words as long as we have only a ordinary wave in the medium we do not get any reorientation. This is the classical situation where a threshold behaviour is observed. On the other hand the tight focusing condition gives rise to field depolarization in the focal waist so that the component E_z is always present. As a consequence we should expect in this case a no-threshold behaviour.

A second interesting feature that makes different the not paraxial case is the behaviour of the optical torque Γ_y (we can neglect Γ_x due to the holding condition $E_y \ll E_x$). It changes sign at the focal plane as shown in Fig. 3 where calculation has been made for a focusing objective with N.A. = 0.45. As a consequence we may expect a peculiar behaviour in the molecular reorientation at z = 0.

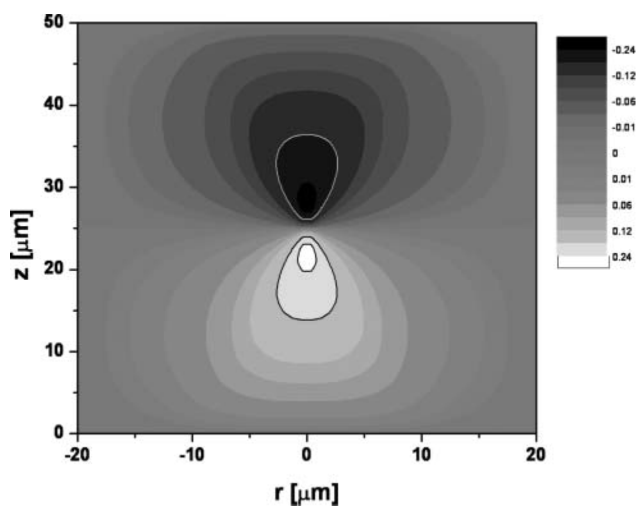

Figure 3. Initial optical torque along y direction (Γ_y) using an objective with NA = 0.45 and P = 1 mW.

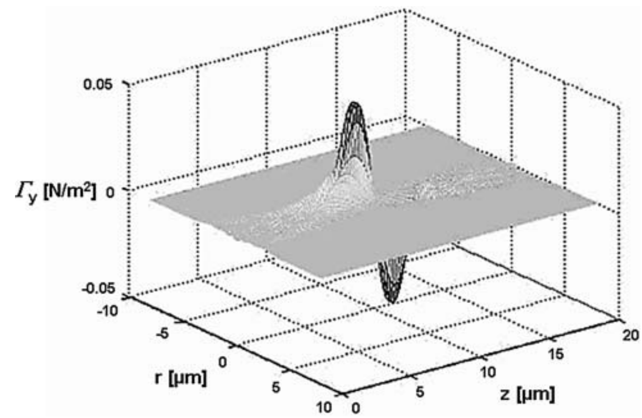

Figure 4. Contour plot of the liquid crystalline director distortion $\theta(r, z)$ induced by a focused Gaussian beam in case of N.A. = 0.45 and P = 30 mW. The values reported in the colorbar are expressed in radians.

This is actually found by integration of the Equation (8), allowing us to calculate the reorientation tilt angle vs optical power P as shown in Fig. 4 for a fixed power. Here the singularity at z = 0 is evident, where due to change of sign of the optical torque, no reorientation occurs. This phenomenon is similar to what observed by Brasselet [6] using circular polarization and tight focusing on nematic liquid crystals.

In Fig. 5 we report the maximum reorientation angle θ_{max} vs P, calculated for paraxial Gaussian beam and for a tightly focused beam, showing that in the last case we have a slower increase of reorientation and overall weaker effect.

While a complete demonstration of this theoretical results has not been carried out yet due to several experimental problems occurring in the tight focusing geometry a first easy check can be done by comparing the area of the molecular reorientation in the two cases. This

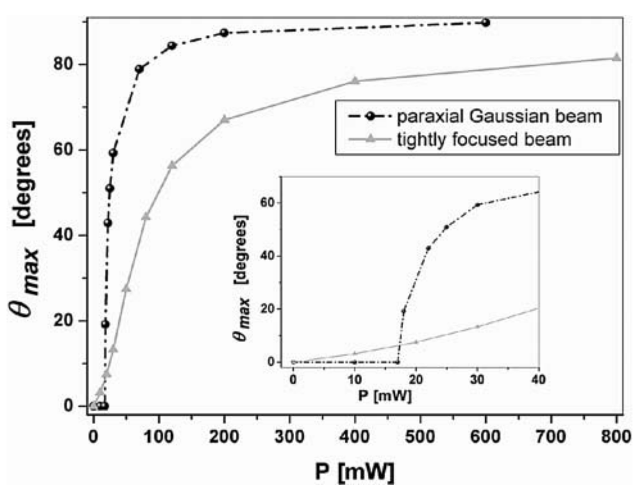

Figure 5. Maximum reorientation angle vs power. The inset shows a magnified graph at the low powers.

is actually the most relevant macroscopic effect of the Gaussian beam as compared to the plane wave: the nonlocality due to liquid crystal elasticity, i.e. the molecular reorientation is wider than the beam cross section. The reoriented area can be detected and measured by microscope observation through crossed polarizers: the onset of a bright field means that director has rotated. This comparison is reported in fig. 11 of reference [5] where the diameter of reoriented area is plotted versus the optical power and fitted by the values calculated for the paraxial and not paraxial case. It is very clear that the second approach gives a very satisfactory fit.

Conclusions

In this paper we have discussed the problem of nonlinear optical reorientation induced in a nematic liquid crystal by a tightly focused Gaussian beam underlining the novel features arising under these conditions and comparing them to the behaviour expected for a Gaussian beam under paraxial approximation. We have found the following main differences occurring in the calculated reorientation for the not paraxial case with respect to the paraxial one are:

1) no threshold effect;
2) reduced reorientation at the same optical power
3) orientational singularity at $z = 0$

A very good fit has been obtained between the calculated values and the experimental data related to the radius of the reoriented area.

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