

Extended summary

Unconventional optical manipulation

in liquid crystals by nonlocal effects of reorientation

Curriculum: Ingegneria dei Materiali, delle Acque e dei Terreni

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Abstract. Optical trapping of low index particles in liquid crystals is a completely different phenomenon as compared to traditional manipulation of colloids by light in isotropic media. The latter is related to intensity gradient forces, that origin, for example, from a fundamental Gaussian laser beam focused by an objective lens; in order to work, this optical trap requires the refractive index of the particle to be higher than the one of the surrounding medium. The former, on the contrary, is due to the interaction, by means of elasticity of liquid crystals, between an optically distorted region around the beam focus and the colloid itself, even if the condition on the refractive indices is not fulfilled. The resulting attractive force shows peculiar properties, the same as the ones of the hosting medium: it is anisotropic in space and active over very long range distances. Our analysis, carried out under experimental conditions that prevent the effect of conventional trapping, has made clear the crucial role of the nonlocality of the optical reorientation, by showing the dependence of the trapping force on the size of the reoriented area. A theoretical model for director configuration, solved with a full numerical approach, was also proposed, taking into account the actual form of the Gaussian focused beam on the liquid-crystalline medium, in good agreement with experimental data.



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1 Problem statement and objectives

The long-range nature of the orientational order in liquid crystals is responsible of many fascinating optic, electro-optic and mechanical properties of liquid crystals.

Recently, there has been an increased interest in novel liquid-crystalline systems, such as the inverted nematic emulsions [1] and nematic liquid crystals colloids [2]. It has been observed that the presence of small particles in an otherwise homogeneous nematic host gives rise to new, anisotropic, structural forces, not observable in any other isotropic media. The unique combination of these kind of forces in liquid crystals and their electro-optical properties could, therefore, lead not only to new self-assembled structures for controllable photonics devices, but also to novel soft solids with unusual mechanical properties.

Some years ago, Musevic et al. reported an unusual mechanism of laser trapping and manipulation of small colloids in the nematic liquid crystal [3]. In that experiment, micronsized glass particles with homeotropic boundary conditions were dispersed in the nematic liquid crystal, with refractive indices that were larger compared to the index of refraction of colloids. Under such conditions, a repulsive force was expected to arise between a strongly focused light and the colloidal particle. As a surprise, the opposite was clearly observed: the colloid was attracted into the laser focus over extraordinary large separations of several microns. Musevic at al. observed that the optically induced distortion of the nematic director field (*optical Fréedericksz transition*, *OFT*) plays an important role, and two possible mechanisms were identified for this forbidden trapping.

Below the *OFT* threshold, the trapping has its origin in the anisotropic dielectric coupling of the optical field with the director field around the colloid. The particle is attracted into the laser focus until the favorable region of the distorted director field is in the most intense part of the optical field, as in the conventional manipulation. One could say that the colloid is dressed with an *isotropic index cloud* and behaves effectively as a high-index particle compared to its surrounding.

Above the *OFT* threshold, the highly localized EM wave creates a region in the liquid crystal, which is elastically distorted; this optically reoriented area interacts with the colloid and can lead to anisotropic forces between the laser focus of the trap and the particle. It should be stressed that in both cases, the colloid is attracted into the trap indirectly and the direct interaction between the low-index object and the optical trap is repulsive.

In a recent paper [4], our group highlighted the strong relationship between the unconventional trapping of low-index particles and the light-induced reorientation in the liquid crystal. It was demonstrated, in particular, that the trapping velocity is related to the optically induced phase shift due to the director reorientation by the laser beam creating the trap. Such a relationship confirms that the trapping mechanism in liquid crystals is different from the "ordinary" one and suggest that it is regulated by parameters not related to the traditional gradient force. Moreover, the need to claim two regimes for trapping is questionable, because one should expect a threshold-less optical reorientation when acting with a strongly focused beam on a liquid-crystalline sample. This aspect has been observed recently by Brasselet [5,6] and was actually confirmed by calculations in our theoretical model.

The objective of the research activity was focused on a better understanding of the origin of this particular trapping phenomenon and on finding a direct relationship between



the measured trapping force and the director reorientation around the focal region of the beam, with comparison between the experimental data and the theoretical values.

2 Research planning and activities

In order to get away from the issue of two trapping regimes in the investigation of optical trapping of low-index particles in higher index nematic, experimental conditions, that prevent the onset of conventional gradient trapping, were chosen. An objective with numerical aperture NA=0.45 was used to create the optical trap. This value of NA is usually below the minimum value required to get stable optical trapping in isotropic media. This point was actually checked by calculating the conventional trapping efficiency in the geometrical optical approximation (which describes well the experimental conditions), showing that usual manipulation is forbidden for all the values of optical power.

The possibility of obtaining long-range stable trapping with this objective was actually demonstrated, showing that the phenomenon is only due to the elastic interaction between the optically distorted trap area and the colloid with its surrounding elastically distorted region. Moreover, the long-range of interaction, as compared to the size of the focal spot, underlines the important role of nonlocality of the optical reorientation in this process.

The nonlinear optical distortion induced by the incident laser beam on the liquid crystal was measured by means of Self-Phase Modulation during the trapping experiment, while tracking of the particle movement was performed.

Director distortion was also analyzed by means of a theoretical model, where the proper expressions of the optical field of a focused beam and its propagation in a nematic liquid crystal were taken into account. Both the amount of the induced director distortion and the width of the involved region have been derived by computer simulations based on the model.

2.1 Optical trapping experiments

Trapping experiments were carried out on thin cells (50 μ m thickness) filled by a mixture of spherical silica particles dispersed in the nematic liquid crystal 5CB; the refractive indices of 5CB were greater than the one of the colloid. The particles' surface was covered by DMOAP, which gives rise to strong homeotropic anchoring at their surface. Cell substrates were also covered by DMOAP, in order to obtain samples with homeotropic alignment.

An *optical tweezers* system was used to manipulate colloids; it is built on a classic inverted microscope configuration, with a frequency-doubled Nd:YVO₄ laser at λ =532 nm as the light source. The Gaussian TEM₀₀ beam was focused on the cell by a 20X microscope air objective with NA=0.45; the optical power of the linear polarized optical field, impinging on the sample, varied in the range 8-180 mW. In order to work in unfavorable conditions for conventional trapping, besides using a numerical aperture less than 0.7, experiments were performed in *under-filling conditions* (that is, the laser beam waist is *lower* than the radius of the back aperture of the objective, resulting in a reduced trapping efficiency). Figure 1 presents a schematic view of the *optical tweezers* system.

Stable trapping of silica particles was observed for all the values of the incident optical power. We selected an isolated particle in the middle of the sample and positioned the trap in the same plane at different distances from the colloid, in the range of several tens of microns.





Figure 1. Optical tweezers system

Different trajectories from several starting points around the trap center were observed and recorded by means of a CCD camera, similar to what was reported in [7], clearly showing the anisotropic behaviour of the manipulation event.

2.2 Trapping efficiency in "forbidden" conditions

In order to check the role of conventional optical trapping in our experiments, the trapping efficiency was evaluated under the geometrical optical approximation, (ray optics regime is well justified in our conditions).

Optical forces, that is, scattering, gradient and total forces were calculated with a numerical integration over the distribution of input rays; results are shown in Figure 2 where the value 1.11 for the ratio *m* between the refractive index of the colloid and the one of the surrounding medium was chosen in agreement with the hypothesis of *dressed particle* [3,7].



Figure 2. Calculated optical forces

The total force is positive for every position on the optical axis, that is, it points in the direction of positive z-axis; this means that no stable conventional trapping was allowed in our experimental conditions. If a stable optical trap along the z-axis is obtained, as we have



seen, it cannot be related to the usual trapping conditions and another mechanism is responsible for the effect.

2.3 Trapping force due to optical reorientation

The dependence of the particle displacement r versus time was recorded, for different values of the optical power; the general trend is very similar to that usually reported for optical trapping in liquid crystals with conventional high NA objectives [4,7].

Following an usual approach [7,9], the force between the laser focus and the silica particle was calculated assuming that it is nearly balanced by the drag force:

$$F_D = 6\pi R \eta \frac{\partial r}{\partial t} \tag{1}$$

where R= bead radius and η = medium viscosity. The particle motion was highly viscously damped, allowing us to directly determine the attractive force from the displacement data by differentiating them to obtain the colloid velocity.

Figure 3 shows the typical behavior of the trapping force versus the particle-trap distance from the trap center (the negative value means that the force points toward the focus of the beam).



Figure 3. Measured optical force versus distance

The reported curve corresponds to the optical power P = 92 mW; the same behaviour is obtained for all powers used in the experiment. The only difference is the equilibrium position, that is the *r*-coordinate where the particle stops its movement and the force is zero; in this situation $r_{eq} = 8.8 \ \mu m$.



3 Analysis and discussion of main results

3.1 Optical force formalism

By looking at Figure 3, we can see that the attractive force increases as it approaches the trap until it reaches a maximum, decreases at short distance with decreasing r, and eventually vanishes, the spatial dependence is on r^a at long distance with α =2. From more than 50 measurements of trapping events, the average exponent α was obtained as 2.11±0.08. At shorter distance, the dependence is different, and the overall curve is well described by the function:

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

where A and B are constants whose value is given by the use of a fitting procedure (the solid line in Figure 3 is the best fit using Equation (2)), and is related to the strength of the two term, F_A being an attractive component, greater at long distances, and F_B a repulsive one.

The interaction between a laser-induced reoriented nematic and a colloid cannot be considered as the interaction between two colloids, even if conceptual similarities are possible, as already pointed out in [8]. On the other hand, it is questionable to treat light-induced reorientation as a defect around a colloid, with dipolar or quadrupolar symmetry, since symmetry, size and amount of induced distortion are supposed to vary with optical power.

Lev et al. [8] were able to justify the Coulomb-like attractive interaction in the case of a spherical particle of dipolar symmetry trapped by a light-induced reoriented area. Possibly, similar arguments could justify the r^3 dependence of the repulsive term.

Following concepts similar to the ones used for interaction between two colloids [9], the attractive part of the force can be written as:

$$F_{A} = -\frac{A}{r^{2}} = -\frac{CKa^{2}(P)}{r^{2}}$$
(3)

where a=radius of the light-induced reoriented area, dependent on the light power; K=elastic constant of liquid crystal and C=dimensionless constant.

From Equation (5), we expected the ratio between the attractive force F_A and the radius a to be independent on the size of the distorted area, that is, independent on the optical power. Measurements of a(P) were performed by looking at the distorted area between crossed polarizers. For each value of P, we considered the curve of the force F(r) for medium-long distance, where the term $1/r^2$ is the dominant one. In this way, the ratio $F(r)/a^2(P)$ was found for different values of P, as reported in Figure 4. Data fall on a single curve, which confirms that the force scales as $a^2(P)$, pointing out that the dependence of the force on the optical power is due to the increasing width of the distorted area with increasing power.

Following a similar approach for the repulsive term, in agreement with the expressions reported in [10] for colloid-colloid interaction, F_B can be written as:

$$F_{B} = \frac{B}{r^{3}} = \frac{C'a^{3}(P)}{r^{3}}$$
(4)

where C'=dimensionless constant and the other parameters were defined before.





Figure 4. Trapping force over the square radius of the lightinduced reoriented area versus the distance from the trap, for three different powers (log-log plot)

We experimentally observed that the particle equilibrium position r_{eq} changes by increasing power; according to our fitting curve (Eq. 2), the equilibrium position, $F(r_{eq})=0$, is given by:

$$r_{eq} = \frac{B}{A} \tag{5}$$

where A,B=constants already defined; Equation (5), according to expression of F_A and F_B (Eqs. 3 and 4), becomes:

$$r_{ea} = C^{\prime\prime}a(P) \tag{6}$$

where C^{*}=dimensionless constant. This means that a linear increase of the equilibrium distance versus the radius of the reoriented area should be expected; this was actually found in our experiments, by means of independent measurements of r_{eq} and a(P) for different values of power, confirming the consistency of the description for the trapping force (Eq. 2).

It is worth underlining, finally, that in our case a single trapping mechanism was considered, without any onset of two different regimes, since the used fitting function remains valid at all used powers.

3.2 Optical reorientation under strong focusing conditions

We have experimentally observed that, while the focal waist of the laser beam is constant (about 1 μ m in our case), the effective area of the reorientation increases with power, reaching a radius close to 30 μ m, at the highest power used; this great nonlocality plays a crucial role in the trapping mechanism and a relationship between the strength of the force and the amount of the optically-induced reorientation was clearly found (Eq. 2). A further step in the analysis of the process was done by evaluating the actual reorientation by the laser beam.



Light-induced reorientation in liquid crystals has been usually considered in the planewave approximation, which is generally fulfilled when the *confocal beam parameter* is much bigger than the sample thickness [11]. On the other hand, the effect of tighter focusing has been investigated by different authors [12-14], who pointed out the strong nonlocality of the reorientation and the peculiar effect arising in the beam wave front. In all previous works, the Gaussian distribution of the light intensity was taken into account, while the depolarization effect due to the wave-front curvature was neglected. In an *optical tweezers* system, the strong focusing due to the objective makes this approximation very weak, because several **k** vectors of the impinging wave are involved in the reorientation process. Recently, Brasselet considered the effect of strong focusing of a circularly polarized beam on a liquid-crystalline film, showing the occurrence of no threshold reorientation [5].

We described the interaction of the optical field with the molecular director \mathbf{n} by using the field components calculated for a focused Gaussian beam.

A focused laser beam, impinging on a homeotropic liquid-crystalline cell of thickness d, was considered. As usual, the induced reorientation can be obtained by minimizing the total free energy, using the *Euler-Lagrange equation* [15]; the total energy comes from integration over the volume of the sample of the free energy density F_{tot} of the system, that includes contribution due to the elastic and the optical torque [11]:

$$F_{tot} = F_K + F_E = \frac{K_1}{2} \sin^2 \theta \left(\frac{d\theta}{dz}\right)^2 + \frac{K_3}{2} \cos^2 \theta \left(\frac{d\theta}{dz}\right)^2 - \varepsilon_\perp \frac{\left|\vec{E}\right|^2}{4} - \frac{\Delta \varepsilon}{4} \left(\vec{n} \cdot \vec{E}\right) \left(\vec{n} \cdot \vec{E}^*\right)$$
(7)

where $\theta(r,z)$ =reorientation angle; K_i =elastic constants of the liquid crystals; **n**=director; ε_{\perp} =ordinary dielectric constant, **E**=optical field and $\Delta \varepsilon$ =dielectric anisotropy at optical frequencies.

As mentioned, the optical field **E** to be used in the last term of Equation (7) is the field associated with a focused beam [16]. When a linearly x-polarized plane wave is focused by an aplanatic lens with numerical aperture $NA=nR_L/f$ (R_L =aperture radius of the lens; f=focal length; n=index of the medium after the lens), the field in the focal regions is given by (x, y, z components):

$$\vec{E}(r,\varphi,z) = \frac{ikf}{2} \sqrt{\frac{n_1}{n_{eff}}} E_0 e^{-ikf} \begin{bmatrix} I_{00} + I_{02}\cos 2\varphi \\ I_{02}\sin 2\varphi \\ -2iI_{01}\cos\varphi \end{bmatrix}$$
(8)

where $r, \varphi, z=$ cylindrical coordinates; k= wave-vector modulus, f= lens focal length; n_i =refractive index of the medium before the lens; n_{eff} = effective refractive index of the medium after the lens (in our case there were three different media: air, glass and liquid crystals) and $I_{ij}=$ integral functions to be evaluated for each value of r and z. The three components of the field were numerically calculated; the beam profile of the focused field **E** is shown in Figure 5, where the square modulus of the field on the longitudinal xz-section and yz-section is presented.

Equation (8) was included into Equation (7) and minimization of the total free energy was performed by applying the *Euler-Lagrange equation* in the one elastic constant approximation. In this way, we got a differential equation for the light-induced reorientation angle



 $\theta(\mathbf{r},z)$, respect to the initial homeotropic configuration of the liquid crystals molecules (z-axis):

$$K(\theta_r + r\theta_{rr} + r\theta_{zr}) + \frac{\Delta\varepsilon}{16}k^2f^2\frac{n_1}{n_{eff}}rE_0^2[(D-H)\sin 2\theta + 2F\cos^2\theta] = \frac{\Delta\varepsilon}{16}k^2f^2\frac{n_1}{n_{eff}}rE_0^2F$$
(9)

where K=average elastic constant of the liquid crystal; θ_r , θ_z =derivatives of θ with respect to *r* and *z* and *D*,*H*,*F*=constant parameters coming from the integral functions $I_{ij}(r,z)$.

Equation (9) was solved numerically using the finite difference method; we got the tilt angle distribution $\theta(r,z)$ in the sample volume as a function of the power incident on the liquid crystal. Computer calculation was carried on by using the same parameters' values as those used in the experiments. Figure 6 reports a three dimensional plot of the reorientation calculated angle for an optical power P=30 mW in our experimental conditions.



Figure 5. Square modulus of the focused field **E** on the xz and yz longitudinal planes calculated from Equation (8) for NA=0.45 and P=30 mW



Figure 6. Three dimensional plot of the director distortion $\theta(r,z)$ induced by a focused Gaussian beam in case of NA=0.45 and *P*=30 mW (angle values are indicated in radians)



By looking at Figure 6, we have noticed the peculiar behaviour of the reorientation around the focal region: the tilt angle changes the sign crossing z_{β} , that is the location of the beam focus ($z_{f}=25 \ \mu m$ in the middle of the sample). However, the important feature to be analyzed is the nonlocality of the optical reorientation; from the calculated values of $\theta(r,z)$, we obtained, finally, the theoretical diameter 2a(P) of the reoriented region (imposing the condition $\theta(r,z) > 0.1$ rad) that was compared with the experimental one, taken by observation of microscope images under crossed polarizers. A good agreement between measured and theoretical values was found, showing the consistency of the adopted model.

From $\theta(r,z)$, we were able to calculate the optically *self-induced phase shift* $\Delta \psi$ between the ordinary and extraordinary waves in the optical field **E**, due to reorientation process; this was done applying the following expression:

$$\Delta \psi = \frac{2\pi}{\lambda} \int_0^d \Delta n(r=0,z) dz \tag{10}$$

where $\Delta n = n(\theta) - n_0$ = optically-induced birifrangence; *d*=sample thickness and λ =wavelength. Self-Phase Modulation measurements [11] allowed us monitoring the nonlinear optical reorientation induced by the trapping beam; the maximum induced phase shift, the same calculated in Equation (10), was experimentally obtained in this way. Also in this case, the comparison between measured data and theoretical values, coming from numerical solutions, was very satisfactory; this means that the model is suitable to represent the interaction of the focused beam with the liquid-crystalline medium. Moreover, it is worth emphasizing that our model showed the absence of the threshold for optical reorientation; this is not surprising, since ,when a focused beam is used, a wide spectrum of incident wave vectors should be considered and a non-threshold behaviour can be expected.

4 Conclusions

We have investigated unconventional optical trapping of silica particles with a refractive index lower than the ones of the surrounding nematic liquid crystals, under experimental conditions that prevent the effect of classic manipulation, originated by optical gradient forces. Stable and long-range trapping was observed, showing that this phenomenon in liquid crystals is regulated by a completely different mechanism, with respect to that working in isotropic media. The trapping force has a range (several tenths of microns) longer than the conventional ones, and it includes an attractive term scaling as r^2 and a repulsive term scaling as r^3 . This behaviour shows a stronger interaction than the one observed for colloids with associated dipolar defects. However, this comparison suggests an analysis of the power dependence of the attractive and repulsive terms, that demonstrates that the size *a* of the optically reoriented area affects the trapping force. In other words, the strength of interaction is regulated by the nonlocality of optical reorientation, since the width of the distorted area is more than one order of magnitude bigger than the focal waist.

A model for evaluating the light-induced reorientation was also developed, by taking into account the depolarization effect due to strong focusing of a Gaussian beam. We got a very satisfactory agreement with experimental data for both the width of the optically distorted area and the nonlinear phase shift, that can be measured on the trapping beam.



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