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Total Optical Torque and Angular Momentum Conservation in Dye-Doped Liquid Crystal Droplets Spun by Circularly Polarized Light

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Micrometric droplets of nematic liquid crystal dispersed in water were trapped and made to spin in the focus of a circularly polarized laser beam. The measured rotation speed is directly proportional to the angular momentum exchanged per unit time by the droplet with the electromagnetic field, that is to the total optical torque acting on the droplet. We searched for a possible enhancement of this total torque when the liquid crystal of the droplets was doped with a light-absorbing dye. In contrast to the local optical torque acting on the molecular director, which is known to be strongly enhanced by the presence of the dye, the total torque acting on the droplet as a whole did not show any enhancement. This null result is in agreement with the leading theory of the dye-assisted (or photoinduced) optical torque, which postulates that this torque is associated with an internal angular momentum exchange between different material degrees of freedom, namely orientational and translational. Moreover, our findings confirm the predicted existence of a photoinduced stress-tensor acting on the dye-doped liquid crystal.

INTRODUCTION

It is well known that light is capable of exerting a torque on a birefringent anisotropic material, leading to an exchange of angular momentum between the optical field and matter. For example, a suspended small solid particle made of a birefringent anisotropic material, such as a crystal, can be easily made to spin by an impinging circularly polarized light beam [1,2]. An entirely analogous whole-particle rotation can be induced by circularly polarized light in the case of nematic liquid crystal (NLC) droplets immersed in a low-viscosity fluid such as water [3]. However, in contrast to ordinary solid crystals, NLC's are both anisotropic and fluid, and this implies that their axes of rotational symmetry, i.e., the molecular director, can be also rotated independently of the material fluid displacement, that is without inducing a significant flow or matter displacement. For example, the continuous rotation of the molecular director induced by circularly polarized light was demonstrated in a homeotropic NLC cell [4,5], where the fixed cell walls frustrated the fluid flow (although a small residual flow is still present [6]).

In the early 90's, it was serendipitously discovered that dissolving a small fraction (0.1–1% w/w) of certain dyes in the NLC's leads to a dramatic enhancement of the light-induced torque acting on the molecular director (by a factor of several hundreds) and of the corresponding rotation speed of the molecular director, at given light intensities [7–9]. This torque enhancement was so far demonstrated only in the case of fixed sandwich-cells, and therefore it concerns the rotation of the molecular director only, with almost no accompanying flow or matter displacement.

This leads naturally to asking whether a similar enhancement of the light-induced rotation speed can be achieved with dye-doped NLC also when the material system is rotated as a whole, such as in the case of suspended liquid crystal droplets. Indeed, one would naively expect the light-induced total torque acting on the whole NLC droplet to correspond to the integral of the specific torque per unit volume acting on the molecular director within the droplet. If the latter is strongly enhanced, one would expect a similar enhancement of the total torque as well. This question is addressed in this paper, where we report the results of our experimental investigations of the rotation dynamics of dye-doped NLC droplets under the effect of circularly polarized laser light. To highlight the effect of dye photoinduced contributions, we compared the case of light having a wavelength falling within the dye absorption band with that of light which is not absorbed by the dye. Moreover, we compared the case of dye-doped NLC with the case of pure undoped NLC.

EXPERIMENT

We prepared emulsions of pure and dye-doped NLC's in bidistilled water. By properly choosing the relative quantities of liquid crystal and water, we could obtain sufficiently stable emulsions containing many liquid crystal droplets in the 1–20 μm diameter range. The liquid crystal employed was the commercial mixture E63, provided by Merck, Darmstadt, Germany. The dye used in doped materials was the 1,8-dihydroxy 4,5-diamino 2,7-diisopentyl anthraquinone (HK271, provided by Nematel, Mainz, Germany), known as one of the most effective dyes in the photoinduced effects. We prepared dye-liquid crystal solutions at a concentration of 2% in weight, leading to molecular director torque enhancement factors of several hundreds [8,9].

For performing the optical rotation experiments, a small volume of emulsion was placed on a microscope glass slide and covered with a thin (about 0.18 mm thick) glass coverslip, thus forming a thin cell (open on the sides). A 100× oil-immersion microscope objective (Carl Zeiss, NA = 1.25) was used both to image the droplets on a CCD camera (using lamp illumination from below the cell, in combination with suitable imaging optics) and to focus the input laser beams that were used to trap the droplets and induce their rotation, as discussed in the following (see Fig. 1). The objective was attached, via an oil thin film, to the glass coverslip. When needed, the microscopic imaging could be made in the crossed or nearly-crossed polarizers geometry, in order to visualize the droplet birefringence. CCD images and movies (acquisition rate of 50 frame/s) could also be recorded on a PC, for subsequent frame-by-frame analysis.

Our setup for the optical trapping and manipulation, shown in Figure 1, is a dual-wavelength optical-tweezers apparatus. It differs from usual optical tweezers arrangement in that two laser beams, respectively generated by a diode-pumped solid state laser (wavelength of 785 nm, subsequently called "IR" beam) and a He-Ne laser (wavelength of 633 nm), are brought to a common focus at the specimen plane of the microscope objective. Both beams were able to achieve trapping of droplets for diameters in the 2–12 μm range, as proved by a sudden stop (or confinement) of the droplet Brownian motion after trapping.

The common focus of the two lasers allowed us to switch the controlling beam from one wavelength to the other (e.g., to switch the He-Ne beam on and the IR off, or vice versa) without changing the trapped liquid crystal droplet. Since the He-Ne wavelength is close to the maximum of the dye absorption band, while the IR wavelength falls completely outside the absorption band, this corresponds to adding or removing the dye photoinduced effects, so as to allow for a direct comparison of the behavior obtained in the two cases with the same droplet.
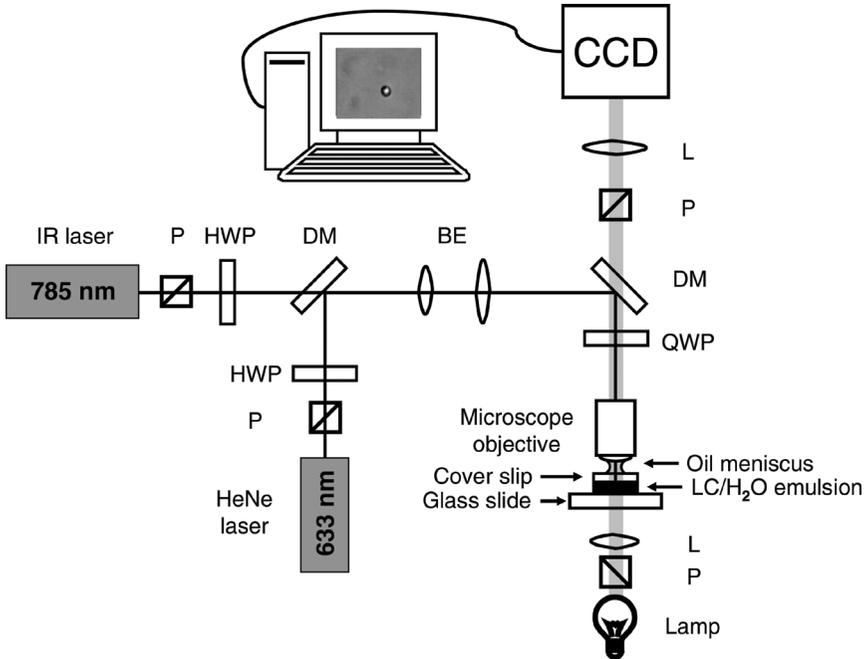
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FIGURE 1 Experimental apparatus, P: polarizer, HWP: half-wave plate, DM: dichroic mirror, L: lens, CCD: ccd camera, BE: beam expander for objective back aperture overfilling, QWP: quarter-wave plate (for the appropriate laser wavelength).

Besides for trapping, the two beams were also used to induce the electromagnetic and photoinduced torques acting on the droplet and driving its reorientation or continuous rotation. By inserting along the beam (just before the microscope objective) a suitable birefringent wave plate, we could control the polarization of each beam (separately). In particular, in this paper we focus only on the experiments performed with circular polarization, while additional experiments with elliptically and linearly polarized light will be discussed elsewhere.

Small NLC droplets are known to show either one of two possible internal spatial distribution of the molecular director [10]: the cylindrically symmetric axial (or bipolar) distribution, in which the molecular director is approximately uniform in the centre of the droplet with some distortion toward the droplet surface (see, e.g., the schematic pictures in the lower panel of Fig. 2), and the spherically symmetric radial distribution, with a hedgehog defect at the droplet centre. The images of axial and radial droplets under a polarizing microscope
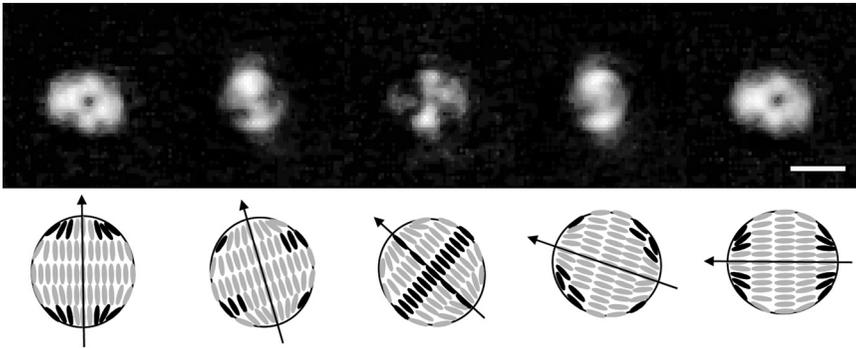
FIGURE 2 Upper panel: sequence of frames of an acquired movie showing a nematic liquid crystal micrometric droplet observed between crossed polarizers, during its laser-induced rotation. Scale bar: 1 μm. The polarizers axes were oriented at about ±45° from the figure vertical direction. Lower panel: schematic representation of the corresponding molecular director configuration occurring within the droplet, as deduced from the crossed-polarizer images (black ellipsoids correspond to regions in which the molecular director is roughly aligned with the polarizer axes and light is blocked). The only region of the droplet in which the dark (crossed) regions are not well explained by our simple graphical analysis is its centre, possibly because of some director reorientation induced by the optical torque itself.

can be similar, but we could identify axial droplets either by checking that the image changed while rotating both the microscope polarizers (keeping them crossed), or by looking at the droplet dynamical behavior under the laser beam, as only axial droplets could be set in rotation (for radial droplets the total optical torque vanishes identically if light absorption is neglected, due to the droplet radial symmetry). About 80–90% of the droplets of our emulsions were found to be axial. In the following we will only refer to them.

We measured the spinning of droplets by analyzing the frames of a droplet rotation movie. Using the crossed-polarizers geometry we could image the birefringence rotation, corresponding to the director motion, as a periodic pattern modulation in time, as shown in Figure 2. For the smallest droplets, we could observe a significant modulation of light intensity during the rotation even without the analyzer, presumably due to the anisotropic scattering cross-section of the droplets. In certain cases, we could also measure the droplet fluid rotation (i.e., regardless of director rotation) when a smaller satellite droplet (or another small object) happened to get trapped close to the rotating droplet and was thereby dragged around. The rotation speed measured in this way

was found to be consistent with that measured by observing the modulation of the polarization pattern.

RESULTS

In order to see if the photoinduced effects give rise to a rotation speed enhancement, we measured the rotation frequency induced with the He-Ne and the IR beams as a function of droplet diameter. We also measured the rotation frequencies induced in pure (undoped) liquid crystal droplets, although of course in this case the comparison could not be done with exactly the same droplet sizes. In these measurements, the two laser beams were adjusted for having a roughly equal power (or, more precisely, an equal optical torque due to dielectric anisotropy). In particular, the light power P_0 measured after the microscope objective was about 4.1 mW for the He-Ne beam and 2.8 mW for the IR. Taking into account the respective laser (angular) frequencies ω, these two powers correspond, within 15%, to the same photon flux $P_0/(\hbar\omega)$ and therefore to the same input flux of angular momentum $M_{z0} = P_0/\omega$.

In Figure 3 we show the rotation frequency observed for several pure NLC (open symbols) and dye-doped NLC (filled symbols) droplets of different sizes, using the IR laser beam. Here, with IR light, we do not
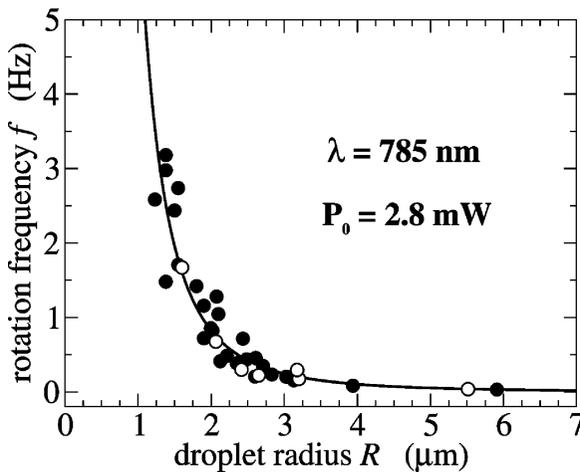

FIGURE 3 Frequency of droplet rotation induced by circularly polarized IR light having a wavelength that falls outside the dye absorption band, for the case of dye-doped (filled circles) and pure (open circles) liquid crystal, as a function of droplet radius. The laser power on the sample was 2.8 mW. The solid line is a theoretical prediction based on the model reported in the text.
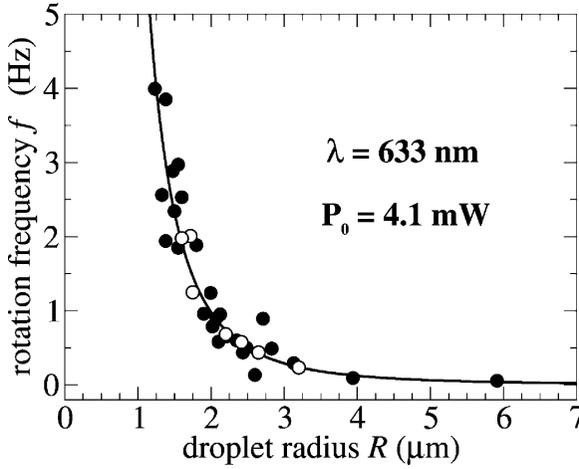
FIGURE 4 Frequency of droplet rotation induced by circularly polarized light from a He-Ne laser, which is absorbed by the dye, for the case of dye-doped (filled circles) and pure (open circles) liquid crystal, as a function of droplet radius. The laser power on the sample was 4.1 mW. The solid line is a theoretical prediction based on the model reported in the text.

expect to see a difference in the doped and undoped cases, as the IR light falls outside the dye absorption band and therefore photoinduced effects are not possible. This is indeed what we find, as seen in the figure.

Figure 4 shows the rotation frequencies obtained with the same (pure and dye-doped) droplets when using the He-Ne laser beam, which is well absorbed by the dye (a few droplets are not present in both figures because they escaped the trap during the switch). As clearly seen in this second figure and comparing the two figures to each other, no significant rotation speed enhancement takes place in the dye-doped LC case with the He-Ne beam as compared both to the IR beam case and to the pure LC case. Droplets of the same radius exhibit roughly equal rotation speeds in all three cases.

The experimental points have been compared with the predictions of a simple model in which the nematic droplets were considered as rigidly rotating spherical bodies. According to the well-known Stokes equation, the angular velocity Ω of such an object immersed in a surrounding fluid with viscosity η is given by the relation

$$\Omega = \frac{M_z}{6\eta V_d}, \tag{1}$$

where V_d is the volume of the droplet and M_z is the total external torque acting on the droplet about the rotation axis. In our calculations,

M_z was identified in all cases with the standard dielectric part of the total electromagnetic torque [1,2] and approximated with the input angular momentum flux $M_{z0} = P_0/\omega$ (the output flux of optical angular momentum seems to give a negligible contribution, although the reason for this is not completely clear). As seen from the figures, a good agreement is obtained with this assumption for both dye-doped and undoped droplets, both for the absorbing and the non-absorbing wavelengths (solid lines), with no adjustable parameters. The details of the model will be published elsewhere.

We emphasize that the enhancement factor that was expected from the corresponding enhancement of the optical torque acting on the molecular director is of several hundreds. Therefore our key result reported here is a "null" one: the droplet of dye-doped NLC as a whole does not experience any significant enhancement of the total optical torque, as compared to transparent droplets where the photoinduced dye effects are negligible.

DISCUSSION

Let us now discuss the physical meaning of our findings. We started from the simple idea that the total light-induced torque acting on the whole droplet is simply the sum (integral) of the torques acting on the molecular director in each small volume inside the droplet. If the terms which are added up are all enhanced by the same large factor, why should the sum not be enhanced?

A qualitative answer to this question, in our opinion, goes as follows. In dye-doped NLC's, besides the photoinduced torque acting on the molecular director, the absorbed laser light actually generates also other significant forces. These photoinduced forces are applied to the fluid displacement degrees of freedom and not to the orientational degrees of freedom, that is the molecular director. From a molecular point of view, these forces can be seen as acting on the molecule centres of mass. Therefore, relative to each molecule centre of mass, these forces do not correspond to an additional torque. However, relative to the droplet centre, these forces do contribute another light-induced torque, which must be included in the total torque acting on the droplet, and which will presumably balance all the (enhanced) sum of the molecular director torques.

These photoinduced forces, which can be formally described by introducing a photoinduced stress tensor [11], have not been directly measured thus far. Our findings that the total torque acting on a dye-doped NLC droplet does not show any enhancement is therefore also the first experimental demonstration of their existence. In other

words, the existence of these photoinduced forces is highlighted by their contribution to the balance of all torques acting on the droplet, being all other contributions to this balance already known.

But why do these photoinduced forces cancel so precisely the effect of the photoinduced molecular director torque? The answer is that it is a consequence of angular momentum conservation. Indeed, the presence of dye in the droplet clearly cannot enhance the angular momentum carried by light. Although the absorption of light alters slightly the transfer of angular momentum between the field and the droplet, this effect is quantitatively small and cannot account for the large enhancement of the light-induced torque. Therefore the additional angular momentum transferred to the molecular director in the droplet cannot come from the light itself, which plays only the role of driving the transfer. Instead, it must come from other degrees of freedom, such as those associated with fluid displacement (or, equivalently, with the molecule centre-of-mass positions). One can still doubt here if the angular momentum could be finally exchanged, at least in part, with the *external* fluid surrounding the droplet (water, in our experiments), or if instead the exchange is only with the *internal* fluid (the liquid crystal itself). In the former case, one could still find some form of total torque enhancement, as the cancellation mentioned above could be only partial. However, our data show that this is not the case, i.e., the angular momentum exchange appears to be fully internal to the droplet. This conclusion is also in agreement with the predictions of a phenomenological theory based on the torque constitutive law and with the currently most accredited microscopic model of the dye-induced optical effects [11–14]. If the enhanced angular momentum exchange is all internal to the droplet matter (apart from the ordinary light torque due to the birefringence), one clearly cannot have any dye-induced enhancement of the total torque acting on the whole droplet.

In summary, the photoinduced dye torque acting on the molecular director has a reaction force acting on the fluid displacement, which is represented by the photoinduced stress tensor. The total torque acting on the whole droplet includes both action and reaction, and therefore there is a perfect cancellation of all dye photoinduced effects.

A more quantitative detailed analysis of the light-induced torques and forces acting in the NLC droplet based on the above considerations will be presented elsewhere.

CONCLUSIONS

We have experimentally searched for a possible enhancement of the total optical torque acting on a droplet of nematic liquid crystal which

has been doped with dyes that are known to strongly enhance the internal torque acting on the molecular director. We found that no torque enhancement is present for the whole droplet. This null result confirms the validity of current microscopic models that associate the enhancement of light-induced torque to a light-driven exchange of angular momentum between internal degrees of freedom of matter [11–14], namely the orientational and translational molecular ones. This confirms the validity of the angular momentum conservation law, as expected. Moreover, our null result proves for the first time the actual existence of the photoinduced stress tensor associated with light absorption, which was postulated only theoretically a few years ago [11].

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