Continuously rotating chiral liquid crystal droplets in a linearly polarized laser trap

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Abstract: The transfer of optical angular momentum to birefringent particles via circularly polarized light is common. We report here on the unexpected, continuous rotation of chiral nematic liquid crystal droplets in a linearly polarized optical trap. The rotation is non-uniform, occurs over a timescale of seconds, and is observed only for very specific droplet sizes. Synchronized vertical motion of the droplet occurs during the rotation. The motion is the result of photo-induced molecular reorganization, providing a micron sized opto-mechanical transducer that twists and translates.

OCIS Codes: (160.3710) Liquid crystals; (350.4855) Optical tweezers or optical manipulation; (160.1585) Chiral media

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Using light to move objects has wide appeal, though the scale of the momentum transfer limits applications to microscopic particles. In particular, it is possible to transfer angular momentum from light to birefringent media, as was first demonstrated in the 1930s [1], becoming far easier with the development of laser tweezers [2], which combined high optical fields with microscopic particles. Friese *et al* [3] first controlled the optical rotation and alignment of a calcite plate by varying the polarization of the incident beam. The mechanism relies on a phase shift occurring between polarization components of the input and output light, causing a change in the angular momentum of the light and hence imparting a torque to the material. The phenomenon has the potential for impact in areas including sensing and microfluidics.

Liquid crystal droplets are obvious candidates for optical manipulation and the transfer of optical angular momentum; the materials are highly birefringent ($\Delta n \sim 0.2$), and uniform though relatively complex structures can readily be formed. Circularly polarized light has been shown to induce rotation of nematic liquid crystals [4] and nematic droplets [5] with the relative contributions of absorption and anisotropic scattering being considered [6]. In an earlier paper we demonstrated that wave-plate behavior is the dominant mechanism causing optical angular momentum transfer from a circularly polarized beam to a nematic droplet [7]. When a nematic droplet on the micron scale is held in a *linearly* polarized optical trap it lies close to the trapping-beam focus with the bipolar axis in the plane normal to the beam (droplets of this type typically adopt a bipolar director structure [8]). The droplet then rotates, usually until the bipolar axis and the plane of the incident polarization are coincident and, with the additional influence of damping, remains stationary since it experiences no further torque in this orientation [9]. In this paper, by contrast, we will discuss the *continuous* rotation of long-pitch chiral nematic droplets in plane polarized optical traps. We previously reported an observation of anomalous rotation of chiral droplets [10], but at that time neither the conditions required for rotation, nor the mechanism responsible were known. Here we present a full account of the phenomenon, including the new observation of a synchronous vertical motion of the rotating droplet. We explain why only very few chiral droplets can be rotated in the linearly polarized light and give a full explanation of our observations, resulting in the description of a new opto-mechanical transducer.

We controlled the helicoidal pitch of the chiral nematic materials by using mixtures of the nematic and chiral nematic materials MDA-1444 and MDA-1445 [11]; six mixtures were made with natural pitch lengths (p) of 15.6 μm, 11.4 μm, 7.8 μm, 5.7 μm, 3.4 μm and 3.1 μm. The helicoidal pitch and refractive indicies of these mixtures are known to be relatively temperature independent [12], avoiding any effects due to heating in the optical trap. Suspensions of droplets with diameters (D) between 1 and 20 μm were created by dispersing and agitating a small quantity of each mixture in distilled, deionized water. Our previous work [10] had indicated that rotation was only observed in droplets with $D < 20$ μm, and that the director adopts a twisted bipolar configuration as has been observed previously in both nematic [13] and chiral nematic droplets [14], the appearance of which is illustrated in the inset of Figure 1.

The optical trap was formed using a strongly focused (NA = 1.3), linearly polarized 1064 nm laser beam. The laser power was fixed at 50 mW (measured at the back aperture of the objective) and the distance between the coverslip and the trapping position was 15 μm. A high degree of linear polarization was ensured in the trapping beam by including a plane polarizer immediately prior to the objective and verified *via* measurement of the Stokes parameters. The trapped liquid crystal droplet could be illuminated with white light and the image viewed via a charge-coupled device (CCD) camera. A photodiode was set to measure the transmission of the droplet between crossed polarizers as it rotated.

In the vast majority of cases, the droplet axis (see Figure 1) was observed to rotate to a fixed orientation and then remain stationary. Further rotation was only observed if the plane

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of incident polarization was rotated. The final angle between the droplet axis and the polarization plane was found to vary with the droplet diameter (*D*) as shown in Figure 1. Sudden changes in the orientation of the 7.8 μm pitch droplets are observed at droplet diameter to pitch length ratios (*D/p*) of around 0.5 and 1. As will become clear, these sudden changes in orientation as a function of *D/p* are a requirement for the rotation of chiral nematic droplets in linearly polarized light. Indeed, no such sudden changes are found in the 11.4 μm pitch mixture, and no rotation is observed in droplets of this material.

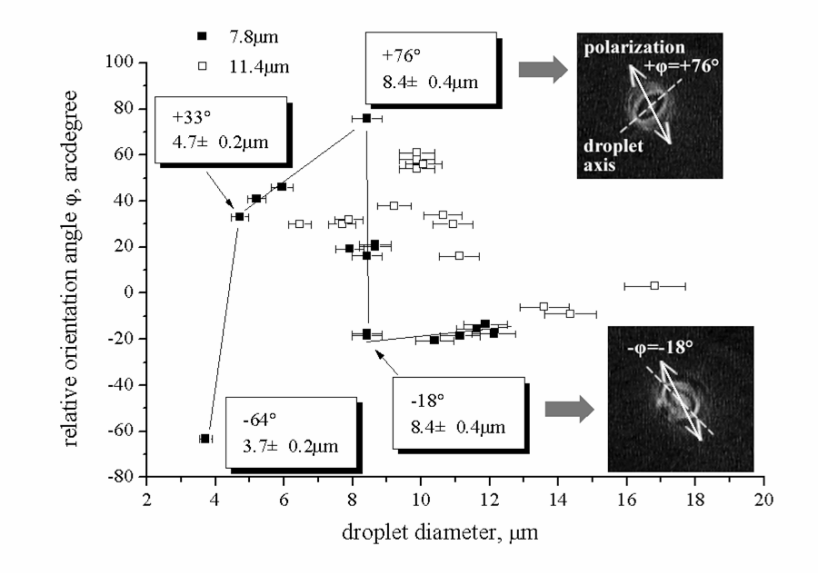

Fig. 1. The relative orientation of the droplet axis (dashed line) to the polarization axis of the light, φ (the range of φ has been restricted to ±90°). The open and closed squares represent data for materials of natural pitch 11.4 μm and 7.8 μm respectively. The two droplets shown in the insets have measured diameters of 8.4 ± 0.4μm, and adopt different preferred orientations in the trapping beam.

Continuous rotation of droplets was observed only in the mixtures with pitch lengths of 7.8 μm and below. Even in these mixtures, only droplets that satisfy the condition *D/p* = 0.5 and 1 rotate continuously in the linear light; the remainder rotate to the optimum orientation for the specific droplet size (and hence director structure), as shown in Figure 1 and then stop. Indeed the continuously rotating droplets fall in the regime shown in Figure 1 where there is a sudden change in the optimum orientation of the droplet axis; in the case of the mixture with natural pitch of 7.8 μm, droplets of diameter 3.9 ± 0.4 μm and 8.2 ± 0.4 μm. Details of the continuous rotation are shown in Figure 2; it is distinctly non-uniform, has 180˚ symmetry, and is accompanied by vertical movement of the droplet of around 0.3 μm away from the focus of the optical trap. Conversely, the rotation of nematic liquid crystal droplets in circularly polarized light is uniform [5,9] and is due largely to wave-plate behavior, with some contribution from other mechanisms [7]. We note that all of the chiral nematic droplets can be continuously rotated in circularly polarized light with the features of the rotation identical to those observed for achiral droplets. We confirmed that the droplet as a whole rotates, as opposed to the director structure alone rotating, by noting that droplets suspended in glycerol, a far more viscous medium, never rotate. We believe this observation to be valid even though the trap strength is weaker in glycerol (because of its higher refractive index) since optically induced rotation of the *director* structure in the droplet would occur even in a

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weaker trap, while rotation of the *droplet* occurs only if the drag due to the surface is less than the optical torque.

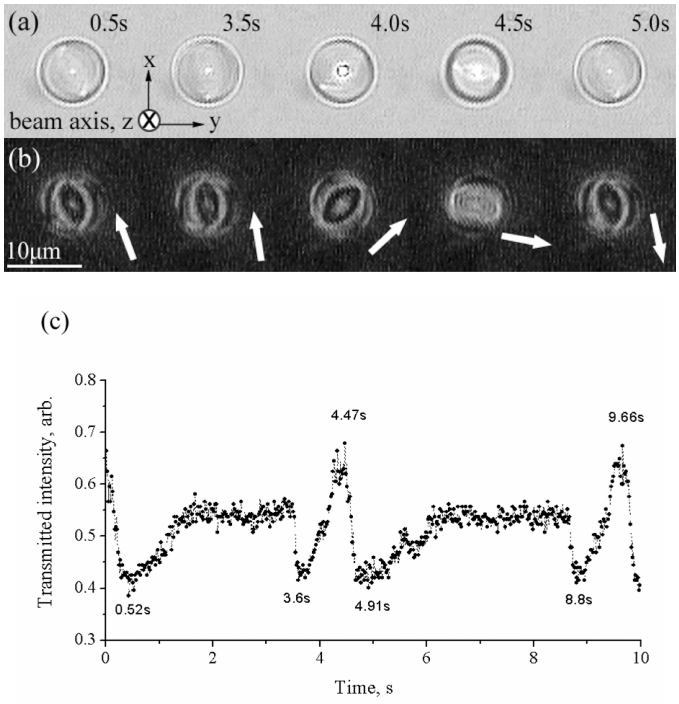

Fig. 2. Details of the time-evolution of rotation for a twisted bipolar chiral nematic droplet in a linearly-polarized laser trap. The natural pitch of the droplet is 7.8 μm and the droplet diameter is 8.2±0.4 μm. (a) shows images with uncrossed polarizers in which the vertical motion of the droplet is apparent, *z* is the beam axis; (b) shows equivalent images under crossed polarizers; the white arrows indicate the orientation of the droplet director. (c) shows the light transmission of the rotating droplet measured between crossed polarizers, corresponding to the photomicrographs in (a) and (b).

We now consider the mechanism that is causing the rotation of the chiral droplets. An analysis of the contributions to the optical torque shows that there are terms resulting specifically from the chiral structure that could induce a torque. In a chiral, anisotropic medium subject to a time-dependent electric field, $D_i = \varepsilon_{ij} E_j + g_{ij} H_j$, where D_i, E_j and H_j are the components of the displacement, **D**, the electric field, **E** and the magnetic field, **H**, and g_{ij} is the gyration tensor that accounts for the optical activity of the medium [17]. Consider an anisotropic chiral droplet that is uniaxial for simplicity, in which the director is along the optical axis. The dielectric tensors can then be written in the form $\varepsilon_{ij} = \varepsilon_o \delta_{ij} + \varepsilon_a n_i n_j$ and $\varepsilon_{ij} = g_o \delta_{ij} + g_a n_i n_j$, and the torque can be expressed as $\Gamma = \mathbf{E} \times \mathbf{D} = \varepsilon_a (\mathbf{n} \cdot \mathbf{E})(\mathbf{n} \times \mathbf{E}) + g_o (\mathbf{E} \times \mathbf{H}) + g_a (\mathbf{n} \cdot \mathbf{H})(\mathbf{n} \times \mathbf{E})$. The first term is the standard dielectric torque, the second is a chiral torque, which *could* result in the rotation of a chiral droplet and the third term is an insignificant correction proportional to the anisotropy of the local optical activity. The 'dielectric' torque is exactly the torque that rotates a non-chiral droplet until the director is parallel to the field; in the case of a chiral droplet, this torque is still important but the final orientation of the droplet with respect to the field will depend on the average dielectric axis in the droplet. If the contribution from the chiral torque was sufficiently large, it could make this orientation unstable over time and cause rotation of a

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chiral droplet. However, although this analysis clarifies the unusual nature of chiral systems, it applies to all of the droplets we consider experimentally. Since continuous rotation is observed only for critical droplet sizes, another mechanism must be responsible.

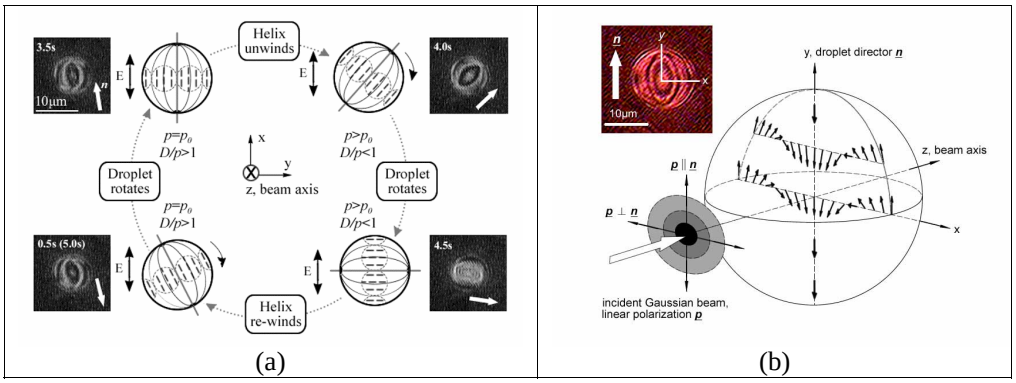

Fig. 3. (a) The proposed cyclic mechanism for droplet rotation. The electric field from the laser trap induces helix unwinding in the droplet, shown in (b). This causes a change in *D/p* which then changes the stable droplet orientation (see Figure 1). Once the new stable orientation has been reached, the helix resumes its natural state, once again moving the stable droplet orientation and the process continues indefinitely.

Consider a droplet of natural pitch p_o that satisfies the condition D/p_0 slightly larger than 1 (the argument for $D/p \sim 0.5$ is equivalent). Such a droplet will be trapped and, as shown in Figure 1, will rotate to an orientation such that the droplet axis is approximately parallel to the polarization plane of the incident laser. We suggest that the optical field density in the centre of the droplet due to the laser trap (the size of which is diffraction limited in the medium) is sufficiently great to partially unwind the helix [15, 16]. Because of this change in pitch, *D/p* will have decreased below axis and so no longer acts to unwind the helix. Thus the original pitch will be restored through an elastic relaxation process. The droplet is again in a condition to rotate and the cycle continues. This mechanism would certainly result in a non-linear rotation rate, as observed. It would also only be possible to continuously rotate droplets with a natural pitch just slightly larger than the droplet diameter (or radius); these critical droplet sizes are also as observed. The fact that no rotation is observed in longer pitch droplets, which also do not experience sudden changes in stable droplet orientation as *D/p* changes, also supports this argument. One notes that the chiral torque, created by the linearly polarized light in a chiral medium, still plays a role in this mechanism. It is responsible for the continuous rotation in the same direction and does not allow the droplet to stop at any intermediate angle.

The vertical motion of the droplet can be explained straightforwardly by considering the optical forces that hold the droplet in a position close to the focus of the trap. Because the laser trap produces an inhomogeneous distribution of intensity proportional to $\mathbf{E}^2$, there is a force along the gradient, which moves the droplet into the equilibrium position of maximum electric energy. Our system employs linearly polarized light, which gives a contribution to the effective pressure and the force described by this term. The force depends on the coupling between **E** and **D**, and as the relation between **E** and **D** changes with time (because the droplet and hence the dielectric tensor rotates), the position of the droplet in the trap will change slightly. A similar motion has been reported only once previously [18], resulting from changes in birefringence in nematic droplets.

In conclusion, we have described the continuous, non-linear rotation of chiral nematic droplets in a linearly polarized laser trap. The rotation occurs only for small droplets of a critical size and is accompanied by a distinct vertical motion of the droplet in the trap. The

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mechanism responsible for the anomalous motion of the chiral droplets is quite different from any previously reported that result in the rotation of birefringent particles in optical traps. It relies on a photo-induced molecular rotation that induces a critical reorganization of the liquid crystal in the droplet, followed by elastic relaxation of the photo-induced structure. A cycle of molecular reorganization and relaxation follows. This fascinating combination of continuous rotation and linear motion in laser traps has not been seen before, and could have interesting implications for microfluidics.

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