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High-efficiency optical transfer of torque to a nematic liquid crystal droplet

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We report on the determination of the difference in the refractive index between the ordinary and extraordinary rays in droplets of nematic liquid crystal E-44 at the wavelength of laser trapping, 1064 nm. This difference was calculated by measuring the ellipticity of laser tweezers for the moment at which rotation of the trapped droplet starts. The precision of this determination of the refractive index difference is approximately 5×10^{-3}. Hydrophobic nematic liquid crystal E-44 formed 0.5–5-μm droplets when cast into D_2O. The efficiency of the transfer of light torque to the liquid crystal droplets was determined to be 70% for the no-slip-boundary condition. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1588366]

The momentum and angular momentum of light can be transferred to an object by light absorption and refraction. The angular momentum carried by light can be characterized by the "spin" angular momentum associated with circular polarization[1] and the "orbital" angular momentum associated with the spatial distribution of the wave.[2] Each photon of a Gaussian circularly polarized beam has an angular momentum $\sigma\hbar$, where $\sigma=\pm1$ for the left- and right-handed polarizations, respectively. The equation of motion that links torque $\mathbf{\Gamma}$ exerted on the object with its angular velocity $\mathbf{\Omega}$ and moment of inertia I is then $\mathbf{\Gamma}=I\,d\mathbf{\Omega}/dt+D\mathbf{\Omega}$, where D is the damping factor (the drag coefficient for the rotational movement). For a spinning sphere $D=8\pi\eta r^3$ and $\frac{32}{3}\eta r^3$ for a disk of the radius r in the medium of viscosity η. In the steady state, the time derivative drops out and the terminal angular velocity is proportional to the torque divided by the damping factor, which is highly dependent on the object's shape and the properties of the environment in which it moves.

Gauthier[3] evaluated numerically the torque acting on an object shaped as a four-blade windmill trapped by single-beam laser tweezers. Calculations well explained the earlier reported experimental results[4] on the spinning of such an object in a laser trap (0.37-Hz rotation was observed at 80-mW laser power). The direction of spinning can be reversed by inversing an optical contrast $n_{\mathrm{rel}}=n_s/n_m$, which is the ratio of the refractive index of a sample (n_s) to that of the surrounding medium (n_m).[3] An extended theory[5] predicts that the direction of rotation can be controlled by adjusting the spot size of illumination. Another approach was introduced for the optically driven rotor.[6] Two polystyrene beads of 2 and 0.94 μm in diameter were linked via avidin-biotin binding. The smaller bead was half-coated with gold/palladium. This geometry allowed the harvesting of the gradient pressure of a laser trap made by a focused linearly polarized beam, as well as the conversion of this pressure into a spin torque. An average rotation frequency of 2.6 Hz was observed at 29-mW laser power ($\Gamma\simeq10^{-19}$ Nm). Rotation frequencies higher than 10 Hz were reported for a 3–4-μm windmill-type structure at 20 mW.[7] The complete transfer of angular momentum from light to a particle is expected for a 100% absorbing particle. However, the absorbing particles have shortcomings in practical applications due to overheating and unwanted axial forces. Higher rotation frequencies can be obtained when birefringent particles are used, as was demonstrated in the case of grinded calcite particles[8] and nematic liquid crystal (LC) droplets[9,10] laser manipulated in water.

Here, we report on a determination of: (i) the difference of refractive indexes between ordinary and extraordinary rays in droplets of nematic E-44, and (ii) the efficiency of torque transfer from light to a droplet. The difference of refractive indexes is a key factor in the transfer of angular momentum from a circularly polarized light to a laser-trapped particle. A high-efficiency (70%) angular momentum harvesting by birefringent LC droplets is demonstrated when the no slip boundary condition was assumed.

The typical setup to perform all-optical switching experiments by laser-trapped droplets of LCs is shown in Fig. 1. A combined angular tuning of λ/2 and λ/4 plates allows the precise control of the polarization of the laser beam at the focus. Both wave plates are necessary to compensate for a birefringency introduced by the dichroic mirror. The polar-

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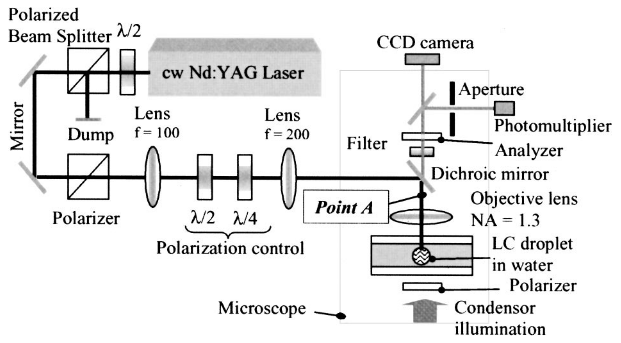

FIG. 1. (a) Setup for optical manipulation of liquid crystal droplets. The eccentricity ϵ of the laser beam is determined at point A.

ization was analyzed by a polarizer prism at the point A before the objective lens.

Hydrophobic nematic liquid crystal, E-44 (Merck), forms droplets of 0.5–5-μm diameter with a bipolar internal structure when dispersed in heavy water (D_2O). In order to avoid absorption at elevated laser powers, D_2O is a preferable environment for laser manipulation, since the absorption at $\sim$1-μm wavelength is almost an order of magnitude lower than that in H_2O.[11] Almost no temperature rise ($<2\,°C$) at the focal volume of $1\,\mu m^3$ was observed at 0.5-W laser power in D_2O.[11]

Most droplets possess a polar-like molecular arrangement due to the so-called anchoring effect; that is, molecules at the outer regions tend to be oriented along the surface of the droplet, while those at the inner regions tend to sustain their bulk-like orientation. This brings about the final polar-like (bipolar) structure of a droplet. However, some LC droplets have a radial and irregular segmented structure. When viewed in a polarizing microscope,[9,10] the image is that of the so called Maltese cross, as shown in Fig. 2(a). The dark regions correspond to those areas where the passing light is least scattered (depolarized), and vice versa. However, the distinction between the bipolar and radial structures is not straightforward from such imaging, since both molecular orientations cause similar cross-like images. The internal molecular arrangement can be further distinguished by judging the response of a LC droplet to laser manipulation.[9,10] The underlying mechanism of the response can be understood in terms of the birefringency of the droplet. The particles with a polar orientation can be easily laser-manipulated {oriented to a preset angle [Fig. 2(a)] or spun [Figs. 2(b) and 2(c)]} by the polarization control of a laser beam.[9,10] The spinning can be evidenced by a viscous drag of the nearby free-floating LC droplet,[9] as shown in Figs. 2(b) and 2(c). Eight video frames were combined using the image processing routines (MATLAB 6.1) in order to superimpose consecutive snapshots, while in the central polygon (c), the image of a spinning droplet from the first frame (b) was numerically inserted [recognizable by a contrast difference in (c)]. If the structure of the LC droplet is perfectly radial, there should be no birefringence, and the droplet should not respond to a change in the polarization of the trapping beam.[12] The only mechanism of torque in such a case would lie in the astigmatism of the laser trapping beam.[13]

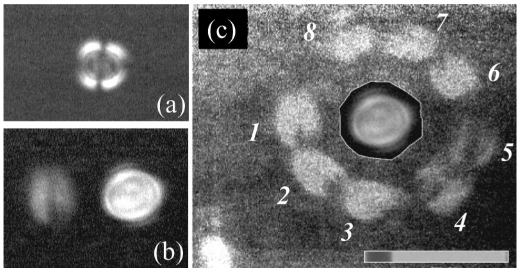

FIG. 2. Video snapshots of a E-44 liquid crystal particle trapped by linearly (a) and circularly [left particle in (b)] polarized tweezers. (c) Combined image of eight consecutive snapshots made with 33-ms separation. Numbers mark the locations of a dragged particle; image shown in (b) corresponds to that marked by *1* in (c). (Scale bar, 10 μm.)

The frequency of optical switching can be measured experimentally by the setup shown in Fig. 1, where the frequency of optical transmission modulation is fourfold that of the droplet's rotation:

$$f_{\mathrm{opt}} = \nu \cdot 4 f(d,\phi), \tag{1}$$

where the factor ν accounts for the efficiency of torque transfer from light to a droplet, and $f(d,\phi)$ is derived according to Friese *et al.*[8] [see Eq. (4)]. Let us consider an elliptically polarized laser beam incident on a particle. Elliptically polarized light can be described by $\mathbf{E} = E_0 e^{i\omega t} \cos\phi \cdot \mathbf{x} + i E_0 e^{i\omega t} \sin\phi \cdot \mathbf{y}$, where ω is the angular frequency, E is the electric field of light, and x with y are Cartesian coordinates defining the lateral spot size. Here, ϕ describes the degree of ellipticity of the light ($\phi = 0; \pi/2$ indicates plane-polarized and $\phi = \pi/4$ indicates circularly polarized light). The ellipticity phase angle ϕ is related to the experimentally measured eccentricity of the laser beam by $\phi = \tan^{-1}\epsilon$, where the $\epsilon = \sqrt{P_{\min}/P_{\max}}$ is defined by the ratio of minimum and maximum laser power passed through a polarizer (the angular dependence of the passed laser power is measured).

To calculate a change in the angular momentum of the light after it passes through a birefringent material, the incident elliptically polarized light is first expressed in terms of components parallel and perpendicular to the optical axis of the material:

$$\begin{aligned}\mathbf{E} \equiv \mathbf{E}_i + \mathbf{E}_j = {} & E_0 e^{i\omega t}(\cos\phi \cos\theta - i \sin\phi \sin\theta)\mathbf{i} \\ & + E_0 e^{i\omega t}(\cos\phi \sin\theta + i \sin\phi \cos\theta)\mathbf{j},\end{aligned} \tag{2}$$

where θ is the angle between the fast axis of the quarter-wave plate producing the elliptically polarized light and the optical axis of birefringent material. The phase shift due to passing through a thickness d with refractive index n is kdn, where $k = 2\pi/\lambda$ is the free space wave number. Thus, the emergent light field will be: $\mathbf{E} = \mathbf{E}_i e^{ikdn_e} + \mathbf{E}_j e^{ikdn_o}$, where $n_{e,o}$ are the refractive indexes of extraordinary and ordinary rays, respectively. The angular momentum of a plane electromagnetic wave is defined as $\mathbf{J} = [\epsilon/(2i\omega)] \int d^3 r \mathbf{E}^* \mathbf{E}$, where $\mathbf{E}^*$ marks the complex conjugate of electric field vector $\mathbf{E}$, and integration is made over all spatial elements $d^3 r$. The changes in the angular momentum of the light cause a reaction torque per unit area on the thickness d of material:

$$\begin{aligned}\tau = {} & -\frac{\epsilon}{2\omega} E_0^2 \sin kd(n_0 - n_e) \cos 2\phi \sin 2\theta \\ & + \frac{\epsilon}{2\omega} E_0^2 (1 - \cos kd(n_0 - n_e)) \sin 2\phi.\end{aligned} \tag{3}$$

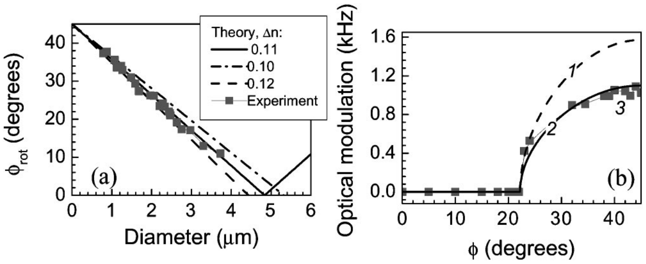

FIG. 3. (a) Determination of effective refractive index change of a birefringent spherical particle [Eq. (5)]. (b) Frequency of the optical modulation by a laser-manipulated liquid crystal droplet versus the ellipticity of light, ϕ [Eqs. (4), (1)]. The diameter of a droplet was $d=2.45\pm0.08$ μm, the laser power at the focal point was 198 mW, and the environment was heavy water. Linear velocity for a 1-kHz switching frequency corresponds to 2 mm/s at the angular velocity of 1570 rad/s.

The first term in Eq. (3) is the torque due to the plane-polarized component of elliptically polarized light; we can call this the alignment torque. Meanwhile, the second term of Eq. (3) is due to the change in polarization caused by passage through a birefringent media. For a plane-polarized light, $\phi=0$ or $\pi/2$, the torque on the particle is proportional to $\sin 2\theta$. The particle will experience torque as long as $\theta \neq 0$, and will be at equilibrium when the fast axis of the crystal is aligned with the plane polarization ($\theta=0$).

For a steady-state spinning particle, the torque exerted on the particle by light [Eq. (3)] is counterbalanced by the environment's drag torque according to $\tau=D\Omega=D\times 2\pi f$ when the no-slip-boundary condition was assumed. The frequency of a particle's rotation (f) for a trapping power P and optical frequency ω is

$$f(d,\phi)=P\sqrt{A}/(2\pi\omega D), \tag{4}$$

where

$$A=[1-\cos kd(n_0-n_e)]^2\sin^2 2\phi$$
$$-\sin^2 kd(n_0-n_e)\cos^2 2\phi.$$

The fastest spinning is observed when particle thickness corresponds to a λ/2 plate. Hence, the periodic dependence of frequency versus particle size is observed. Every local maximum in the rotation frequency obeys the dependence on the particle size $f\propto r^{-3}$, which follows from Stokes' law. The rotation of a birefringent calcite microparticle of ~6-μm radius was demonstrated[8] at 50-mW laser trapping, but the rotation frequencies were only a few hertz due to a large viscous drag produced by irregularly shaped particles. The frequency limit for the smaller particles is actually imposed by the size of a laser spot, which at its diffraction limit approaches a diameter of 1.22λ/NA (NA is the numerical aperture of the objective lens, typically NA is 0.8–1.3); the frequency is also limited by increased light scattering, which decreases the efficiency of torque transfer from the laser beam to the particle [the factor ν in Eq. (1)].

The typical refractive index difference between the ordinary and extraordinary rays in bulk birefrigent nematic LCs is $\Delta n=0.2$ ($\Delta n=0.256$ for E-44). When a nematic LC is spherical, its molecular arrangement changes. Thus, the effective refractive index contrast Δn needs to be established experimentally. This can be done by measuring the ellipticity of a laser beam (angle ϕ) at which a birefringent particle starts to spin. This can be found from Eq. (3) for the case when the alignment torque (the first term of the equation) is maximum and the total torque is zero. This is when

$$\phi_{\rm rot}=(\pm m\pi\mp kd\Delta n)/4,\ \text{where}\ m=1,2,3,\ldots. \tag{5}$$

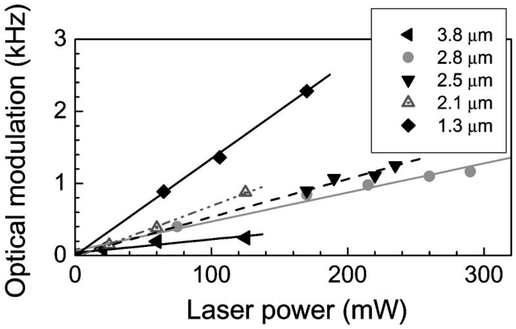

FIG. 4. Dependence of the frequency of optical transmission modulation on the trapping laser power for nematic E-44 LC droplets of different sizes. LC droplets were laser manipulated in D_2O (heavy water was used to exclude any absorption at 1064 nm).

The best fit of experimentally measured $\phi_{\rm rot}$ gives the value of effective refractive index, $\Delta n=0.11$, with a precision greater than 5×10^{-3} [Fig. 3(a)]. With this established value of Δn, the experimentally observed dependence of optical switching frequency $f_{\rm opt}$ on the degree of ellipticity of a laser beam can be fitted by Eq. (1), as shown in Fig. 3(b), where the curve *1* is plotted for an ideal 100% torque transfer from a laser light to a spherical droplet. The experimental data of $f_{\rm opt}$ can be fitted by Eq. (1) with efficiency factor $\nu=0.7$ [curve *2* in Fig. 3(b)]. Considerably high frequency of optical switching for a mechanical system can be achieved by a rotating LC droplet [Eq. (4)], as shown in Fig. 4.

We have demonstrated a technique to precisely measure the changes in the refractive index between ordinary and extraordinary beams in droplets of nematic liquid crystal E-44. The same measurement allows us to calculate the efficiency of torque transfer from circularly polarized light to a droplet, which was found to reach 70%.

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