

Laser manipulation based on a light-induced molecular reordering

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Abstract: We report on a novel principle of actuation of micrometer-sized liquid crystal droplets. It is based on a light-induced reordering of liquid crystal molecules inside the droplets. Polariscopes allowed to evaluate the birefringence change inside the micro-droplets. Directional actuation of the trapped droplet was achieved by cycling laser power with the direction defined by the polarization of the tweezing beam. Micro-actuation resulted from optically-induced birefringence; i.e., a nonlinear optical effect was utilized for mechanical manipulation of the micro-droplet. This principle of actuation can be used to induce molecular flows in sub-micrometer volumes.

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References and links

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Control over actuation and positioning, exerted force and torque are basics for the development of macro- or nano/micro-mechanical tools, characterization methods, and machinery. Current technologies, especially those related to bio-oriented research, require such tools for microscopic characterization, imaging, and fabrication at 0.1 – 100 μm feature sizes. The application of laser tweezers [1] has been increasingly successful in these fields, providing nm-fN resolution [2].

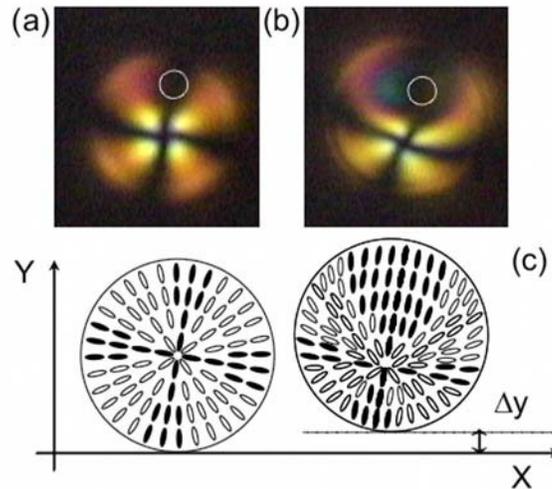


Fig. 1. Droplet of liquid crystal 5CB (radial structure) trapped at 62 (a) and 310 mW (b), respectively, in linearly (vertically) polarized laser tweezers. Diameter of droplet $5.2 \mu\text{m}$. Circles depict approximate diameter at the focus. (c) Schematic presentation of internal molecular alignment and principle of actuation.

Here, we report on an optically-controlled actuation of sub-micrometer-sized volumes of molecules ordered by the laser trapping beam. This principle is demonstrated by a polarization-controlled positioning and directional actuation of a laser-trapped liquid crystal droplet. The method is applicable for any micelle forming dipole molecules, whose alignment the resulting birefringence can be optically controlled by changing the laser trapping intensity.

Micrometer-sized droplets of nematic liquid (LC) crystal 4-pentyl-4'-cyanobiphenyl (5CB, Aldrich) were used in our experiments. Molecular alignment of nematic liquid crystal inside droplets renders them optically anisotropic; hence, they possess birefringence, which is the key optical property allowing the angular momentum transfer from light to a micro-object trapped by laser tweezers [3, 4]. Birefringent micro-objects can harness the angular momentum of light, i.e., are responsive to the polarization of the trapping beam; thus, optical alignment and spinning become possible [5, 4, 6]. The 5CB droplets in water (H_2O or D_2O) have a typical diameter distribution of $1 - 10 \mu\text{m}$ and their internal molecular alignment is bipolar and homeotropic-like. Droplets with internal radial (homogeneous-like) molecular alignment were obtained by the addition of the cationic surfactant cetyl-trimethyl-ammonium bromide (CTAB) at concentrations just above that of the critical micelle concentration of $\approx 7 \mu\text{M}$. The surfactant decreased the surface tension, allowing for LC molecules to acquire a more isotropic radial structure. D_2O was used as a solvent to exclude absorption at the laser trapping wavelength of 1064 nm ; its absorption coefficient is $\alpha \approx 0.3 \text{ cm}^{-1}$, i.e., ten times smaller than that of water H_2O . Spring constant of the tweezers was proportional to the laser trapping power and was $k \approx 10 \text{ nN/m}$ for 100 mW and droplet of $4 \mu\text{m}$ in diameter. It was measured directly by oscillation transients around the trapping center when laser was switched on.

Figure 1 shows a view of the lateral (vertical) displacement of the radial droplet imaged through a crossed polarizer and analyzer, a polariscope. The intensity of light transmitted through a polariscope with birefringent material in between is given by [7]:

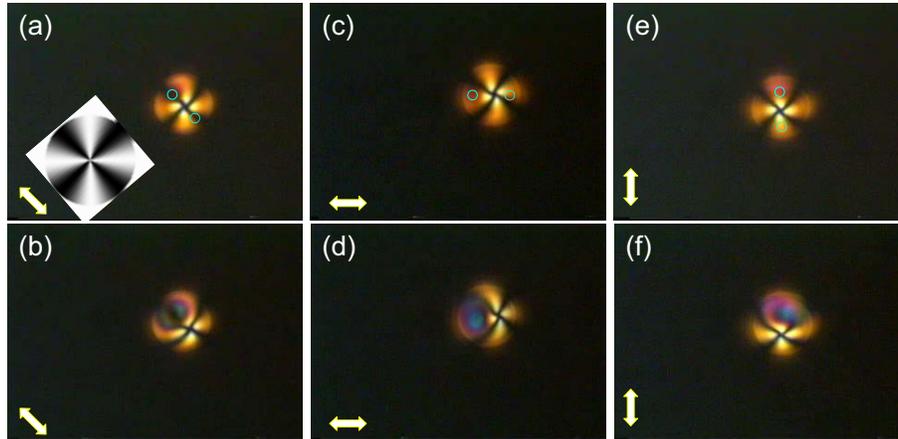


Fig. 2. Polariscope images of the radial 5CB droplet trapped at 62 mW (a, c, e) and at 300 mW (b, d, f), respectively. Polarization is shown by arrows. The circle marks show two possible alternative laser trapping spots. Diameter of the droplet, $4.4 \mu\text{m}$. The inset in (a) shows a simulated "Maltese" cross (Eq. 1), the intensity distribution through the crossed polarizer and analyzer with a birefringent material in between.

$$I(\theta, \Delta n) = I_0 \sin^2(2\theta) \sin^2(\pi n d / \lambda), \quad (1)$$

where θ is the angle between the polarizer and the fast axis of a birefringent sample, $n = n_e - n_o$ is the birefringence ($n > 0$ for 5CB), d is the thickness of sample, and λ is the wavelength of light used for imaging. Equation 1 represents the well known "Maltese" cross intensity distribution (inset in Fig. 2(a)), where the principle directions on the fast axis of the birefringent sample coincide with the dark lines and points ($\theta = 0$) in the polarization micrograph. The brightest regions are those where the strongest depolarization due to light scattering took place, at the $\theta = \pm\pi/4$ angles in the image.

When laser power increased, the droplet was moved along the polarization of the trapping beam (Fig. 2). An increased birefringence, n , due to the molecular dipolic alignment, favored the relocation of the laser trapping center (Fig. 1(c)). At any given orientation of linear polarization, two out-of-center locations of stable laser trapping exist (marked by circles in Fig. 2). The stable trapping occurs at the locations where the most laser beam occupies the volume with preferentially same director orientation (it has the largest birefringence). This is because of the hedgehog-type molecular alignment inside the droplet. The director has the most changing direction in the droplet's center, hence, there is no stable trapping position at the center. A free-moving droplet can be trapped at either of those two points (Fig. 2) by a targeted illumination. If the droplet is trapped at some other location, it usually re-locates itself to one of those points of stable trapping within few milliseconds. Hence, once trapped, the droplet can be actuated along the direction of polarization by changing laser trapping power (i.e., changing the volume where molecules are aligned). When the larger volume has the same molecular orientation (director) at higher laser power it becomes a new center of laser trapping. For the outside observation, it appears that the droplet moves along the polarization when laser trapping power is cycled. The polarization can be tuned to the desired orientation by a $\lambda/2$ -plate set into the incident beam and allowing to achieve lateral displacement in any direction. The shape and color changes in the polariscope images (Figs. 1-2) signify the corresponding texture alterations due to the molecular re-alignment inside the liquid crystal droplet.

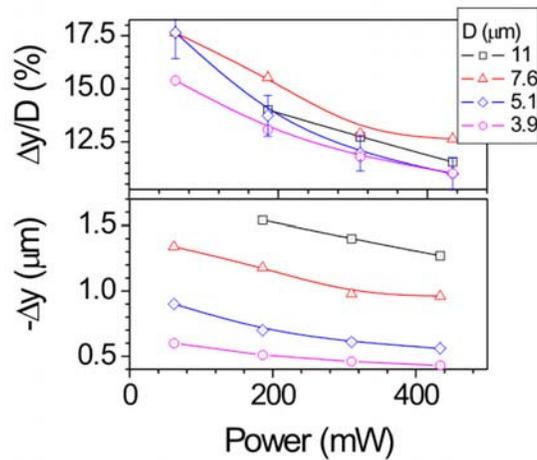


Fig. 3. Relative (a) and absolute (b) actuation by a laser-trapped radial 5CB droplet vs. the trapping power for droplets of different diameters, D . Polarization of tweezers was linear.

The actual light path inside the droplet is complicated; a simple Gaussian beam tracing can only serve as an estimate. Indeed, the Mie scattering strongly affects the intensity distribution inside/outside the droplets [8], since their typical size parameter is $x \equiv 2\pi r/\lambda \simeq 10$. Despite the unknown actual light intensity distribution inside the droplet, its polariscope image shows homeotropical alignment, with the director prevalently perpendicular to the surface of the droplet. Hence, the beam of laser tweezers mostly propagates along the director (slow axis of the droplet) forming the Maltese cross image (Fig. 2). According to Eq. 1, the same image would result from a plane wave passing through a plate of birefringent crystal. For example, the bipolar alignment would form another image resembling that of homogeneous alignment with a director parallel to the surface of the droplet [4].

The consequence of the light-induced molecular realignment causes a ponderomotive action when applied to the micrometer-sized objects. Figure 3 shows a summary of the actuation data with droplets of different sizes. If the absolute actuation stroke is normalized by the diameter of the droplet, a similar behavior is evidenced (Fig. 3(a)) for all droplets. The uncertainty of the results is primarily caused by the error of measurements of the diameter and displacement. The trapped particles showed reproducible actuation with the same stroke-power dependence and no degradation.

The ordering of LC molecules inside a nematic LC in an applied electric field takes place when the strength of field is higher than a certain critical value, the Freedericksz limit, for the homeotropical director perpendicular to the electric field. This limit is the maximum field strength necessary for a field-induced alignment of LC molecules. There is no such limiting E-field strength for the other dipole orientation with respect to the field [9]. Once the dipole interaction of LC molecules with the electric field is strong enough to overcome the elastic and visco-elastic forces, molecular re-alignment occurs. Let us compare the field strengths in our laser trapping experiments with the Freedericksz limit, which can be calculated by balancing the elastic free energy with the free energy of the LC in the electric field $E_i = \frac{\pi}{d} \sqrt{\frac{K_i}{\epsilon_0 \Delta\chi}}$ [9], where $K_{1,2,3}$ are the Frank elastic constants for splay, twist, and bend deformations, respectively, and

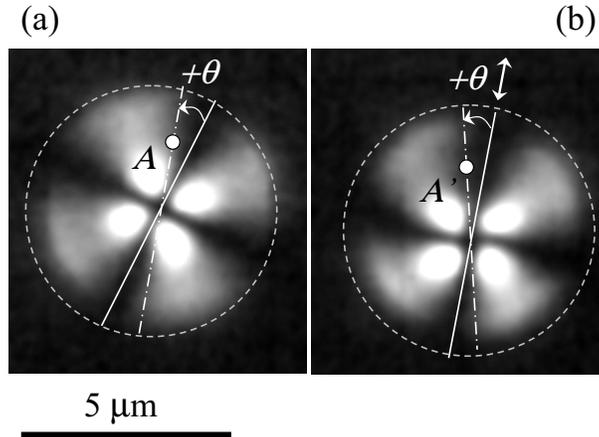


Fig. 4. Optical transmission images of a radial 5CB droplet observed through a pair of crossed polarizers at laser trapping power 0 (a) and 62 mW (b). Circles at points A, A' mark the location on the droplet where transmission measurements were carried out. Arrowed line in (b) shows the polarization of the laser tweezers. A CCD camera was calibrated for a linear response.

d - thickness of LC, $\Delta\chi = \chi_{\parallel} - \chi_{\perp}$ is the anisotropy of susceptibility, while susceptibility is related to the dielectric constant by $\epsilon_{e,o} \equiv \tilde{n}_{e,o}^2 = 1 + \chi_{\parallel,\perp}$. For typical values of 5CB material $\Delta\chi = 0.6$, $K \simeq 10$ pN, and $d = 10$ μm , one can find $E \simeq 0.5$ V/ μm . In our experiments, the lowest laser power of laser trapping was about 30 mW, which corresponds to the strength of the electric field $E = \sqrt{2I/(\epsilon_0 cn)} = 0.75$ V/ μm , when focused into a spot with a diameter equal to 2λ (a good approximation for our experimental conditions). This is already over the Freedericksz limit. Thus, optical ordering should be present when an LC droplet is held in the laser tweezers and the molecular alignment is expected to occur.

The polariscope image showed the onset of internal ordering at a laser power of about 60 mW (Fig. 4). The dark region to the right of point A' is a new relocated laser trapping center, where the light-induced ordering of LC molecules took place. Due to the ordering, the region appears dark; also, the initial radially symmetric Maltese cross became distorted. The molecular ordering in an optically applied electric field is, in fact, an optical nonlinear effect of giant optical nonlinearity [10]. Let us evaluate the relevant value of nonlinear susceptibility $\chi^{(3)}$. The transmission through a birefringent particle is given by Eq. 1. We measured the transmission at point A, T_A , (Fig. 4(a)) for a 5CB droplet at minimum laser trapping power and fitted it by Eq. 1. The angle θ was measured directly as the azimuth angle from the transmission minimum. This value of θ defines the angle between the unperturbed director and the direction of polarization. Thus, the value of n can be found for the known particle diameter d . In this way a functional relation $T = f(\theta)$ was established for the fixed value of n . For the known laser power transmission, $T_{A'}$, the point A' was again fitted by Eq. 1, only now, the angle was $\theta' = \theta \pm \Delta\theta$ and $n' = n \pm \Delta n$.

In order to calculate the light-induced rotation of the director, $\Delta\theta$, and the corresponding change in birefringence, Δn , we must use an additional functional relationship between the two. Such a relationship has been established for a homeotropically aligned nematic liquid crystal [9]. When the thickness of the crystal is d , the optically induced rotation of the director

$\Delta\theta$ along the propagation of light (z-axis) is given by [9]:

$$\Delta\theta(z) = \frac{1}{4\xi^2} \sin(2\theta)(dz - z^2), \quad (2)$$

where $\xi = 4\pi K_1 / (\Delta\epsilon \langle E_{op}^2 \rangle)$ and $\Delta\epsilon = \chi_{\parallel} - \chi_{\perp} = \Delta\chi$. At the same time, the light induced refractive index defined by $\tilde{n} = n_0 + n_2 I$ for the same geometry is given by:

$$n_2(z) = \frac{(\Delta\epsilon)^2 \sin(2\theta)^2}{4K_1 c} (dz - z^2). \quad (3)$$

By use of Eqs. 2 and 3, the laser-induced change of birefringence can be calculated as the function of director rotation, assuming $n_2 \equiv \Delta n$. This is the nonlinear refractive index change induced by the light field of the linearly polarized laser tweezers. We can now calculate Δn from the measurement of the transmission change at point A:

$$T_A - T_{A'} = \sin\left(\frac{\pi d n}{\lambda}\right)^2 \sin(2\theta)^2 - \sin\left(\frac{\pi d (n + \Delta n)}{\lambda}\right)^2 \sin(2(\theta + \Delta\theta))^2. \quad (4)$$

Equation 4 defines a way of measuring the light-induced refractive index change due to director reorientation from the experimentally measured transmission change. If we define polarization as $P = \epsilon_0 \chi^{(1)} E + \epsilon_0 \chi^{(3)} EEE$, the nonlinear refractive index is given by $\Delta n = \chi^{(3)} I / (\epsilon_0 n_0 c)$ [9], where $\chi^{(3)} [(m/V)^2] = 4\pi (3 \cdot 10^4)^{-2} \chi^{(3)} [esu]$. For a 5CB LC droplet (Fig. 4), we found the value of $\chi^{(3)} = (1.0 \pm 0.3) \times 10^{-8}$ esu, which is about two times larger than the known bulk constant of 5CB, because this evaluation is strictly correct for isotropic media only.

In conclusion, we have shown that an optical ordering of LC molecules can be achieved inside droplets with diameters smaller than 10 μm . This ordering yields a birefringence change via giant optical nonlinearity at the focal spot of the laser tweezers exerting a force large enough to actuate the laser-trapped droplet. Also, elastic properties of "soft" (ductile) materials can be investigated by applying calibrated force or angular momentum using polarization-controlled laser-tweezers with polariscope imaging.