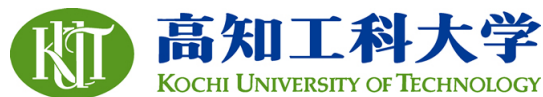


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Optically Induced Rotation of Microcylinders Made of Photopolymerizable Nematic Liquid Crystal

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We report for the first time the use of photopolymerizable liquid crystals in fabricating microsized cylinders that are rotated by a circularly polarized laser beam. They were fabricated by two-photon photopolymerization from liquid crystals whose molecules had been deliberately oriented on a rubbed surface. The rotational torque showed similar oscillatory structures to those observed in the intensity of light transmitted through crossed Nikols as a function of cylinder height. From the structures, the birefringence of the cylinders was estimated and the main mechanism underlying the rotational torque transfer was determined.

The linear and angular momenta of photons can be transferred to micro- or nanoparticles when light is absorbed or refracted. Since the pioneering work of Ashkin¹⁾, the refraction of linear momentum has been widely used to trap and manipulate small particles. Dissymmetrically shaped micro-objects have been shown to rotate in optical traps by means of radiation pressure²⁻⁶⁾. The transfer of photon angular momentum to various types of objects has also been a subject of interest since the early experiments of Beth⁷⁾, who measured the spin angular momentum transferred from circularly polarized light to quartz plates. Each photon in a circularly polarized light beam carries an angular momentum of $\pm\hbar/2\pi$, depending on the handedness of the polarization. Friese *et al.*⁸⁾ showed that an optical torque is induced on microscopic birefringent particles of calcite when held by optical tweezers. In contrast to such types of spin angular momentum, another type of angular momentum sometimes referred to as “orbital angular momentum” has also been investigated^{9, 10)}.

Since 1999, the manipulation of liquid crystals (LCs) by laser tweezers has been investigated exclusively in droplet form¹¹⁻¹³⁾. Since LC droplets have inherent birefringence, the polarization of circularly polarized light passing through such droplets changes accompanied by the transfer of angular momentum for the conservation of angular momentum.

In the present work, we used polymerizable LC materials that solidified when irradiated with UV light to fabricate microsized rotors. We investigated the characteristics of the rotational torque that developed when circularly polarized light was used. To the best of our knowledge, this is the first investigation of the rotational driving of microphotofabricated structures made of LCs.

The fabrication system used a mode-locked Ti:sapphire laser (Spectra Physics,

Tsunami), which provided a wavelength of 730 nm and a pulse width of 130 fs at a repetition rate of 83 MHz¹⁴⁾. Instead of using UV light, however, ultrashort pulses of near-infrared laser light were used which made it possible to fabricate structures with high resolution via two-photon absorption^{15,16)}. The laser beam was sent to an inverted microscope (NIKON, TE-2000) and tightly focused with an oil-immersion objective of 100× magnification and high numerical aperture (NA = 1.3). After passing through the objective, the laser power was 8 mW. The focal spot was steered in the horizontal plane by a pair of computer-controlled galvanomirrors. A focusing unit (Chuo Precision, MSS-FMC,) was attached to the microscope, enabling control of the vertical position of the objective. The accuracy in the vertical positioning of the focal point was 0.1 μm. Figure 1 shows the thus-fabricated cylinder of 5 μm diameter, observed using a secondary electron microscope, together with a micro-wheel gear to give insight into the fabrication accuracy.

The nematic LC materials were prepared by mixing a 4 wt.% solution of photoinitiators (BASF, Lucirin TPO) in reactive monomer LCs (BASF, Paliocolor LC242) as the main component. Since the materials change to nematic phase at 63-70 °C, they were heated just before photopolymerization to change them into LC phase. On cooling, they keep the LC phase with supercooling even below 30 °C¹⁷⁾. They consist of rod-like nematic LC molecules with attached chains, which carry polymerizable endgroups. They are transparent to near-infrared light, but absorb photon energy via a two-photon process and can thus be photopolymerized. A drop of the LC monomer mixture was placed on a glass plate that had been covered in advance with a polyvinyl alcohol (PVA) film and rubbed so as to align the LC molecules in the rubbing direction¹⁸⁾. Photopolymerization was started from a point 30 μm above the glass

surface, and the focal spot was moved downward at 0.3 μm intervals. In this way, several circular cylinders with a diameters of 5 μm and heights ranging from 7 to 22 μm at 1 μm intervals were fabricated. The cylindrical form was selected in the present study to avoid unwanted effects of the rotational drive supplied by radiation pressure and to observe pure spin effects of circularly polarized light. After photofabrication, the unsolidified LC was washed out with ethyl acetate. The LC orientational order was frozen indefinitely by photopolymerization. The fabrication process was monitored by illuminating a sample area with white light that was passed through a longpass filter to remove short wavelengths and prevent unwanted polymerization.

For optical manipulation, we used nearly the same system as that for the fabrication, except for a few replacements (Fig. 2). We used a 1064 nm beam from a CW Nd:YAG laser (Laser Compact , LCS-DTL-322-1100), whose power was 1100 mW at the output and 180 mW immediately after the objective. An optical trap was formed using a 100 \times oil-immersion objective with NA = 1.3 and 70% transmission at 1064 nm. The LC is transparent at both the fabricating wavelength (730 nm) and the trapping wavelength (1064 nm), so that no noticeable laser-induced heating of samples was observed. The laser beam was converted to circularly polarized light after passing through a linear polarizer and a quarter-wave plate. The handedness of circular polarization was easily controlled by adjusting the quarter-wave plate.

The rotating samples were placed between crossed polarizers and imaged onto a charge-coupled device (CCD) camera. By analyzing the periodic change in image intensity, rotational frequency was measured. They are shown as a function of beam power in Fig. 3 for circular cylinders with heights of 5 μm (closed circles) and 7 μm (open squares). It is seen that both frequencies are linearly dependent on laser power in

the present power range. The driving torque Γ [N m], which is balanced by the drag on the cylinder rotating in ethyl acetate at a rotational frequency ν [s⁻¹] and is proportional to the laser intensity, is, therefore, represented by

$$\Gamma = K\nu,$$

where K [N m s] is a constant. The linearity of rotational frequency with respect to laser power indicates that drag torque is proportional to the frequency at least in the present range of rotation.

In Fig. 4(a), the rotational frequency is shown as a function of the height of the cylinders. The data show a modulation, as expected, for a viscous torque that counteracts the optical driving torque originating from the wave plate-like behavior. A similar behavior was reported for LC droplets as a function of droplet diameter by Wood *et al.*¹²⁾. Since the rotational frequency falls with minima at both 10 and 19 μm , cycle length is estimated to be 9 μm .

In order to clarify the rubbing effect on rotational torque, we made similar measurements on photofabricated cylinders made on an unrubbed glass plate. The rotational frequencies averaged over four different samples for each height are plotted as open circles in Fig. 4(a). They show no noticeable oscillatory structure, in contrast to the cylinders made by the rubbing process. Without rubbing, the orientation of the molecules is not linked to a specific in-plane alignment direction, and the birefringence of the samples fabricated from LCs in different domains is different even for cylinders of the same height. We therefore found a broader distribution of rotational frequencies even for a fixed height, but a rather monotonic decrease in the average rotational frequency as cylinder height increases.

Birefringence is said to be the mechanism responsible for optical angular

momentum transfer from a circularly polarized beam to a nematic LC droplet¹²⁾. To confirm that birefringence is also effective on a solidified LC, we measured the intensity of light passing through the fabricated cylinders along the axes using a cross-polarizer technique. In this measurement, cylinders of various heights were fabricated on a rubbed glass plate such that they were fixed to the plate. A linearly polarized CW YAG laser beam was passed through the cylinders. The polarization axis was 45° to the rubbed direction, i.e., the orientation axis of the samples. The intensity of the transmitted light is plotted as a function of cylinder height in Fig. 4(b). As seen, a modulation is also observed in this figure, with minima at heights of 10 and 19 μm , which are coincident with the minimum rotational frequencies observed in Fig. 4(a). A minimum transmission intensity corresponds to the lack of change in the polarization plane after passing through the cylinders, which means that the phase change is an integer multiple of π . In such cases, the circularly polarized light suffers no change in polarization, thereby exerting no torque on the cylinders from the conservation of angular momentum. In contrast, at heights of 5 and 14 μm , where the transmitted intensities are maxima, the polarization direction is changed by 90° (the phase changes by an odd multiple of $\pi/2$). When a circularly polarized beam passes through such media, the handedness of the polarization is reversed. In such cases, the change in the angular momentum of light reaches its maximum, and the maximum torque is transferred to the samples, causing the maximum rotational frequency. As shown in Fig.4(b), the modulation in transmitted intensity is well preserved in the present range of measurements, indicating that the molecules in the cylinders are well oriented when the height of the cylinder fabricated over the rubbed surface is at least less than 20 μm .

Optical retardation reaches one full wavelength (in the present case, $\lambda = 1.06 \mu\text{m}$)

when the height d of the cylinder is equal to the cycle length of $9\ \mu\text{m}$. When n_x and n_y represent the refractive indices for polarizations perpendicular and parallel to the orientation direction of the samples, respectively, the following relation holds:

$$d |n_x - n_y| = \lambda .$$

From this relation, the birefringence $|n_x - n_y|$ is estimated to be 0.12 at a wavelength of 1064 nm. This value is comparable to the value of 0.1470 for birefringence at 589 nm reported for the LC phase¹⁷⁾. In general, birefringence tends to be smaller at longer wavelengths¹⁹⁾.

In summary, we fabricated cylinders from photopolymerizable LCs via two-photon absorption to investigate the rotational torque induced by circularly polarized light. Before polymerization, a LC droplet was placed on a rubbed glass plate to align the molecules in the rubbed direction. Both rotational torque and the intensity of the image observed through crossed polarizers were found to show similar oscillatory behaviors as a function of cylinder height. This was explained by the birefringence of the samples. The present results indicate that the structures fabricated from LCs have the potential to act as efficient photodriven microsized rotors.

Acknowledgments

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Figure Captions

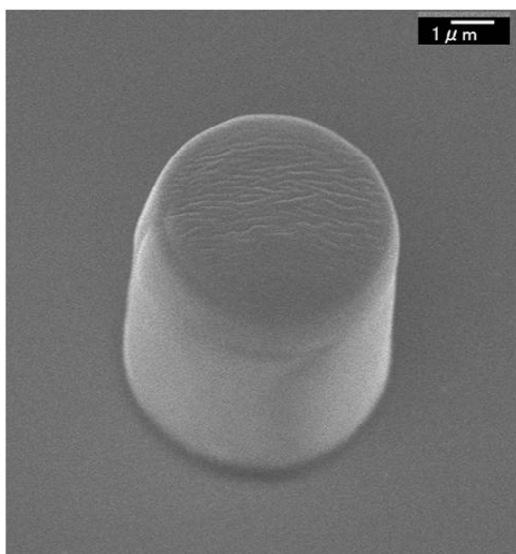
Fig. 1 Examples of microfabrication using two-photon absorption from photopolymerizable nematic liquid crystals, observed obliquely through a secondary electron microscope. (a) a 5- μm -diameter cylinder of 5 μm height, and (b) a micro-wheel gear to indicate the present fabrication accuracy.

Fig. 2 (Color online) Schematic of the optical trapping setup for rotating microcylinders. The YAG laser beam was converted into circularly polarized light through a linear polarizer and a quarter-wave plate. The cylinders were placed between crossed polarizers and imaged by a CCD to monitor their rotation.

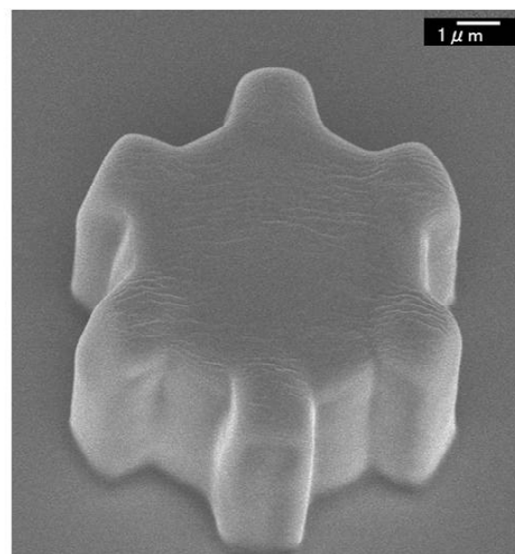
Fig.3 Rotational frequency as a function of power of circularly polarized laser. Samples made of solidified liquid crystals are cylinders of 5 μm diameter. Solid circles and open squares represent frequencies for 7- and 14- μm -high cylinders, respectively. The lines are the least-square fits of the measured points.

Fig.4 (a) Rotational frequency of microcylinders of 5 μm diameter as a function of cylinder height (in μm). The solid and dashed curves are guides for the eyes. The cylinders were fabricated from LC droplets placed on a rubbed glass plate (solid circles) and an unrudded glass plate (open circles). (b) Intensities of the laser light transmitted through the cylinders of various heights fabricated on the rubbed glass plate. They were placed between crossed polarizers for the measurements. The axis of the polarizer was 45° to the rubbing direction. The solid line is a guide for the eyes.

Fig. 1



(a)



(b)

Fig. 2

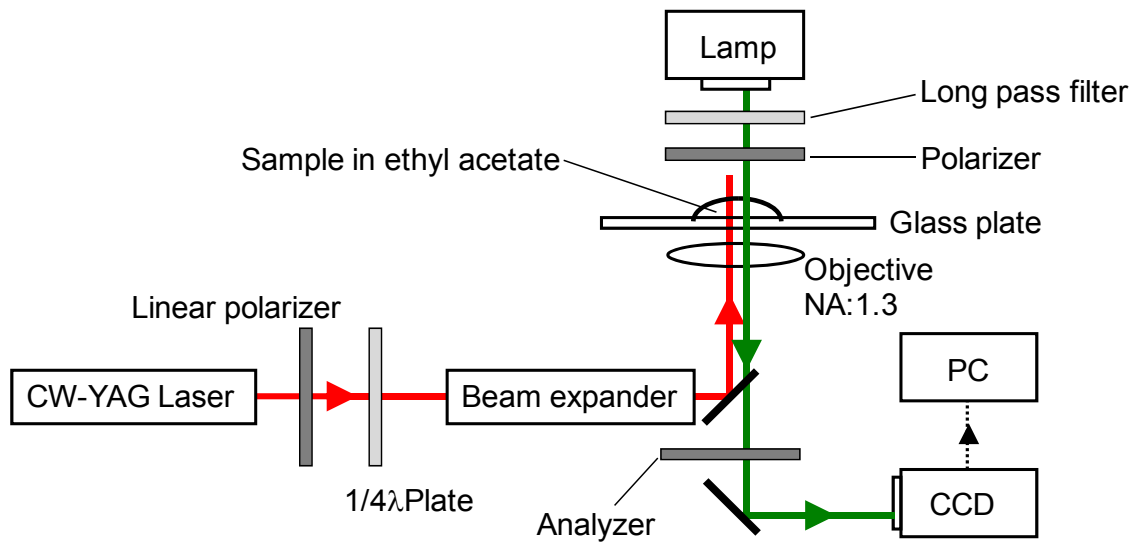


Fig. 3

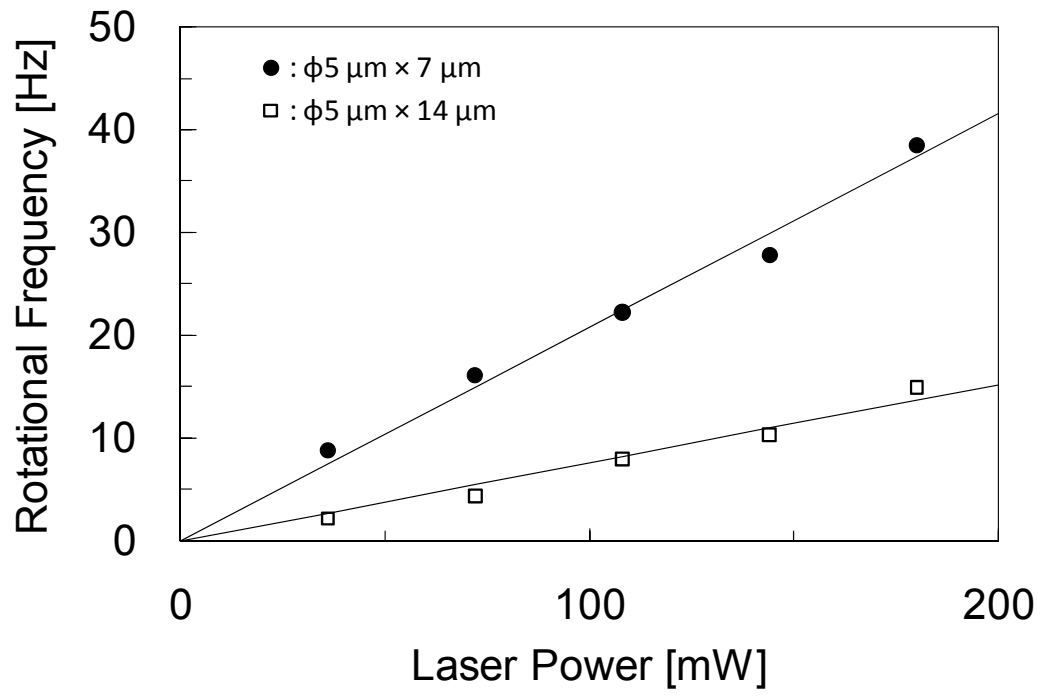


Fig. 4

