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Fabrication of a Liquid Crystal Microlens Array Film with Oligothiophene-Doped Polymerizable Liquid Crystals

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Fabrication of a microlens array film was explored by means of photoinduced reorientation of dye-doped polymerizable liquid crystals (LCs). LC monomer/photoinitiator mixtures doped with a small amount of an oligothiophene derivative were irradiated with a 488-nm beam from an Ar⁺ laser. Then the sample cell was periodically moved. After these two processes were repeated for a few tens times, remaining photoinitiators were quenched by irradiation with UV light. Observation of the cells with a polarizing optical microscope revealed that the molecular alignment of LCs induced by the Ar⁺ laser beam remained unchanged after the beam was turned off and a microlens array with a square-lattice type was obtained. This indicates that formation of the microlens and photopolymerization of the LC monomers are brought about simultaneously in a small area. The fabricated microlens has polarization selectivity that reflects molecular alignment. The focal length of the microlens arrays could be changed by controlling the irradiation intensity. Furthermore, a freestanding film with microlens arrays was obtained by peeling from the sample cell.

Keywords: liquid crystal; microlens array film; oligothiophene; photo alignment

INTRODUCTION

Microlenses whose diameter is smaller than several millimeters have attracted much interest over the past decade because of the growing miniaturization of optical components in optoelectronics and photonics devices [1]. They are used as practical applications such as an optical pickup for CD and DVD, coupling elements in optical

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interconnecters, image sensing arrays for charge-coupled devices and confocal microscopes [2], light emission arrays for vertical-cavity surface-emitting laser array and light emitting diodes [3], 3-D image displays [4] and so on.

In particular, gradient index (GRIN) lenses have attracted much attention because they have an advantage in that their surfaces are flat and other optical components can be connected on their surfaces. Ion exchange in glasses is currently the most common technique for the production of such GRIN microlens arrays, as well as electromigration [5], stumping replication [6] and diffusion polymerization [7]. Although these methods give high-quality microelements, their fabrication requires expensive equipments, multistep processes, and a long time. Additionally, they involve health hazards.

As liquid crystals (LCs) show a large birefringence which is controlled by an external field, various optical components using LC materials have been applied to practical applications [8,9]. LC microlenses and microlens arrays could be a new type of planar microlens composed of LC materials and driven by an electric field [10] and optical field [11–13]. Recently, we developed a novel fabrication method of planar microlens arrays by using photoinduced reorientation of a dye-doped LC system and successive photopolymerization with a single light source [14]. However, we could not obtain a freestanding film with microlens arrays because the main component was a low-molecular-weight LC. In this study, we investigated the photoinduced reorientation behavior of dye-doped polymerizable LCs to fabricate planar microlens array films.

EXPERIMENTAL

Sample Preparation

Chemical structures of the compounds used in this study are shown in Figure 1. 5-5"-Bis(5-butyl-2-thienylethynyl)-2,2':5',2"-terthiophene (**TR5**) has been proved to be an effective dye for photoinduced reorientation of LC [13]. By mixing **A0T5** and **A0PC3**, the mixture shows LC behavior, the alignment of which can be stabilized by polymerization after the LC alignment is controlled precisely. **TR5**, the polymerizable LCs (**A0T5**: **A0PC3** = 46 mol: 54 mol) and 0.5 mol% of the photoinitiator **Irgacure 184** were dissolved separately in chloroform and the solutions were mixed together at several dye concentrations. After the solvent was removed completely under vacuum, the LC mixture was sandwiched between two parallel transparent glass substrates with a gap of $60 \,\mu\text{m}$ by silica particles as a spacer. The inner surfaces

FIGURE 1 Chemical structures of the compounds used in this study.

of the two substrates were coated with lecithin to obtain homeotropic alignment of LCs. The monodomain structure of the LC cell was confirmed by polarizing optical microscopy (POM), and highly homeotropic alignment was observed. This cell was used as a sample for optical measurement.

Optical Setup

Preparation of the microlens was performed by the experimental setup shown in Figure 2. A linearly polarized Gaussian beam at 488 nm from an Ar^+ laser was focused normally onto the sample cell by a lens with a focal length of 20 cm to fabricate microlens. Beam waist at the focal plane was $8.2\,\mu m$ calculated from the focal length of the lens and the beam diameter. The polarization direction of the laser beam was controlled with a quarter-wave plate and a polarizer. The transmitted beam pattern created after the sample cell was observed on a screen behind the sample. When the formation of microlens was confirmed, photopolymerization was performed with 366-nm light from a high-pressure Hg lamp through glass filters (Asahi Technoglass, IRA-25S +UV-D36A +UV-35). Additionally, linearly polarized light at 633 nm from a He–Ne laser (NEC, GLG5370) was used as a probe beam. The polarization direction of the probe beam was maintained parallel to that of the Ar^+ laser beam.

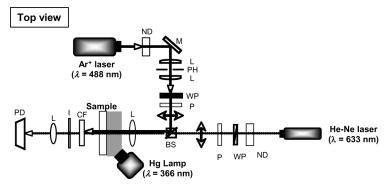


FIGURE 2 Schematic representation of the experimental setup. BS, beam splitter; CF, color filter (cut off below 505 nm); I, iris; L, lens; M, mirror; ND, neutral density filter; P, polarizer; PD, photodetector; PH, pinhole; WP, half-wave plate.

Polarizing Optical Microscopy

The gradient of the refractive index was investigated by observing the interference fringes by POM. An interference green filter (wavelength $\lambda = 560 \, \text{nm}$, Asahi Technoglass, KL55) was inserted to reduce axial chromatic aberration caused by wavelength dispersion of the light.

RESULTS AND DISCUSSION

Photoinduced Reorientation of Oligothiophene-Doped Polymerizable LCs

Upon photoirradiation, transmitted light through the sample cell formed diffraction patterns on a screen. Figure 3 shows a typical diffraction pattern observed for a **TR5**-doped cell, which indicates that reorientation of LCs occurs and microlens is formed. Alignment direction of the LCs was parallel to the polarization direction of the beam because the direction of alignment change is determined by the outer product of the electromagnetic vector of a beam and the director of LCs [12]. Depending on the light intensity, the time needed for the appearance of the diffraction rings varied for several seconds. The diffraction rings were clear and in high contrast on the screen. The number of diffraction rings (N) varied by the light intensity. N can be estimated from Eq. (1): [15]

$$N = \frac{\Delta\psi_0}{2\pi} = \frac{1}{\lambda} \int_{-d/2}^{d/2} \Delta n(z) dz \tag{1}$$

where $\Delta \psi_0$ is the phase shift observed by the laser beam in the beam center traversing the sample thickness d, $\Delta n(z)$ is the local refractive-index

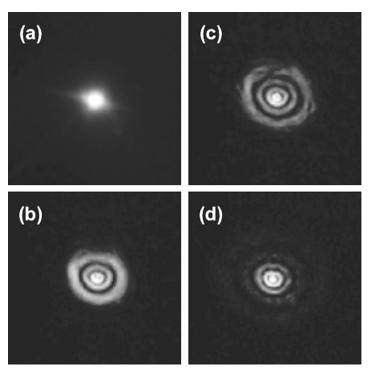


FIGURE 3 Photographs of the diffraction pattern formed on a screen at various irradiation intensities; (a) 4 W/cm^2 , (b) 9 W/cm^2 , (c) 12 W/cm^2 , (d) 15 W/cm^2 . Dye concentration, 0.2 mol%.

change induced by the laser beam, and λ is the wavelength of the laser beam. For simplicity, it is assumed that $\Delta n(z)$ is uniform along the propagation direction, and the average Δn induced in the beam center can be estimated by Eq. (1).

When the light intensity was low, the laser beam passed through the sample without obvious distortion and no rings could be observed. Above a threshold intensity, the rings appeared and the number of rings increased with the light intensity shown in Figure 4. Then, the growth of the rings ceased at a certain intensity. In the sample without polymerizable LCs, this diffraction ring vanished when the 488-nm beam was turned off due to reorientational relaxation of LC molecules. Under the present condition, however, it was found that the stable diffraction rings remained over 10 min after turning off the 488-nm beam (Fig. 5). This result suggests that alignment of LCs is stabilized by photopolymerization of LC monomers and the irradiated area functions as a GRIN lens.

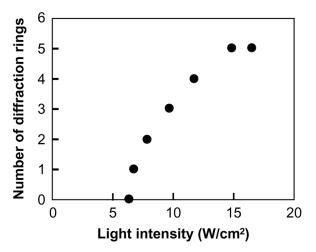


FIGURE 4 The number of observed rings formed on a screen as a function of light intensity of the pumping beam. Dye concentration, 0.2 mol%.

Focal Length of the Fabricated Microlens

The focal length is one of the most important optical properties of the lens. Therefore, the focal length of the microlens was investigated by POM. The height of the stage was set to the ink marked on the glass cell (Fig. 6a) and then the stage was moved on to the microlens, and adjusted until a sharp focal point was observed (Fig. 6b). The focal length was estimated by subtracting thickness (60 μm) from the total distance of move.

The effect of the light intensity of the 488-nm beam on the focal length of the microlens was investigated and the results are plotted in Figure 7. Below the threshold intensity, the sample cell exhibited

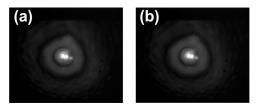


FIGURE 5 Photographs of the diffraction patterns formed on a screen upon irradiation of the sample cell with a probe beam during (a) and after exposure to the pumping beam.

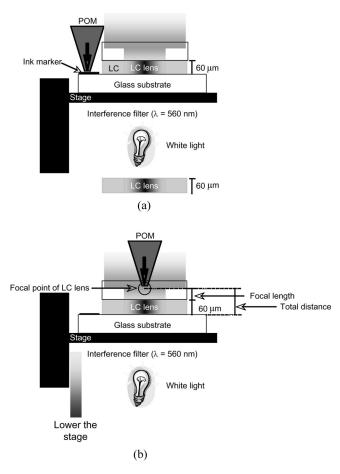


FIGURE 6 Schematic illustration of the experimental setup for measuring the focal length of the microlenses. (a) First step; (b) second step.

an initial homeotropic alignment. Thus, no focusing effect was observed. As the light intensity increased, the focal length became shorter and reached a minimum at 15 W/cm² and then increased again. Assuming that the induced refractive index in the sample is semi-spherical, the focal length of a GRIN lens is determined by three parameters and can be described by Eq. (2) [16]:

$$f = \frac{r^2}{2d\Delta n} \tag{2}$$

where r is the radius of the microlens, d is the cell gap, and Δn is difference of the refractive index between the center and the periphery of

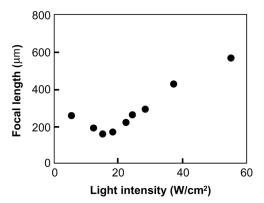


FIGURE 7 Change in focal length of the mircolens as a function of the light intensity of the Ar⁺ beam.

the microlens. This equation means that the focal length increases with an increase of the radius, while it decreases with difference of the refractive index when the thickness is constant. On the basis of this relationship, the results can be interpreted as follows. As given in Figure 8, the diameter of the microlens linearly increased as the light intensity increased. On the other hand, the photoinduced change in refractive index greatly depended on the light intensity below $15\,\mathrm{W/cm^2}$, while it showed little change above $15\,\mathrm{W/cm^2}$. Therefore, it is assumed that, at the light intensity of $<15\,\mathrm{W/cm^2}$, the focal length

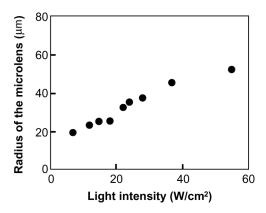


FIGURE 8 Change in radius of the mircolens as a function of the light intensity of the Ar⁺ beam.

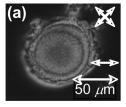
becomes shorter due to the larger contribution of Δn than that of the radius of the microlens. In contrary, the value of Δn reaches constant at the light intensity of $>15\,\mathrm{W/cm^2}$, and the contribution of the radius of the microlens dominates, leading to the increase of the focal length.

Optical Property of Microlens

POM photographs of the microlens between crossed polarizers are shown in Figure 9. The polarization direction of the 488-nm beam was at $\pm 45^{\circ}$ with respect to the polarizer (Fig. 9a). Under this circumstance, a set of concentrically circular rings was observed, which was induced by spatial distribution of the refractive index. When the sample was rotated so that its polarization direction of the 488-nm beam was parallel to the polarizer's axis, the exposed part became dark as shown in Figure 9b, indicating that the LC alignment was maintained unidirectionally homogeneous. By conoscopic observation, it was found that in the unexposed area initial homeotropic alignment was maintained. Thus, the fabricated lens was found to exhibit polarization selectivity.

Fabrication of Microlens Array Films

To fabricate an LC microlens array film with polymerizable LCs, a sample cell containing $0.1\,\mathrm{mol}\%$ of a photoinitiator was placed on an XYZ-axis stage and irradiation with a 488-nm beam at $16\,\mathrm{W/cm^2}$ for 1 min was performed. Then the sample cell was periodically moved. After these two processes were repeated a few tens of times, the whole cell was irradiated with a high-pressure Hg lamp at $5.0\,\mathrm{mW/cm^2}$ for $20\,\mathrm{min}$ to stabilize the alignment in the unirradiated area, producing a freestanding film with microlens arrays as shown in Figure 10.



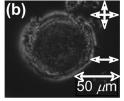
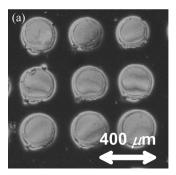


FIGURE 9 POM images of the prepared microlens. Crossed polarizers are at $\pm 45^{\circ}$ (a) and 0° (b) to the polarization direction of the Ar⁺ laser beam (dashed arrow).





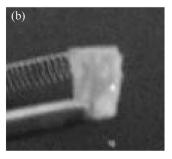


FIGURE 10 Photographs of the fabricated microlens array (a) and a peeled film with a microlens array (b). Crossed polarizers are at $\pm 45^{\circ}$ to the polarization direction of the Ar⁺ laser beam.

CONCLUSIONS

We have explored a simple fabrication of a planar microlens array film by combining photoinduced reorientation of a dye-doped polymerizable LC system and simultaneous photopolymerization. Observation by POM revealed that the molecular alignment of LCs induced by the 488-nm beam remained unchanged after the beam was turned off. The freestanding film with microlens arrays developed in this study is a promising candidate for broad applications, for example, as key components in light-emtting diodes, optical parallel-processing systems and large-scale free-space networks.

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