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Rollable ferroelectric liquid crystal devices monostabilized with molecularly aligned polymer walls and networks

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This paper describes a flexible monostabilized ferroelectric liquid crystal (FLC) device using molecularly aligned polymer walls and networks, which are sequentially formed with a two-step photopolymerization-induced phase separation. When ultraviolet (UV) light was irradiated through a photomask onto a heated nematic phase solution of FLC and monomer, monomer molecules that had flowed into the irradiated areas were photopolymerized, and polymer walls aligned along the rubbing direction of the polyimide alignment layers on plastic film substrates were formed. After uniform UV irradiation without a photomask, polymer networks aligned along the rubbing direction were formed in the FLC. The FLC molecules were monostabilized in the rubbing direction by the strong anchoring of the polymer networks. The device, which was sandwiched between crossed polarizers, exhibited a monostable electro-optic characteristic with a high contrast ratio of over 100:1 and a response time of less than 1 ms. The FLC device exhibited spatially uniform operation even when it was rolled up with a radius of curvature of 1.5 cm.

1. Introduction

Ferroelectric liquid crystal (FLC) devices using thin plastic substrates have great potential for future flexible devices that have faster electro-optic responses compared with conventional nematic liquid crystal (LC) devices [1]. Recently, an FLC device using etched spacers formed on a plastic substrate by photolithography [2] and a device using FLC polymer with high mechanical stability [3] were proposed as flexible devices. However, these devices have a disadvantage in regard to grey-scale display operation, because of the bistable switching behaviour of the FLC.

With the goal of realizing flexible FLC devices with a grey-scale capability, we previously developed an FLC device containing polymer walls [4, 5] and networks that could be formed by using a two-step ultraviolet (UV) irradiation method [6]. The polymer walls, which adhered to the surfaces of the two flexible substrates, kept the cell gap constant and maintained the FLC alignment, even when the device was bent [7, 8]. The polymer networks, whose molecules were aligned along the rubbing direction, ensured a spatial grey-scale capability because they anchored the FLC molecules [9] and changed the threshold voltage for bistable FLC switching in small areas [10].

In that device, however, the FLC alignment was disordered near the non-aligned polymer molecules of the wall surfaces, and the contrast ratio of the device was therefore limited to approximately 30:1 due to light leakage [6]. To improve the FLC alignment near the polymer walls, it is preferable to align the polymer molecules of the walls and networks in one direction. If the FLC molecules are also stabilized in the same direction, it is expected that FLC devices with polymer walls will have a continuous grey-scale capability with an excellent dark-state level, which can be seen in devices without polymer walls that use the V-shaped switching mode [11, 12] or half-V-shaped switching mode [13–15].

In this paper, we describe a high contrast flexible device consisting of an FLC monostabilized in the rubbing direction by aligned polymer networks, and walls in which polymer molecules are aligned along the rubbing direction [16]. We report the device structure, fabrication method (including formation of molecularly aligned micropolymer structures), and the monostable switching mechanism of the FLC molecules. We also discuss in detail the flexibility and electro-optic characteristics of the fabricated device.

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2. Operating principle

The structure of the device is shown in figure 1. The composite film of FLC and polymer microstructures is sandwiched between rubbed polyimide alignment layers on plastic film substrates with transparent ITO electrodes. The polymer molecules of the lattice-patterned walls are aligned along the rubbing direction. The polymer networks are also aligned along the rubbing direction to align the FLC molecules in this direction by surface anchoring of the polymer networks. The FLC molecules are therefore stabilized in the rubbing direction by the polymer networks. The device is sandwiched between crossed polarizers, and one of the optical transmitting axes of the polarizers lies in the direction of the FLC molecular alignment, namely the rubbing direction.

When no voltage is applied between the ITO electrodes, retardation of incident linearly polarized light is unaffected by the birefringence effect of the FLC areas of the device. Light transmitted from the polymer walls also does not sense the polymer birefringence. As a result, the light transmitted through the device is absorbed in the polarizer on the output side, and hence the device is expected to exhibit an excellent dark-state transmittance.

By applying a direct current voltage to the ITO electrodes, the FLC molecules rotate on the conical surface and are tilted from the optical transmitting axes of the polarizers. By applying a reverse polarity voltage, the FLC molecules rotate in the opposite direction and are also tilted from the optical transmitting axes. As a result, the light is transmitted through the output side polarizer because of the birefringence effect, resulting in bright states. In addition, the device can exhibit an analogue light-modulation function because the tilt angles of the FLC molecules depend on the electric field strength and its direction [17].

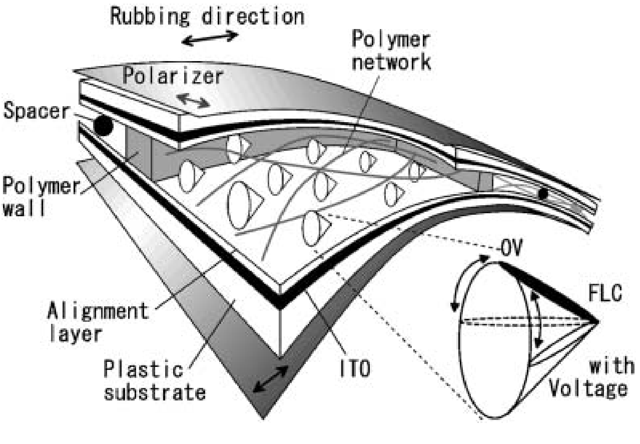


Figure 1. Schematic diagram of the flexible device using a monostable FLC layer containing polymer walls and network.

3. Fabrication method

The composite film of the FLC and polymer structures is formed by a photopolymerization-induced phase separation method using two-step UV irradiation [6]. FLC material (CS-2005, Chisso) with a 45° tilt angle is mixed with a liquid crystalline aclylate monomer (UCL-011, DIC) and 1.5 μm spacers. A solution with monomer concentration of 30 wt% has the phase sequence chiral smectic C–7°C–chiral nematic–60.3°C–isotropic. The FLC/monomer solution is coated onto a rubbed polyimide alignment layer on a 100 μm thick polycarbonate substrate (HT100-B60, Teijin) with an ITO electrode by a flexographic printing method [18].

After lamination with another plastic substrate of the same type, the solution film is heated to a temperature at which it exhibits a nematic phase, where both slender LC and monomer molecules become aligned in the rubbing direction under the narrow cell gap of 1.5 μm. The solution is then irradiated with UV light through a photomask. The monomer molecules that flow into the unmasked areas by thermal diffusion or thermal convection are photopolymerized by the UV light, as shown in figure 2. Polymer then aggregates in the irradiated areas because it cannot dissolve in the solution, due to its high molecular weight, and molecular-aligned polymer walls were formed. After

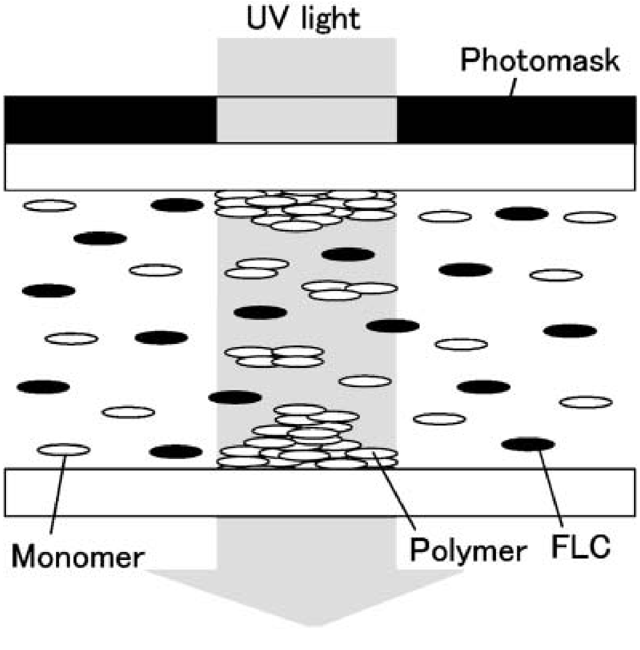


Figure 2. Method of UV irradiation using a photomask for forming polymer walls.

that, uniform UV light is irradiated onto the heated solution in the nematic phase without the use of a photomask. Unreacted monomer molecules left in the solution are photopolymerized, and hence, polymer networks aligned along the rubbing direction are formed in the FLC [19].

First, we formed the polymer walls at 40°C, where the solution with 30 wt% of the monomer exhibited the nematic phase before the walls were formed. The width and interval of the formed polymer walls were 15 µm and 250 µm, respectively. Figure 3 (*a*) shows a polarizing micrograph of the solution after the polymer walls were formed without polymer networks. Polarizing microscopy was performed for the solution in the nematic phase, because the uniformity of the LC alignment in the solution would be affected by the molecular alignment of the formed polymer. In this figure, one of the optical transmitting axes of the polarizers is in the rubbing direction of the device, and dark areas show the polymer and LC molecules aligned in the rubbing direction. Although the solution was heated to obtain the nematic phase, light was transmitted through the solution in areas near the polymer walls. When the device was rotated by about 45°, the areas that had transmitted light turned dark. This means that the LC molecules near the walls aligned in a direction other than the rubbing direction. When the monomer concentration was below 15% at 40°C, the solution exhibited the chiral smectic C phase where LC and monomer molecules were tilted from the rubbing direction. We therefore considered that while the polymer walls were growing, the monomer concentration was reduced and the solution changed to the chiral smectic C phase. This caused an obliquely aligned polymer to be formed at the edges of the walls.

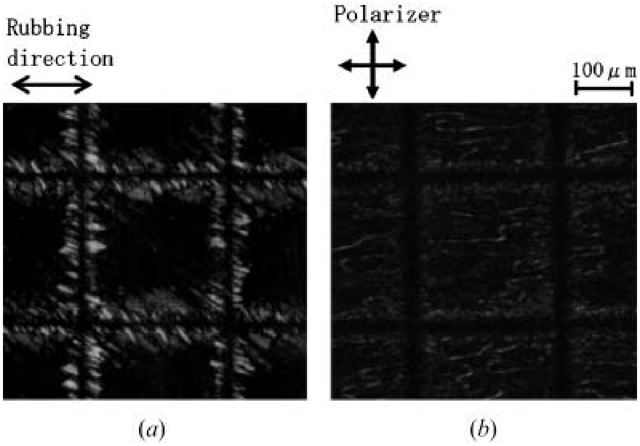


Figure 3. Polarizing micrographs of the solution heated to obtain a nematic phase after polymer wall formation (no polymer network formation) under different temperature conditions. The polymer walls were formed (*a*) at a constant temperature and (*b*) with the temperature increasing.

In this case, obliquely aligned polymer networks were formed near the polymer walls after UV irradiation without the photomask. These polymer networks stabilized the FLC molecules in a direction other than the rubbing direction, and consequently, the cone axis of the FLC molecules was tilted away from the rubbing direction, as shown in Fig. 4 (*a*). As a result, under crossed polarizers, light leakage due to the FLC molecules stabilized in this oblique direction was observed near the polymer walls as shown in Fig. 5.

In order to reduce the amount of obliquely aligned polymer material, the solution was irradiated with UV

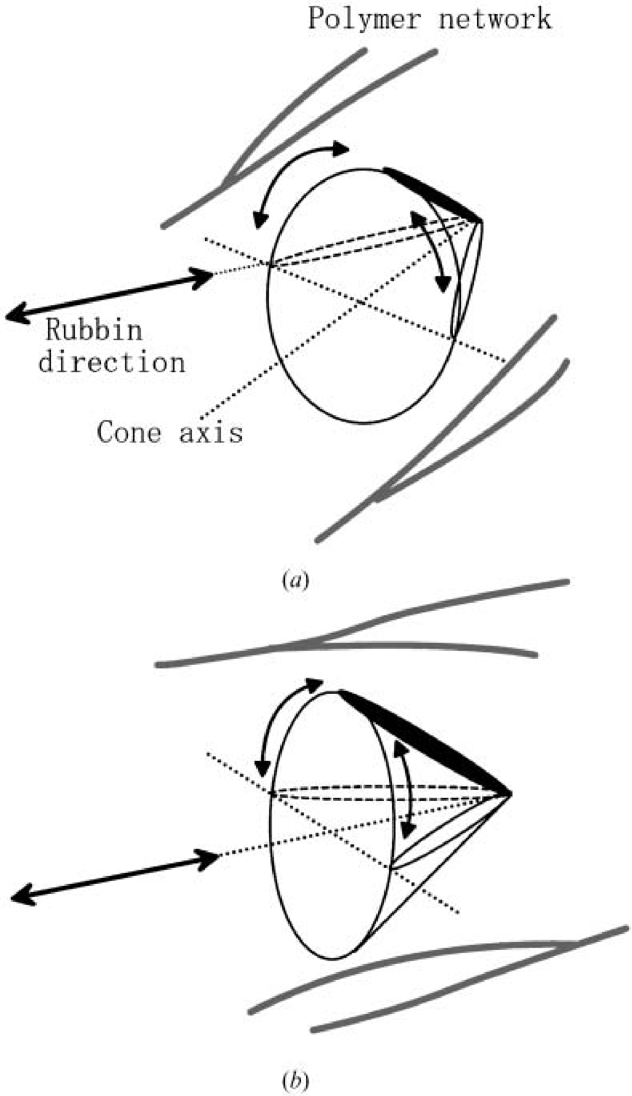


Figure 4. FLC alignment model stabilized by polymer networks. (*a*) and (*b*) show the FLC alignment tilted from the rubbing direction and along the rubbing direction, respectively.

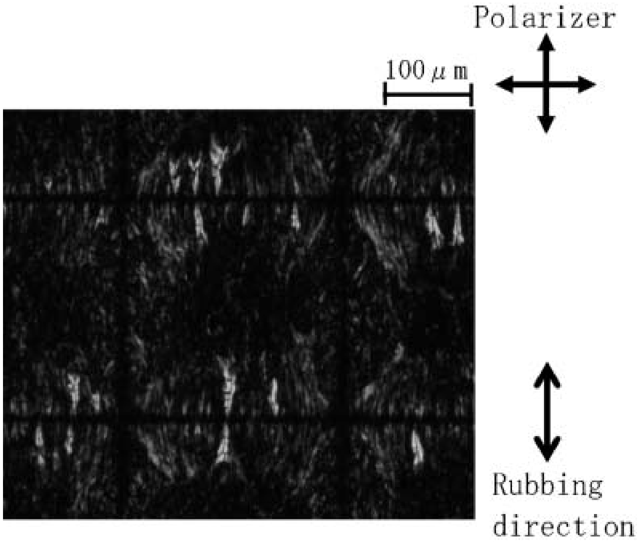


Figure 5. Polarizing micrograph of the device with disordered FLC alignment (chiral smectic C phase) near the polymer walls.

light through the photomask while the temperature of the solution was raised at a rate of 0.2°C s^{-1} to 50°C, where the molecular alignment in the solution was in the isotropic phase before UV irradiation. As a result, the molecules in the solution were uniformly aligned along the rubbing direction when the solution was heated in the nematic phase after polymer wall formation, as shown in figure 3 (*b*).

After that, the polymer networks were formed in the FLC by uniform UV irradiation. We observed the uniformity of the composite film with a polarizing microscope. Figure 6 (*a*) shows a micrograph of the FLC alignments in the dark state under crossed polarizers and a FLC switching model for the fabricated device. Figure 6 (*b*) shows those of our previous device consisting of bistable FLC and optically isotropic walls of non-aligned polymer molecules [6]. In the previous device, the polymer networks needed to be aligned in the rubbing direction and the FLC molecules switched bistably between the two directions tilted from the rubbing direction according to the electric field strength. However, the FLC molecules were disordered by the non-aligned polymer material that had formed near the walls and, hence, light leakage occurred near the polymer walls. On the other hand, both the polymer and FLC molecules in the new device could be aligned uniformly in the rubbing direction, and, consequently, light leakage was eliminated near the polymer walls. Figure 7 shows an electron micrograph of the polymer structures formed on a plastic film substrate in the monostable FLC device. It was confirmed that the polymer molecules of the walls and networks were aligned along the rubbing direction. We considered that the FLC molecules could be uniformly aligned in the rubbing direction by the uniformly aligned polymer molecules.

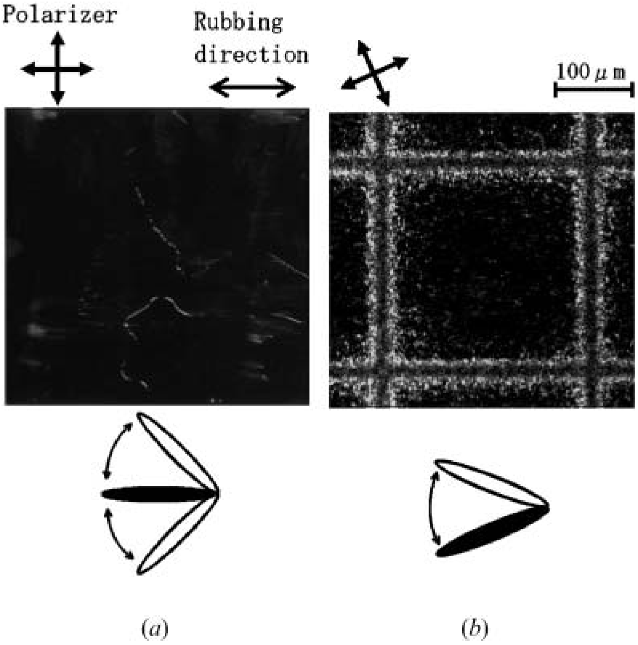


Figure 6. Polarizing micrographs of the FLC and polymer alignment and models of FLC switching. (*a*) Present device using monostabilized FLC and aligned polymer walls; (*b*) previous device using bistable FLC and walls of non-aligned polymer.

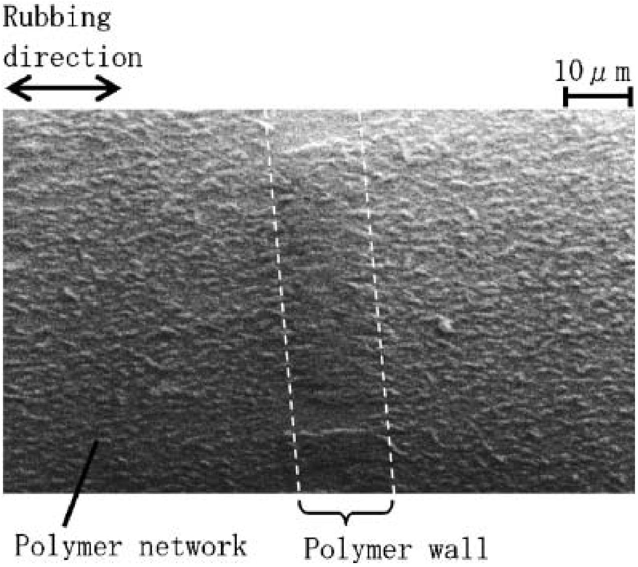


Figure 7. Electron micrograph of the polymer walls and networks formed on the plastic film substrate.

4. Electro-optic properties

Figure 8 shows the measured light transmittance through a fabricated device sandwiched between crossed polarizers under different applied voltages. Although FLC devices without polymer networks usually have bistable switchability, the fabricated device exhibited a V-shaped electro-optic characteristic based on the monostable FLC switching behaviour shown in figure 4(*b*). We thought that the FLC molecules could be monostabilized in the rubbing direction by the strong anchoring of the fine molecularly aligned polymer networks [20]. In addition, the device had a high contrast ratio of more than 100:1, because light leakage near the polymer walls was suppressed by the aligned polymer structures and monostabilized FLC. Polarizing microscopy also showed that the grey-scale capability was due to analogue alignment changes in the FLC molecules according to the electric field strength, as shown in figure 9.

We examined the FLC tilt angle while the temperature was changed from 25 to 65°C. The tilt angle was 40° at 20 V, and this was held almost constant until the temperature of the device reached 55°C (figure 10). In the case of conventional bistable FLC devices, dark-state transmittance increases when the temperature of the FLC materials increases, because the direction of the stabilized FLC alignment tilts from the direction of the optical transmitting axes of the polarizers through a reduction in the tilt angle. On the other hand, the dark-state transmittance of this monostablized FLC device does not depend on the tilt angle, because the FLC molecules can be aligned along the rubbing direction under zero applied voltage. This means that, compared with bistable FLC devices, the monostable device has a wider range of operating temperatures for which a high contrast ratio is available.

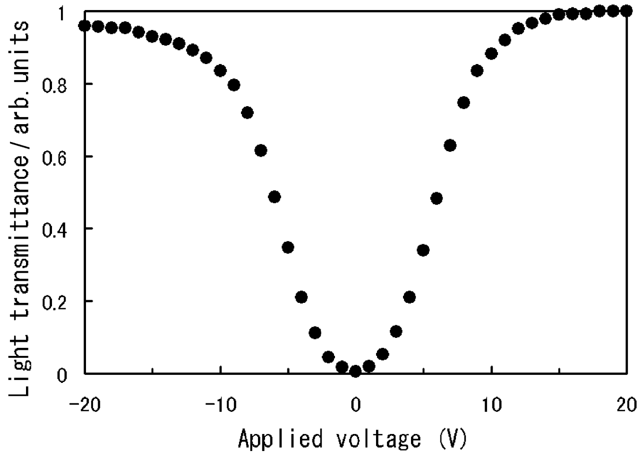


Figure 8. Dependence of light transmittance of the device on applied voltage.

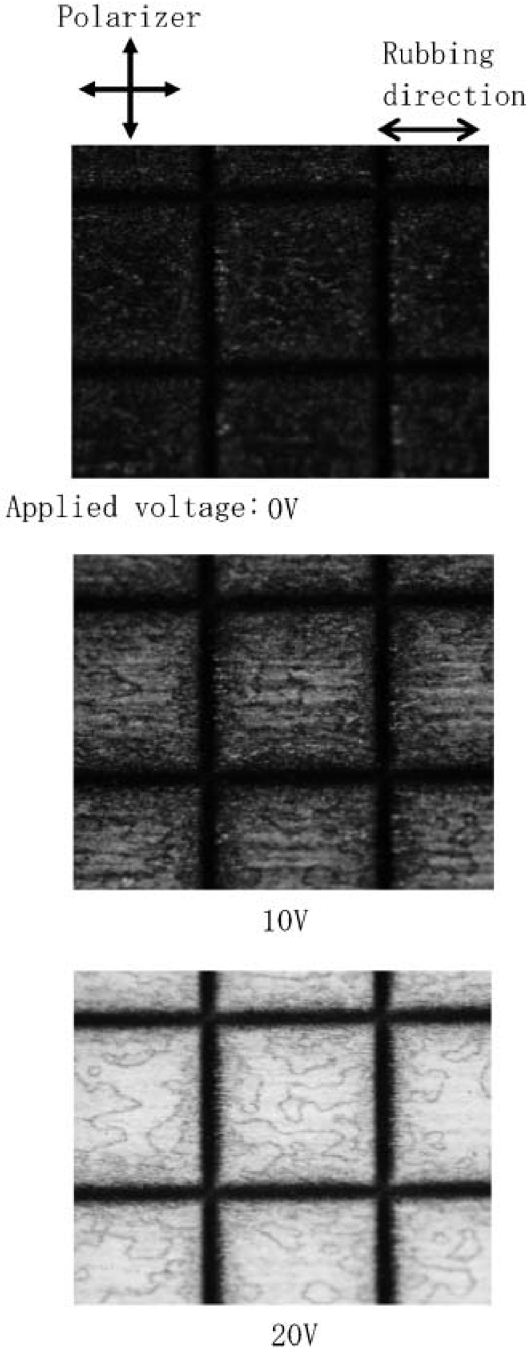


Figure 9. Polarizing micrographs of the FLC device driven by different applied voltages.

We also confirmed that the intensity of transmitted light can be controlled by using various high speed pulse voltages, as shown in figure 11. Figure 12 shows the response time of the FLC device under different applied

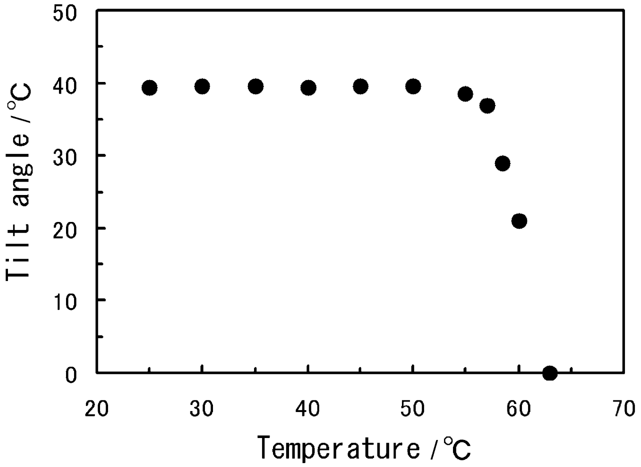


Figure 10. Temperature dependence of tilt angle of FLC switching in the device.

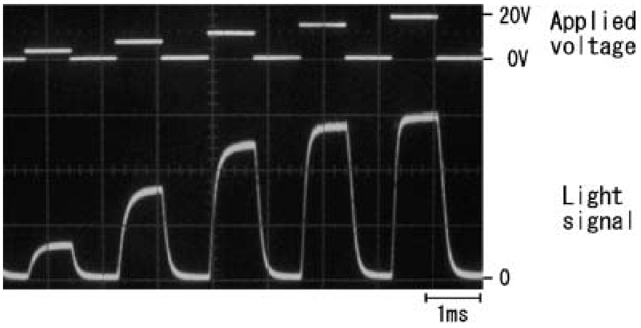


Figure 11. Electro-optic response of the FLC device to various pulsed voltages.

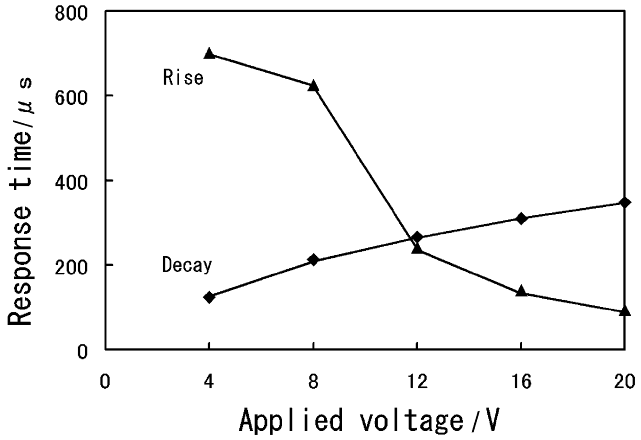
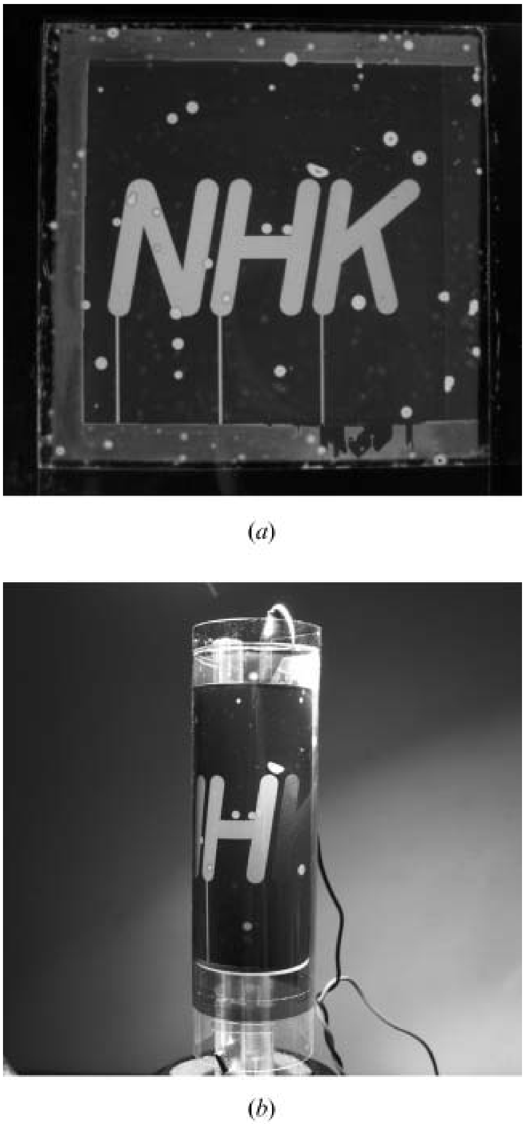


Figure 12. Dependence of response time of the FLC device on applied voltage.

voltages. The rise time and decay time show the time in which the light transmittance changed from 0% to 90% and from 100% to 10%, respectively. We found that the device exhibited a response time of less than 1 ms, including rise and decay times.


(*a*)

(*b*)

Figure 13. Appearance of the FLC device driven by applied voltages: (*a*) in the flat state, and (*b*) in the rolled state.

5. Bent-state operation

We fabricated an FLC device ($10 \times 10\,\mathrm{cm}^2$) with patterned ITO electrodes. When a device sandwiched by polarizers was driven by different applied voltages, it exhibited uniform light modulation in the flat state, as shown in figure 13 (*a*). The bending tolerance of a device without polarizers was evaluated by measuring the minimum radius of curvature that could be achieved without disordering the FLC alignment (the device was bent around solid columns of different radii) [7]. We found that the FLC alignment was maintained even after the device had been rolled up with a radius of curvature of 1 cm. We also confirmed that the bending tolerance of the device does not depend on the FLC alignment direction, although the FLC and polymer structures were aligned along the rubbing direction.

The high tolerance to bending enables the device to be formed into a curved display. Figure 13 (*b*) shows a fabricated rollable FLC device sandwiched by polarizers. A thin light diffusion film, which was located on the inner polarizer, was illuminated with white light. The device was able to demonstrate light modulation with grey-scale capability at a small radius of curvature (1.5 cm). We considered that the reason for the stable display operation in the bent state was that the polymer networks can strongly stabilize the fragile smectic layer structure.

6. Conclusions

We have developed a rollable device containing FLC monostabilized with molecularly aligned polymer walls and networks. The polymer molecules were aligned along the rubbing direction by keeping the FLC and monomer solution in the nematic phase in a two-step UV irradiation process. These polymer networks stabilized FLC molecules in the rubbing direction, and consequently the device exhibited V-shaped electro-optic properties with a high speed response of less than 1 ms. The FLC alignment was uniform near the polymer walls and, hence, the device exhibited a high contrast ratio of more than 100:1. The device also demonstrated light modulation with grey-scale capability, even when it was rolled up with a radius of 1.5 cm.

In future work, we plan to study in detail the mechanism of polymer wall formation, and enhance the uniformity of the FLC alignment by optimizing the conditions of polymer network formation to acquire a higher contrast ratio. We also wish to advance the research on active driving methods for FLC devices using plastic substrates [21].

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