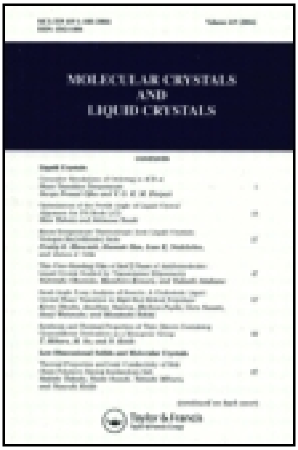

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Fabrication of Optically Designed Multidomain Layer Structure using Azo-Dye-Doped Ferroelectric Liquid Crystal Having N*-C* Phase Sequence

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We proposed an optical fabrication method of designed multidomain structure in an azo-dye-doped ferroelectric liquid crystal with chiral nematic (N*) – chiral smectic C (SmC*) phase sequence (NC-FLC). This layer alignment control is based on the N*-SmC* phase transition induced by the photoisomerization of doped azo-dye. This photo-assisted layer switching can be applied to the fabrication of the designed multidomain structure in the electrooptic elements such as display, optical memory and optical grating. Anomalous layer switching upon the electric field has also been observed.

Keywords: ferroelectric liquid crystal; N*-C* phase sequence; azobenzene; photoisomerization; layer structure; optically designed multidomain

1. INTRODUCTION

Recently, fabrication of a designed multidomain structure has attracted considerable attention from technological point of view such as improvement of a viewing angle in a liquid crystal (LC) display and fabrication of a grating. In this study, we have proposed here the photoassisted fabrication technique of the designed multidomain structure using an azo-dye-doped ferroelectric liquid crystal having chiral nematic (N*) - chiral smectic C (SmC*) phase sequence (NC-FLC) [1].

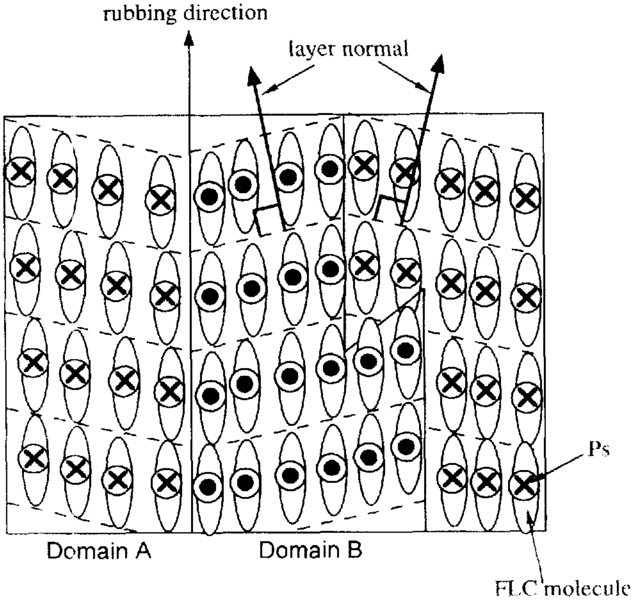


FIGURE 1　Two possibilities of layer arrangements in the SmC* phase of NC-FLC

In an NC-FLC planer cell, there are two types of layer arrangements whose layer normals make angles of $\pm\theta$ (tilt angle) with respect to the rubbing axis in the SmC* phase as shown in Fig. 1. Therefore, NC-FLC has been regarded not to be favorable for application to display devices, because of difficulty of realizing uniform monodomain and bistable switching. However, the interaction of spontaneous polarization (P_s) and an electric field determines the tilt direction of LC molecules in the layer, so that layer arrangement can be controlled by choosing the polarity of the applied electric field in the cooling process from the N* to the SmC* phase. So far, we have proposed the layer alignment control using the N*-C* direct transition which was caused by a thermal process due to laser irradiation [2].

On the other hand, photochromic molecules whose colors can be changed by light irradiation have attracted much interest. Not only colors, but also their molecular structures, dipole moments and so on can be changed optically, so that many studies have been carried out for the application to the optical memory device and so on [3]. Especially, azobenzene chromophore changes its molecular structure markedly due to the photoisomerization. In this study, in order to induce the N*-C* transition, the photoisomerization of doped azo-dye is adopted, which induces the shift of the phase transition temperature of the host LC.

2. MATERIALS

Figure 2 shows the molecular structures of liquid crystal and azo-dye used in this study. An achiral smectic-C liquid crystal, 4-hexyloxyphenyl 4-octyloxybenzoate (HOPOOB), doped with a chiral molecule (S811) of 1wt% concentration, was used for the NC-FLC. 4-diethylaminoazobenzene was doped as a photochromic azo-dye. The concentration of the azo-dye in the FLC was varied from 3wt% to 7wt%. These mixtures show an isotropic - N* - SmC* phase sequence with cooling temperature.

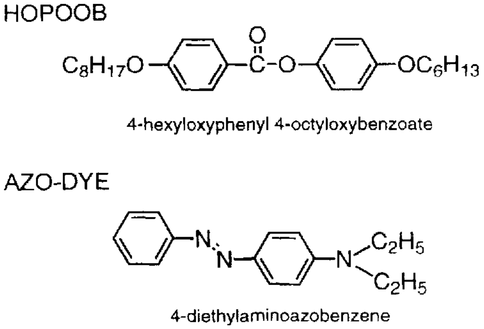
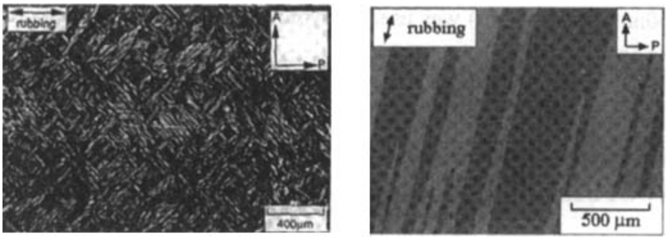


FIGURE 2 Molecular structures of achiral liquid crystal and azo-dye.

As mentioned above, in an NC-FLC planer cell, there are two types of layer arrangements whose layer normals make angles of $\pm\theta$ with respect to the rubbing axis in the SmC* phase. Generally speaking, two types of domains having different layer alignment are randomly distributed in the cell as shown in Fig.3 (a). However, some compounds including HOPOOB show stripe domains with a long axis parallel to the rubbing direction as shown in Fig. 3 (b). Using this charecteristic stripe domain, fine grating structure can be expected.



(a) (b)

FIGURE 3 Polarizing microphotograph of the NC-FLC in the SmC* phase; (a) random domain which can be seen generally, (b) charecteristic stripe domain of HOPOOB.

3. EXPERIMENTAL

The sample was filled by a capillary action into a sandwich cell, which consists of two indium-tin-oxide (ITO)-coated glass plates. The cell gap was 2 μm. In such a thin cell, no texture due to the helix was observed.

To obtain a homogeneously aligned cell, the surfaces were coated with a polyimide (AL1254, Japan Synthetic Rubber) and rubbed in the anti-parallel direction to each other. The sample was set between crossed polarizers and observed by a polarizing optical microscope. The texture was recorded using a CCD digital camera (HC-300, FUJIFILM).

The temperature of the sample was controlled using a hot bath and a temperature controller (FP21, SHIMADEN). Light illumination was performed using a 160W metal-halide lamp (IMH-160, SIGMA KOKI). In order to induce *trans-cis* photoisomerization of the azo-dye, a UV-pass filter C-40C (TOSHIBA) was installed in front of the sample, which transmitted light having a wavelength region from 280nm to 420nm. For the observation of textures, a long-pass filter O-56

(>560nm; orange light) was used to avoid *trans-cis* photoisomerization of the azo-dye.

4. FABRICATING METHODS OF OPTICALLY DESIGNED STRUCTURE

Figure 4 shows the schematic explanation of proposed photo-induced layer alignment control in the azo-dye doped NC-FLC. As mentioned above, if we cool the sample from the N* to SmC* phases in the absence of the electric field, two kinds of layer alignments can be formed in the SmC* phase as shown in Fig.1. Upon cooling down from the N* to SmC* phases under the application of a dc electric field, however, uniform unidirectional layer alignment can be realized in the entire area of the cell (i.e. Domain B in Fig.2 or Fig.4 (a)). When the polarity of the applied field is reversed, the molecules tilt away from the rubbing direction by 2θ (Fig.4 (b)). In this state, if UV light is irradiated, the N* phase is induced due to *trans-cis* photoisomerization of the azo-dye and liquid crystal molecules tend to align along the rubbing direction as shown in Fig.4 (c). Upon subsequentially stopping UV irradiation, an opposite photoisomerization process (*cis-trans*) causes the reentrance into the SmC* phase with keeping molecules parallel to the rubbing direction as shown in Fig.4 (d). Consequently, another domain (Domain A), in which the layer normal tilts to the opposite direction with respect to that in Fig.4 (a), can be obtained.

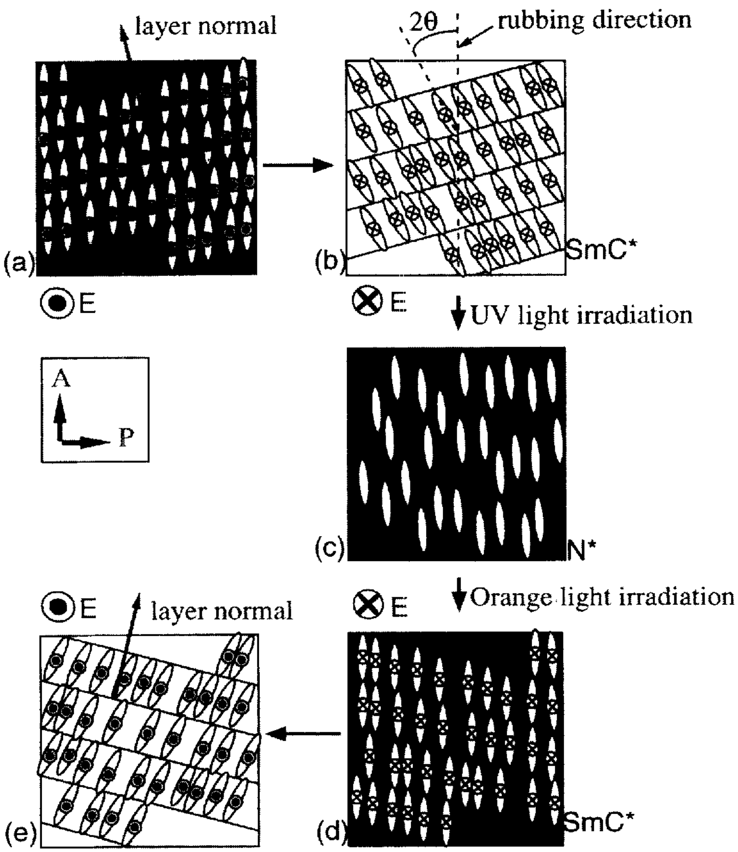


FIGURE 4 Schematic explanation of the photoinduced layer switching proposed in this study.

5. RESULTS AND DISCUSSIONS

5.1 Effect of the photoisomerization of azo-dye to the phase transition of the LC

In advance, we have investigated the effect of the photoisomerization of azo-dye to the phase transition of the FLC. The phase transition temperatures decrease with increasing the concentration of the azo-dye in the host FLC. It should be noted that the N*-C* transition temperature is lowered by the irradiation of UV light. With increasing concentration of the azo-dye, the caused temperature shift increases. Namely, it is expected that the more azo-dye is doped, the more effectively it affects the phase transition temperature of the host FLC. At high concentration of the azo-dye (more than 7wt%), however, the smectic A (SmA) phase appears between the N* and SmC* phases. The study mentioned below was carried out using a sample with a dye concentration of 5wt%.

5.2 Fabrication of optically designed multidomain layer structure

First, the sample was heated to the isotropic phase and then cooled down to the SmC* phase. To obtain uniform unidirectional layer alignment, a negative electric field (-E) was applied at the phase transition from the N* to SmC* phases. The temperature of the sample was kept just below N*-SmC* phase transition point. Polarizer (the polarization of incident light) was set perpendicular to the rubbing direction (Type I). To realize bright state, the polarity of the applied field was reversed to +E, as shown in Fig.5 (a), which corresponds to the state shown in Fig.4 (b). When UV light was irradiated to the sample by changing the long-pass filter into an UV-pass filter, the SmC*-N* phase transition was induced due to the photoisomerization of the azo-dye and the N* phase appeared (Fig.5 (b) and (c)). Upon stopping the UV irradiation, the temperature reentered into the SmC* phase, in which the smectic layer was formed again, as shown in Fig.5 (d). It should be noted that extinction is obtained in the reentered SmC* phase in spite of the constant electric field during the light irradiation. As evident from Fig.4, it means that the smectic layer switching was performed by the irradiation of UV light.

In this study, layer alignment control was succeeded at only 1-3°C below the N*-C* phase transition temperature. However, by selecting a more suitable dye for the purpose (such as one with high solubility, absorbability, and strong interaction between the liquid crystallinity and the photoisomerization), the temperature shift due to the photoisomerization of the dye should be more effectively induced. By utilizing this photo-induced layer switching, the fabrication of designed multidomain layer structure can be performed optically with masking or interfering of incident light.

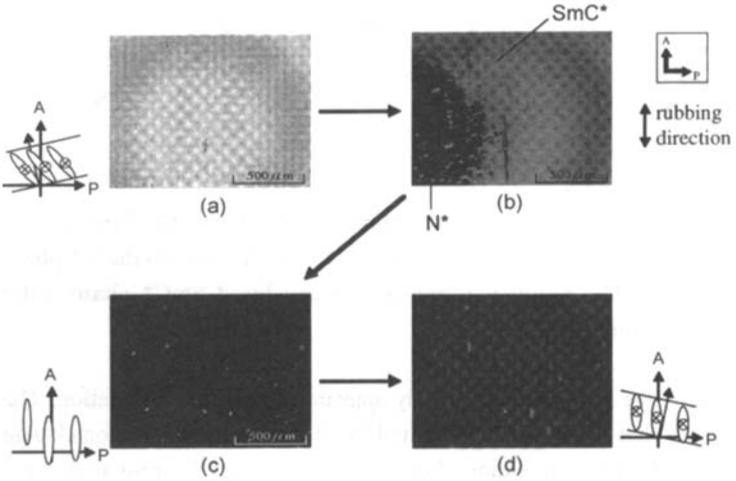


FIGURE 5 Polarizing microphotographs of the azo-dye-doped NC-FLC before and after UV irradiation. (a) the SmC* phase before UV irradiation; (b) and (c) the N* phase induced by UV irradiation; and (d) the SmC* phase after stopping UV irradiation.

5.3 Polarization effect of incident light to the induction of phase transition

In the case of Type I configuration of polarizer, the induced N* phase was stable during UV irradiation. On the other hand, under the polarizer configuration that polarization of the incident light was parallel to the rubbing direction (Type II), the N* phase was also induced (Fig.6 (b)), however, the SmC* phase was reinduced in spite of a continuous UV irradiation (Fig.6 (c)).

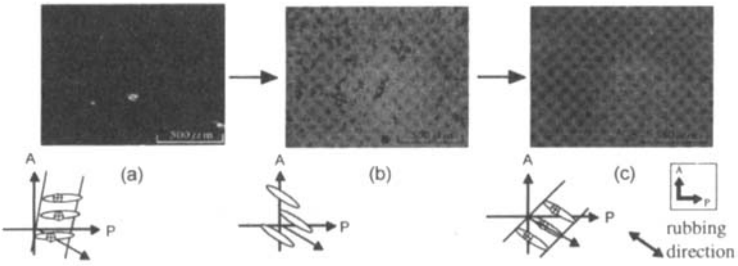
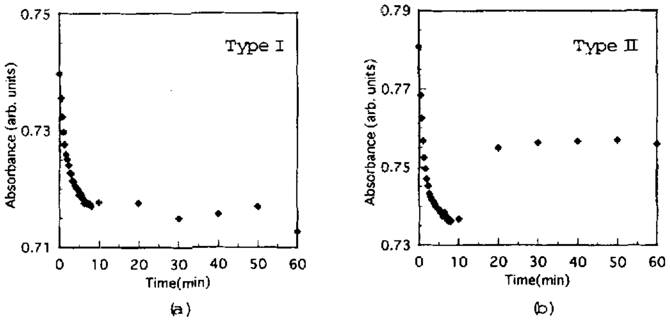


FIGURE 6 Polarizing microphotographs under the Type II UV irradiation. (a) the SmC* phase before UV irradiation; (b) the N* phase induced by UV irradiation; and (c) the reinduced SmC* phase under UV irradiation.

Namely, the N* phase temporally appeared by the UV irradiation. The detail of this effect was investigated by observing the absorption change due to the UV irradiation. Figure 7 shows the UV irradiation time dependence of absorbance of 416 nm in wavelength (corresponding to that of *trans*-form) in Type I and II geometry. In the Type I, upon the UV irradiation, the absorption of *trans*-form was suppressed by the *trans-cis* isomerization, and saturated (Fig.7 (a)). On the other hand, in Type II, although *trans* absorption band was instantaneously suppressed, the absorbance didn't saturate and recovered somehow (Fig.7 (b)). These results seem to be associated with the temporal photo-induced phase transition described above.



(a)

(b)

FIGURE 7 Time dependence of absorbance at 416 nm under UV irradiation.

As shown in Fig.6, the UV irradiation in the Type II polarization geometry cannot generate a stable *cis*-isomer of the doped azo-dye and the shift of the phase transition temperature was not induced. In other words, the SmC* phase was stabilized even upon continuously irradiating UV light. In this reinduced SmC* phase, that is in Type II geometry, it should be noted that anomalous layer switching upon the electric field has been observed. Namely, under UV irradiation, the polarity revarsal of applied electric field induces the phase transition to the N* phase, resulting in the smectic layer switching. When the polarity of the applied field is reversed in the SmC* phase under the Type II UV irradiation, the polarization reversal takes plase accompanying the molecular switching as shown in Fig.8 (b). However, just after the molecular switching, the phase transition to the N* phase is instantaneously induced and finally the SmC* phase appears again as shown in Fig.8 (d). As the result, the smectic layer switching is caused by the polarity reversal of the electric field in the same manner as the photo-induced layer switching discussed in the first half of this paper.

Although the mechanism of this novel electric field induced layer switching phenomenon is not clarified at this stage, this is a quite new phenomenon and may be expected to the novel apprication.

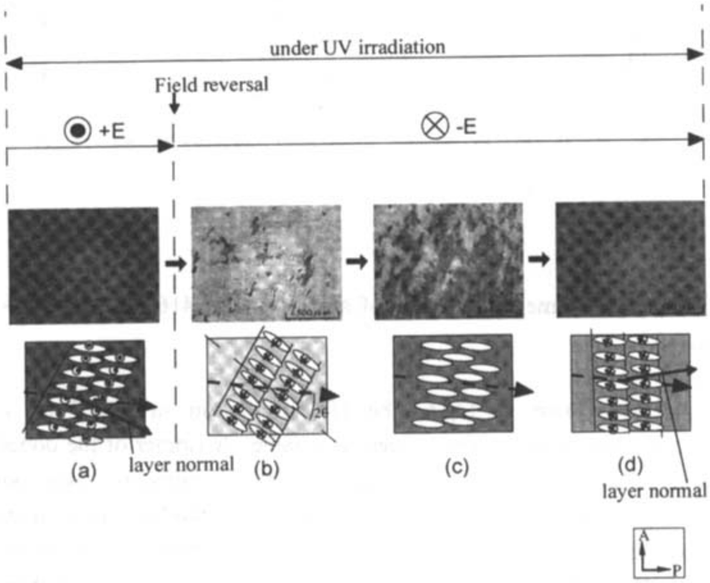


FIGURE 8 The switching of layer structure by reversal of the applied electric field under UV irradiation: before reversal (a); tilting of LC molecules by 2q just after field reversal (b); the N* phase was induced (c) and subsequently, the SmC* phase was reinduced (d).

6. CONCLUSIONS

We proposed a photo-induced layer alignment control in an azo-dye-doped ferroelectric liquid crystal with N*-SmC* phase sequence. This layer alignment control was based on the N*-SmC* phase transition induced by the photoisomerization of doped azo-dye. It is expected that by using masking and interfering method, designed multidomain structure and fine grating structure can be obtained respectively. So, this photo-assisted layer switching can be applied to the fabrication of the designed multidomain structure in the electrooptic elements such as display, optical memory and optical grating.

It was observed that phase transition behaviors were different whether incident light was parallel or perpendicular to the rubbing direction. Anomalous layer switching upon the electric field has also been observed. This novel phenomenon seems to be related with the stability of *trans*-form of azo-dye under UV irradiation.

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