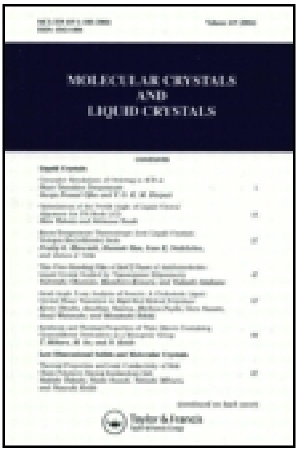

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Photoinduced Polarization-Independent Refractive Index Modulation Using Azo-Dye Doped Liquid Crystal

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We demonstrate polarization-independent refractive index modulation using an azo-dye doped liquid crystal (azo-LC). The azo-LC is aligned homeotropically and irradiated by ultraviolet (UV) light resulting in optically induced nematic-isotropic phase transition. This homeotropic-isotropic transition causes a refractive index modulation, which is independent of polarization for light travelling perpendicularly to the azo-LC layer. In order to evaluate the amount of the modulation, we fabricated an etalon which contains a homeotropically aligned azo-LC layer between the mirrors, and observed the polarization-independent refractive index modulation by 0.03.

Keywords Azo dye; nematic liquid crystal; photoisomarization; polarization independence; refractive index modulation

1. Introduction

It is well known that a refractive index modulation is possible by controlling molecular alignment of liquid crystals (LCs), and this property has been applied to many electro-optic devices. Especially, for homogeneously aligned nematic liquid crystals, a large refractive index modulation from extraordinary refractive index (n_e) to ordinary refractive index (n_o) is attained. However, this modulation is polarization dependent, and polarization control of the incident light using appropriate optical elements (for example, polarizers, wave plates and so on) is needed so that the polarization state of the incident light is parallel to the LC directors. This operation leads to losses and polarization fluctuations [1]. Therefore, the polarization-dependent refractive index modulation is not desirable for some applications.

Several ways for obtaining polarization-independent refractive index modulation have been proposed, for example, twisted nematic cell [2], nanoscale polymer-dispersed LC [3], voltage-biased polymer-stabilized cholesteric texture [4], double-layered structure with two ultra-thin anisotropic polymer films [5] and so

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on. Each method, however, has complex device structures or needs high driving voltage.

In this study, we demonstrate a polarization-independent refractive index modulation using azo dye doped nematic liquid crystals (azo-LCs). This photoinduced polarization independent refractive index modulation has not been reported and the device structure is quite simple. In order to evaluate the amount of the polarization-independent refractive index modulation and apply this principle to a practical device, we fabricate an etalon containing azo-LCs. An etalon consists of two parallel highly reflecting mirrors, and multiple interference is induced. As a result, the transmission spectrum follows the Airy function and sharp transmission peaks appear in it [6]. Since the wavelength of the transmission peak is decided by the optical path length between the two mirrors, it can be controlled by introducing LCs between the two mirrors and controlling the alignment of the LC molecules [7,8]. Based on this theory, we introduce the azo-LCs and observe UV-induced polarization-independent transmission peak shifts.

2. Principle

Azo dyes possess two molecular configurations, trans and cis form. The trans-cis isomerization is induced by UV irradiation, and cis-trans isomerization is induced by heating or irradiation of visible light. In generally, however, since cis form of the azo dye is unstable because of the approaching of two benzene rings, the cis-trans isomerization is induced automatically. In case of azo-LCs, since nematic LCs have rod like shapes, the trans form of the azo dye stabilizes the nematic phase structure in the azo-LC. On the contrary, the cis form of the azo dye is bent and destabilizes the phase structure. As a result, the LC-isotropic phase transition temperature (T_c) of azo-LC with trans form (T_{ct}) is higher than that with cis form (T_{cc}). Therefore, when the temperature of the sample is set between T_{ct} and T_{cc}, on/off control of the UV irradiation enable us to control the phase transition between nematic and isotropic phase [9].

When the azo-LC is aligned homeotropically as shown in Figure 1(a), incident light which is normal to the cell experiences n_o of the azo-LC regardless of the polarization state of the incident light. On the other hand, in case of isotropic phase induced by UV irradiation as shown in Figure 1(b), incident light experiences the

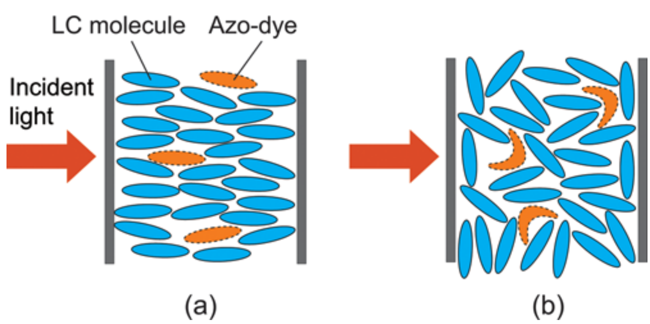

Figure 1. Princile of polarization-independent refractive index modulation. In case of (a) homeotropically alignment and (b) isotropic phase, transmission light experiences n_o and n_{iso}, respectively. Both cases are polarization independent. (Figure appears in color online.)

refractive index of the isotropic phase (n_{iso}) regardless of the polarization state of the incident light. Therefore, polarization independent refractive index modulation between n_o and n_{iso} is attained by the on/off control of the UV irradiation to the homeotropically aligned azo-LC cell.

3. Experiment

3.1. *Device Structure*

In order to confirm polarization independent refractive index modulation and evaluate the amount of the modulation, we fabricated an etalon containing azo-LC. A schematic view of the fabricated etalon is shown in Figure 2. Two dielectric multilayer mirrors, we purchased from Optical Coatings Japan, composed of alternating SiO_2 and TiO_2 layers deposited on a glass substrate was used. The number of SiO_2-TiO_2 pairs on each substrate was five and the center wavelength of the reflection band was adjusted to be 600 nm by setting the optical thickness of both SiO_2 and TiO_2 to be one-quarter of 600 nm. The reflective indices of SiO_2 and TiO_2 are 1.46 and 2.35, respectively, and the thickness of SiO_2 and TiO_2 layers are 103 and 64 nm, respectively.

The top surface of the each dielectric multilayer was coated with polyimide (JSR, JALS-2021-R2) in order to obtain a homeotropic alignment. As an azo-LC, the azo dye, diethylaminoazobenzene (TCI), was doped in the nematic LC, 5CB (4-pentyl-4′-cyanobiphenyl) (Merck), at a concentration of 3.5 wt%. The molecular structures are shown in Figure 3. This azo-LC was sandwiched between two dielectric multilayers using polyethylene terephthalate (PET) spacers with thickness of 4 μm. In order to obtain a good alignment, this device was heated above the clearing point of the azo-LC (30°C), then cooled to the nematic phase prior to measurement.

3.2. *Experimental Setup*

We measured the transmission spectra of light propagating through the fabricated etalon vertically to the surface of the etalon using a charge coupled device (CCD) multichannel spectrometer (Hamamatsu Photonics, PMA-11 Model C5966) and a halogen lamp as light source. Resolution of the CCD multichannel spectrometer is 3 nm. The change in the transmittance was recorded as a UV light ($\lambda \sim 380$ nm) from

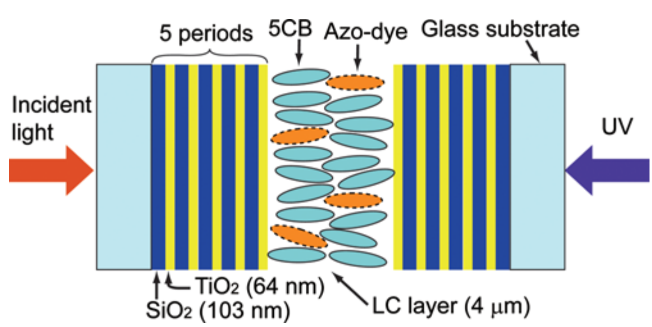

Figure 2. Schematic view of fabricated etalon containing azo-LC. (Figure appears in color online.)

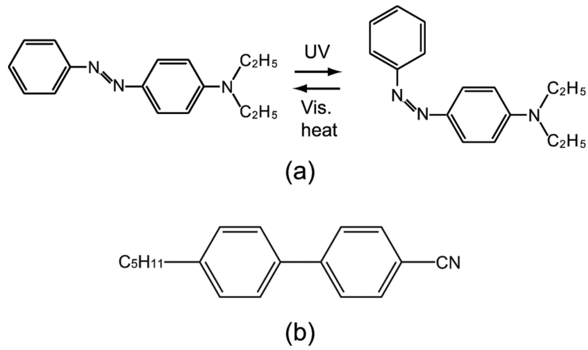

Figure 3. Molecular structure of (a) diethylaminoazobenzene and (b) 5CB.

a light emitting diode (Nichia, NCSU034A) was irradiated coaxially on the etalon at 2.24 mW/cm^2. The device temperature was controlled by hot stage (Linkam, Model 10013), and the phase transition between the nematic and isotropic phase was observed at 29.8 and 27.5°C upon UV-off and -on state, respectively. Therefore, T_{ct} and T_{cc} of the azo-LC was 29.8 and 27.5°C, respectively, and the device temperature was set at 29.2°C, which is between T_{ct} and T_{cc}.

4. Result and Discussion

Figure 4 shows the transmission spectra of the fabricated etalon before and after UV irradiation. Several transmission peaks were observed and redshifted upon UV irradiation by approximately 10 nm. The wavelength (λ) of the transmission peak is determined by a following phase matching condition [10],

$$\lambda = 2\pi \frac{n_{LC}L_{LC} + n_{BR}L_{BR}}{m\pi + n_{BR}L_{BR}k_B}, \quad (1)$$

where n_{LC} and L_{LC} are the refractive index and the thickness of the liquid crystal layer, respectively. n_{BR} and L_{BR} are the mean refractive index and the mean thickness

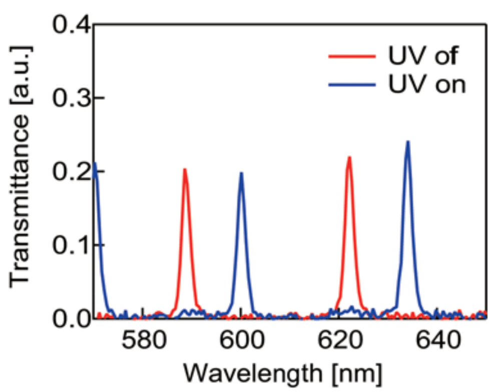

Figure 4. Transmission spectra of fabricated etalon before and after UV irradiation.

of the dielectric multilayer. k_B is the wavenumber that fulfills the Bragg condition. The integer m is the order number of the transmission peak.

Since, L_{LC}, L_{BR}, n_{BR} and k_B are constants with 4 μm, 334 nm, 1.8 and $2\pi/600\,\text{nm}^{-1}$, respectively, and the only n_{LC} is the variable number. Therefore, the red-shifted transmission peaks means the increasing of the refractive index of the liquid crystal layer. Moreover, the transmission spectra shown in Figure 4 did not change regardless of the polarization state of the incident light. This means that a polarization independent refractive index modulation was attained. And the amount of this modulation was 0.03, which was calculated based on Eq. (1).

On the other hand, extraordinary refractive index (n_e) and n_o of the azo-LC was 1.74 and 1.61, respectively, and n_{iso} was 1.65, which was calculated based on a following equation [11],

$$n_{iso}^2 = \frac{n_e^2 + 2n_o^2}{3}. \tag{2}$$

Since the refractive index is modulated from n_o to n_{iso} upon UV irradiation as we stated above, the amount of the refractive index modulation should be 0.04. This discrepancy between measured and calculated data arises mainly from low order parameter of homeotropic alignment. As a result, the refractive index experienced by light propagating through the homeotropically aligned cell was little higher than n_o.

The optical response of the transmission peak shift upon the UV irradiation is shown in Figure 5. The UV irradiation started and ended at 0 ms of Figures 5(a) and 5(b), respectively. The response time of approximate 400 and 600 ms was observed on starting and ending the UV irradiation, respectively. And only in Figure 5(a), double peaks appeared between 200 and 400 ms and transmission peak shifted discontinuously. This is because the UV irradiation induced nematic-isotropic phase transition took place in the local domains as shown in Figure 6 and nematic and isotropic phases coexist at the UV irradiation site. On the other hand, the isotropic-nematic phase transition took place homogeneously at the UV irradiated site. These differences between the nematic-isotropic and isotropic-nematic phase transitions arise from the difference of driving forces which induce the phase transition. In this experiment, UV light was used for nematic-isotropic phase transition, but nothing was used for isotropic-nematic phase transition.

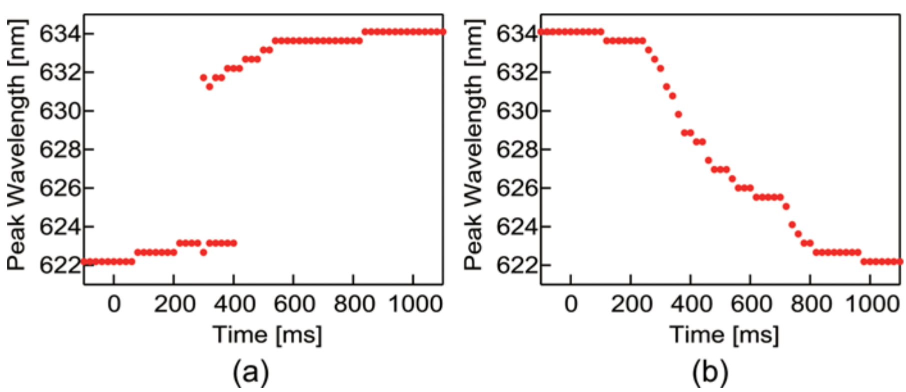

Figure 5. Optical responses of transmission peak shift upon the UV irradiation. UV irradiation (a) started and (b) ended at 0 ms of each figure. (Figure appears in color online.)

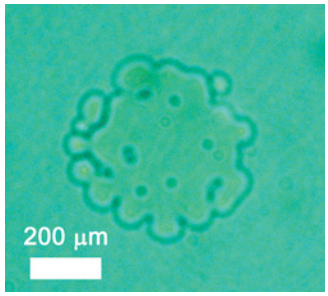

Figure 6. Optical microscope image on starting UV irradiation. (Figure appears in color online.)

5. Conclusion

In conclusion, we have demonstrated UV induced polarization-independent refractive index modulation using homeotropically aligned azo-LC. In order to evaluate the amount of polarization-independent refractive index modulation, we fabricated an etalon containing azo-LCs and observed redshifted transmission peaks induced by UV irradiation. This peak shift corresponded to the change of refractive index by 0.03. Moreover, response time was 400 and 600 ms on starting and ending the UV irradiation, respectively.

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