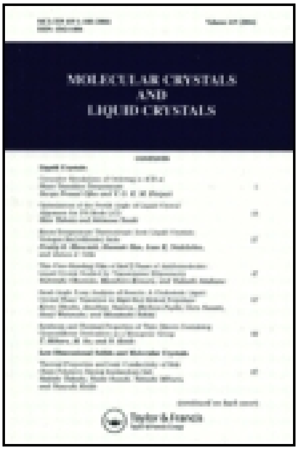

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Generation of Maxwell Displacement Current across Liquid Crystal Monolayers and Control of Liquid Crystal Alignment

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Generation of Maxwell Displacement Current across Liquid Crystal Monolayers and Control of Liquid Crystal Alignment

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The orientational transition process of p-pentyl-p′-cyano-biphenyl (5CB) liquid crystal monolayers at the air-water surface during monolayer compression and the cis-trans photoisomerization process of surface azobenzene monolayer were examined using the Maxwell displacement current measurement. Using this measurement, the orientational transition process of 5CB molecules in monolayers mixed with azobenzene molecules was examined by alternating irradiation with ultra-violet and visible light. Subsequently, we examined the orientational transition process of 5CB molecules in Liquid crystal (LC) cells triggered by the conformational change of the surface azobezene monolayer on polyimide (PI) Langmuir-Blodgett (LB) films. It was shown that the optical transmittance and the capacitance of the LC cells with 1-layer PI LB films changed reversibly by the alternating irradiation of ultra-violet (UV) and visible light, whereas those of LC cells with more than 3-layer PI LB layers did not change reversibly. These results were discussed taking into account the physico-chemical property of PI LB films.

Keywords: orientational transitional process; Maxwell displacement current; LC cells; photoisomerization

1.INTRODUCTION

According to Maxwell's electromagnetic field theory, the displacement current is the transient current, and it is generated when the electric flux

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formed between top and base electrodes changes with time. As the electric flux diverging from the constituent polar molecules in monolayer falls on electrodes even when they are suspended in air, we can detect Maxwell displacement current (MDC) across floating monolayers at the air-water interface by the application of external stimulus such as light and pressure$^{1)}$, possibly because the electric flux density changes in accordance with the orientational motion of the constituent polar molecules. In recent years, much attention has been paid to the photo-regulation of the liquid crystal (LC) alignment triggered by the conformational change of surface azobenzene monolayers, as a new way for an application of LC displays$^{2)}$, where it is essential to gain information on the orientational transition process of LC molecules triggered by the surface monolayers for a profound understanding of the operation of the LC cells. Further it is fundamentally interesting to the fields of physics, chemistry and electronics to clarify the physico-chemical property of surface monolayers.$^{3,4)}$ In conventional LC cells, rubbed polymer films such as polyimide (PI) films are usually used as an alignment layer.$^{4,5)}$ However, the use of non-rubbing process is also expected in order to overcome the electrification problems, etc.$^{4,6,7)}$ For this purpose, the use of Langmuir-Blodgett (LB) film has been proposed.$^{4,7)}$ Fortunately, for the past several years, we have succeeded in preparing PI LB films,$^{8)}$ with a monolayer thickness of 0.4 nm, and examined the physico-chemical property of the PI LB films. We found that PI LB films provide flat surface, and work as a good electrical insulator, although they are charged as-deposited due to the displacement of excessive electronic charges from metals.$^{9,10)}$ Further we have developed a MDC technique for the detection of the polar orientational transition of polar molecules in monolayers. Therefore, in the present paper, we examined the orientational transition process of p-pentyl-p'- cyano-biphenyl (5CB) liquid crystal monolayers at the air-water surface during monolayer compression and the cis-trans photoisomerization process of surface azobenzene monolayer by means of the MDC measurement. Subsequently, using this measurement, the orientational transition process of 5CB molecules in monolayers mixed with azobenzene molecules was examined by alternating irradiation with ultra-violet and visible light. Finally, we discussed the orientational transition process of 5CB molecules in Liquid crystal (LC) cells triggered by the conformational change of the surface azobezene monolayer on polyimide (PI) Langmuir-Blodgett (LB) films.

2.MAXWELL DISPLACEMENT CURRENT ACROSS MONOLAYERS

Molecules used in this study were Nematic p-pentyl-p'-cyanobiphenyl (5CB) liquid crystals and poly(vinyl alcohol)s bearing azobenzene side-chains (6Az5PVA) and polyimide (PI). The chemical structure of these molecules are shown in Fig.1. 5CB and 6Az5PVA were used for the MDC measurement. Monolayers of 5CB and 6Az5PVA were spread onto the water surface in a manner to that described in our previous paper,1) and then measured the MDC during monolayer compression or alternating photoirradiation with ultraviolet and visible light. Figure 2 shows the experimental setup used in the experiment. The trough (Nippon Laser Electronics) has a rectangular shape, where the molecule-covering area (1,050 cm^2) is changed with help of two movable barriers. Electrode 1 is a transparent SnO_2-coated glass slide with an electrode area of 45.6 cm^2. At the center of the trough, a Wilhelmy plate is set to measure surface pressure. Electrodes 1 and 2 are connected to each other with an gold wire through a high sensitivity electrometer (Keithley 617). As shown in Fig.2, charge Q is induced on Electrode 1, and it is expressed as11)

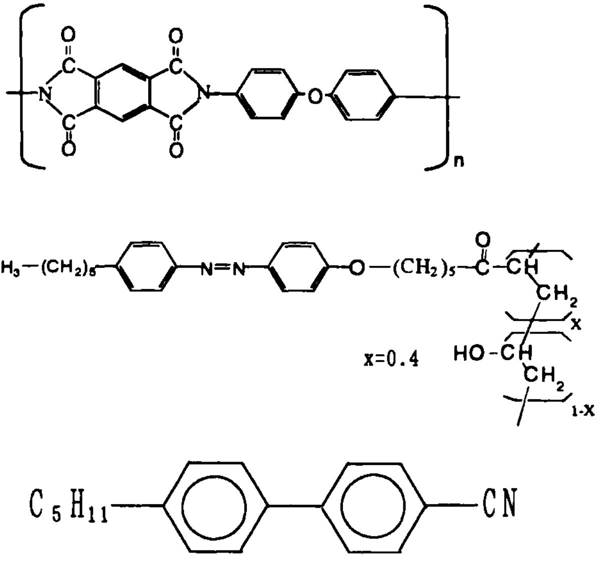

FIGURE 1 Molecules used in this study. PI, 6Az5PVA, 5CB (From top to bottom)

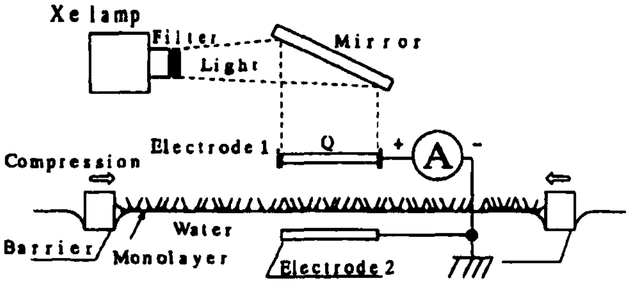

FIGURE 2 Experimental arrangement for MDC measurement

$$Q = -\frac{\mu S}{d}N - \frac{\varepsilon\varepsilon_0 B}{d}\phi_s. \tag{1}$$

Here μ is the dipole moment of the constituent molecules, d is the distance between Electrode 1 and the water surface, N is the number of molecules on the water surface under Electrode 1, ε is the relative dielectric constant of the monolayer, ε_0 is the dielectric permittivity of a vacuum, B is the working area of Electrode 1, and ϕs is the surface potential of water. S is the orientational order parameter defined as S = $\langle\cos\theta_1\rangle$, where θ_1 is the tilt angle of polar molecules away from the normal direction to the water surface and < > represents the thermodynamics average. MDC is generated due to the change of Q with respect to time and it is given by

$$I = -\frac{dQ}{dt}. \tag{2}$$

Thus it is clear from eq.(1) that the MDC is generated due to the changes in the number of constituent molecules N and the orientational order parameter S by either monolayer compression and photoirradiation. In the experiment, 5CB molecules were spread from a 1 mM chroloform solution onto the water surface in the Langmuir-trough using a microsyringe and then compressed from both sides of the LB trough at a temperature of 20 °C, by moving two barriers in opposite directions at a velocity of 0.044 $Å^2$/s. Similarly, After spreading 6Az5PVA monolayers from a 1 mM chroloform solution onto the water surface, we measured the MDC due to the cis-trans photoisomerization of surface azobenzene monolayer by the alternating photoirradiation with UV (360 nm in wavelength) and visible light (450 nm in wavelength). Here a 500 W Xenon lamp (Ushio, UI-501C) and appropriate bandpass filters (Mells griot, 03F1U012 and 03F1V028) were used and the intensities of transmitted UV and visible light were 0.2 mW/cm ^{2} and 2.0 mW/cm ^{2}, respectively. It should be noted that the contribution of the change of the surface potential of water can be assumed very small for monolayers on a pure water surface.11) Similar experiments were performed for the mixture of 5CB and 6Az5PVA (1:1 in molar ratio) on a water surface by alternating photoirradiation, after preparing a stable mixed monolayer.

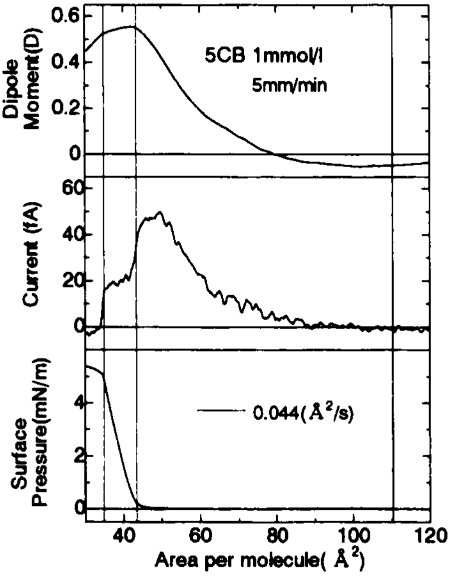

FIGURE 3 MDC across 5CB monolayer by compression.

Figure 3 shows a typical example of MDCs for 5CB monolayers during compression, where MDCs were initiated to flow at the molecular area $A=A_c$ around 110 A^2, due to the phase transition from the isotropic planar alignment phase on the water surface (range 1) to the polar orientational isotropic phase (range 2). One MDC peak appears in the range before the initial rise of surface pressure at 44 A^2.12) We plotted the vertical component m_z($= \mu$ S) of the dipole moment of molecules in monolayers moment of molecules in monolayers calculated from the charge flowing through the circuit, assuming $m_z=0$ at the critical molecular area. As we can see in the figure, the MDC current is generated due to the orientational change in the constituent 5CB molecules.

Figure 4 shows a typical example of MDC current across 6Az5PVA monolayers by alternating photoirradiation, where the molecular area was

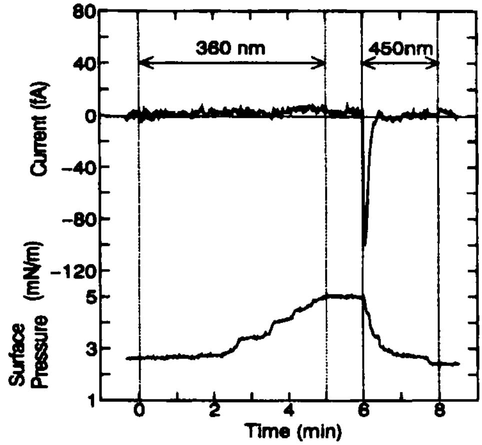

FIGURE 4 MDC across 6Az5PVA monolayer by alternating ultraviolet and visible light irradiation

43 A^2.The displacement current was generated in the positive direction with the UV irradiation, whereas it was generated in the negative direction by the visible light irradiation. The displacement current was found to be generated reversibly in further photoirradiation cycles. The surface pressure was also monitored during the photoirradiation, and found to increase and decrease reversibly. From these results, it is found that MDC is again generated by the molecular motion of azobenzene monolayers due to the cis-trans photoisomerization.

Similar experiments were carried out for monolayers mixed with 6Az5PVA and 5CB at the average molecule area of 100 $\mathrm{Å}^2$ by alternating UV and visible light photoirradiation.

the generation of MDC was not seen for monolayers with trans-form 6Az5PVA, owing to the quenching of the trans-to-cis photoisomerization in the presence of 5CB, which arises from the strong dipole-dipole interaction between the azobenzene side chains and 5CB. In contrast, the generation of MDC was seen for monolayers with cis-form 6Az5PVA, because the the distance between azobenzene and 5CB is sufficient. From this experiment, it is concluded that the transmission of the external stimulation in azobenzene mixed monolayers can be checked by the MDC measurement on a water surface before we fabricate LC cells with surface azobenzene monolayer.13)

3.PHOTOREGULATION OF LCs WITH SURFACE AZOBENZENE MONOLAYER ON POLYIMIDE LANGMUIR-BLODGETT

3.1 SAMPLES AND MEASUREMENT

PI LB films were deposited onto indium-tin-oxide (ITO)-coated glass slides by means of the vertical dipping method in the LB technique at a surface pressure of 35 mN/m, in a manner similar to that in our previous study.9) The number of deposited PI layers was 0, 1, 5 and 11. After the deposition of PI LB films, 1-layer azobenzene 6Az5PVA was deposited at a surface pressure of 30 mN/m from a pure water surface. Both top and bottom ITO-coated glass slides are covered with PI LB films and 1-layer 6Az5PVA monolayer. The top and bottom glass slides were attached face to face with the dipping direction of the two glass slides being anti-parallel to each other at a spacing of 14μm, using a supporting adhesive epoxy resin incorporating beads with a diameter of 14μm. The working area of LC cells was around 1.50 cm^2. The 5CBs have a positive dielectric anisotropy $\Delta\varepsilon (= \varepsilon_{//} - \varepsilon_{\perp})$ of 10.5 and a birefringence Δn (= $n_{//} - n_{\perp}$) of 0.182, where $\varepsilon_{//}$ (= 16.3) and $\varepsilon_{\perp}$(= 5.8) are the relative dielectric permittivity parallel and perpendicular to the long molecular axis, and $n_{//}$ (=1.709) and $n_{\perp}$ (=1.527) are the reflective indices parallel and perpendicular to the long molecular axis, respectively. The pre-tilted angle of 5CBs on 3-, 5-, 11-layer PI LB films was determined as 0.46, 031, and 0.93 degree by an optical method. For the sake of simplicity, the pretilted angle of 5CBs on PI LB films was assumed nearly to be zero in in the following discussion. The capacitance and transmittance of LC cells filled with nematic 5CBs were measured simultaneously during the alternating photo-irradiation by UV and visible lights.[14] Briefly, the UV light has a wavelength of 360 nm and an intensity of 0.4 mW/cm^2, and the visible light has a wavelength of 450 nm and an intensity of 0.8 mW/cm^2. LC cells were connected to an LCZ meter (type-2330A, NF), and the capacitance of the cells was measured at a frequency of 1 kHz. The intensity of transmitted He-Ne laser (632.8 nm in wavelength) beam through the LC cells which were placed between two crossed polarizers was measured using a Si-photo-diode during the alternating photo-irradiation of UV and visible lights. The angle between a dipping direction and a polarized light through the polarizer was set to 45 degrees.

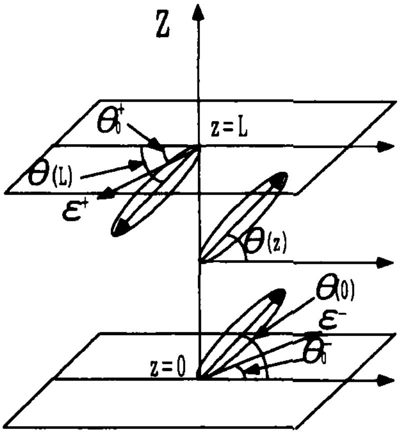

FIGURE 5 Directors in NLC cell.

3.2 ANALYSIS

For analyzing the orientational transition process of 5CB nematic liquid crystal (NLC) cells triggered by the conformational change of the surface azobezene monolayer, we here consider a nematic cell located between two plane electrodes at $z = 0$ and $z = L$, as illustrated in Fig.5. The easy axis directions at the top and bottom substrate surfaces are denoted by unit vectors ϵ^+ and ϵ^-, respectively. According to the Oseen and Frank elastic continuum theory of NLC cells, the total free energy density of the bulk is expressed as[15,16]

$$g_b = \frac{1}{2}[k_{11}(\nabla\cdot\vec{n})^2 + k_{22}\left(\vec{n}\cdot\nabla\times\vec{n} + \frac{2\pi}{p}\right)^2 + k_{33}[\vec{n}\times(\nabla\times\vec{n})]^2] + g_f(\vec{n})\cdot \tag{3}$$

Here k_{11}, k_{22} and k_{33} are the splay, twist and bend elastic constants of the LC, respectively. For 5CBs, $k_{11} = 6.9\times10^{-12}$ N, $k_{22} = 5.73\times10^{-12}$ N, and $k_{33} = 10.19\times10^{-12}$ N. p denotes the pitch of the material. $\vec{n}$ denotes the director orientation in the bulk. $g_f(\vec{n})$ represents the interaction energy between the director and the external field such as an electric field. In the present study, we discuss the case of LC medium not being subjected to the external field and only surface azobenzene monolayer experiences the cis-trans conformational change by alternating photoirradiation. Thus, $g_f(\vec{n})$ is assumed zero in the present analysis. The total free energy F is the sum of the bulk energy and the surface free energy, thus it is given by 15)

$$F = \int g_b dv + \int g_s^+ ds^+ + \int g_s^- ds^-, \tag{4}$$

where dv is the volume element of the bulk and ds is the surface area element. g_s^+ and g_s^- represent the surface energy densities, and they are given by

$$g_s^+ = -\frac{A^+}{2}(\vec{n}\cdot\vec{e}^{\,+}) \quad (z = L) \quad \text{and} \quad g_s^- = -\frac{A^-}{2}(\vec{n}\cdot\vec{e}^{\,-}) \quad (z = 0) \tag{5}$$

in the generalized form, using a simple phenomenological expression for the interfacial energy per unit area for a one-dimensional deformation by Rapini and Papoular (RP).17) Here, A^+ and A^- are the anchoring strength parameter expressing the deviation ability of the director from the easy axis directions at the interface z = L and z = 0, respectively. Minimizing the total free energy yields the stable director distribution. The equilibrium condition is then determined by the variation equation $\delta F = 0$. In our samples, top and bottom electrodes are placed anti-parallel to each other. Thus, for the sake of simplicity, we assume that the second term of Eq. (3) is nearly zero. That is, there is no twist deformation in NLC cells ($\partial\phi/\partial z = 0$ and $p \to \infty$). Under this assumption, it is possible to derive the one-dimensional torque balance equation describing the director orientation distribution in the form

$$(k_{11}\cos^2\theta + k_{33}\sin^2\theta)\frac{d^2}{dz^2}\theta - \frac{k_{11}-k_{33}}{2}\sin 2\theta\left(\frac{d}{dz}\theta\right)^2 = 0 \quad 0< z <L, \tag{6}$$

with the boundary conditions of

$$(k_{11}\cos^2\theta^- + k_{33}\sin^2\theta^-)\frac{d}{dz}\theta\bigg|_{z=0} = \frac{A^-}{2}\sin 2(\theta^- - \theta_0^-) \qquad z = 0 \tag{7a}$$

and

$$-(k_{11}\cos^2\theta^+ + k_{33}\sin^2\theta^+)\frac{d}{dz}\theta\bigg|_{z=L} = \frac{A^+}{2}\sin 2(\theta^+ - \theta_0^+) \qquad z = L. \tag{7b}$$

Here θ_0^- and θ_0^+ represent the angle between the substrate surface and the easy axis direction. During the alternating photoirradiation with UV and visible lights, the surface azobenzene monolayer experiences the cis-trans photoisomerization, and then the easy axis direction changes. Therefore by photoirradiation the NLCs experience the deformation so as to satisfy the above eqs. (6) and (7). The capacitance of LC cells depends on the director orientation distribution and it is given by

$$C = C_0 \frac{1}{\dfrac{1}{L}\displaystyle\int_0^L \frac{dz}{\varepsilon_{//}\sin^2\theta(z) + \varepsilon_{\perp}\cos^2\theta(z)}}, \tag{8}$$

where $C_0 = \varepsilon_0 \dfrac{S}{L}$. Similarly the intensity of transmitted He-Ne laser beam through the LC cells depends on the director

orientation distribution, and it is given by

$$I = I_0 \sin^2\left(\frac{\pi}{\lambda} L\Delta\right) \tag{9}$$

with $\Delta = \dfrac{1}{L}\displaystyle\int_0^L \left\{\frac{n_{//}n_{\perp}}{\sqrt{n_{//}^2\sin^2\theta(z) + n_{\perp}^2\cos^2\theta(z)}} - n_{\perp}\right\}dz.$

Here, λ represents the wavelength of He-Ne-laser (632.8 nm) and I_0 represents the intensity of the input laser beam. 5CB molecules on the surfaces of both top and bottom electrodes change their orientation in accordance with the change of the easy axis direction due to the photoisomerization of azobenzene surface monolayer during the photoirradiation. Under this situation, we can calculate the capacitance and the intensity of transmitted light of LC cells, by changing the easy axis and the intensity of transmitted light of LC cells, by changing the easy axis direction of LC cells in the range between 0 to 90 degrees, which just corresponds to the case of the surface azobenzene monolayer experiencing the cis-trans photoisomerization as aforementioned. In our experiment, we prepared LC cells with top and bottom electrodes to be anti-parallel, thus the relationship $\theta_o^+ = -\theta_0^-$ is satisfied. From Eq.(7), we obtain

$$E(\theta(z), iK) - E(\theta(0), iK) = \sqrt{1 - K^2 \sin^2 \theta(0)} \left(\frac{d}{dz} \theta(z) \right)_{z=0} \cdot z \qquad (i = \sqrt{-1}) \quad (10)$$

with $E(\theta(z), iK) = \int_0^{\theta(z)} \sqrt{1 - K^2 \sin^2 \theta(z)} d\theta(z)$ .Here $iK = \sqrt{1 - k_{33} / k_{11}}$.

Solving Eq. (10) numerically under the boundary conditions given by Eq.(7), we obtain $\theta(z)$ $(0 \leq \theta(z) \leq L)$. Then substituting the result into Eqs. (8) and (9), the capacitance C/C_0 and transmittance I/I_0 can be obtained as a function of θ_0^- $(0 \leq \theta_0^- \leq 90^\circ)$, assuming $L=14\mu$m. For the sake of simplicity, if we assume $\theta^-(0) = -\theta^+(L) = \theta_0^- = -\theta_0^+$, the rigorous solution $\theta(z) = \theta_0^-$ is obtained. In other words, the angle θ_0^- just corresponds to the tilt angle of NLC medium. It is found from simple calculation that 4 peaks appear in the transmittance, and the capacitance gradually increases as the angle θ_0^- increases and it reaches 1.0 at $\theta_0^- = 90^\circ$. Obviously, the valleys and hills appear in the transmittance at the angles where the following relationships are satisfied:

At hills $\frac{\pi L}{\lambda} \Delta = (m + \frac{1}{2})\pi$ (m: integer) (11)

and at valleys $\frac{\pi L}{\lambda} \Delta = m\pi$ (m: integer).

3.3 Motion of 5LCs on PI LB films

Figure 6 shows a typical example of the transmittance and the capacitance of LC cells by alternating UV and visible light irradiation. Figure 6 (a) and (b) shows the results for the LC cells with the number of PI LB films of 1 and 5. During the UV irradiation, 3 peaks appear in the transmittance and the capacitance gradually decreases.

For the LC cells without PI LB film, the transmittance change was not so clear, whereas it wass quite clear for the LC cells with PI LB layers as shown in Fig. 6(a) and (b). This result indicates that 5CB

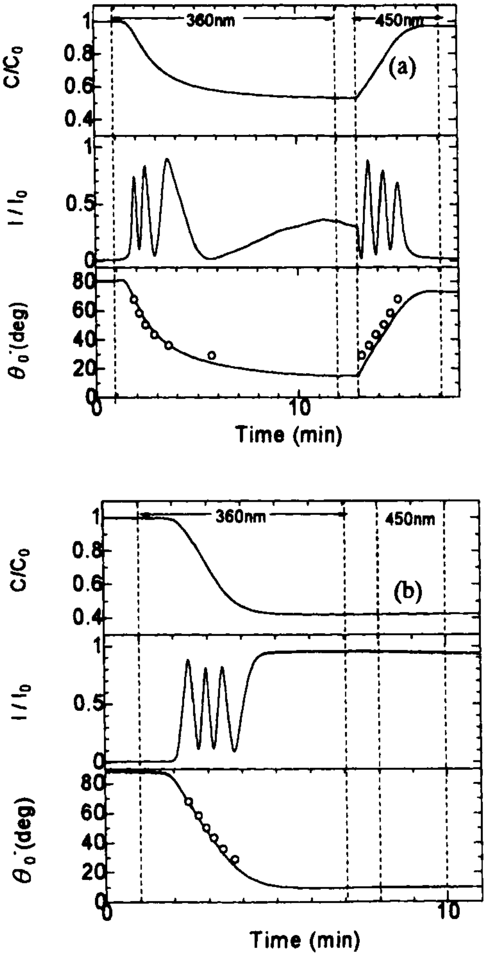

FIGURE 6 A typical example of the capacitance (C/C_0) and optical transmittance (I/I_0) by alternating UV and visible light irradiation. The

tilted angles θ_0^- estimated from the capacitance (C/C_0) are plotted by solid line, and estimated from the optical transmittance are plotted by open circles. (a) with 1-layer PI LB film, and (b) with 5-layer PI LB film.

molecules align homeotropic in the LC cells, and the director orientation transition happens during the UV irradiation on both sides of the electrodes. Further, the deposition of PI LB films is an effective way to prepare LC cells which enable the orientational transition of LC molecules homogeneously. This result was confirmed from the optical microscope observation (not shown here). During the visible light irradiation after the UV irradiation, 3 peaks appear in the transmittance of the LC cells without PI LB films and with 1-layer PI LB films (Fig. 6(a)), whereas the transmittance is nearly constant for the LC cells with 5- and 11-layer film. That is, the director orientational transition happens reversibly in the LC cells without PI LB films and with 1-layer PI LB films (Fig. 6(a)) by the alternating UV and visible lights irradiation. On the other hand, the director orientational transition does not happen reversibly in the LC cells with 5-PI layer LB films (Fig.7(b)). Similar results were obtained for the LC cells with 11-layer PI LB films. These experimental results are tentatively explained by assuming that the 5CBs in LC cells without PI LB films and with 1 layer PI LB films change their conformation from homeotropic to parallel and from parallel to homeotropic by the alternating UV and visible light irradiation. And the 5CBs in LC cells with 5-layer PI LB films change their conformation from homeotropic to parallel by the first UV irradiation and they are then anchored due to the interaction working between 5 CB molecules and PI LB films (or substrate). The anchoring strength of PI LB films with 5 layers is stronger than that with 1- layer and 0-layer films(18) and we explained the relationship between the anchoring strength and the number of PI LB films by a model assuming a van der Waals like interaction working between polar molecules and films. Therefore the 5CB molecules on 5-layer PI LB films cannot change their conformation by visible light irradiation, although their conformation on 1- or 0-layer PI LB films is changed. It is instructive here to note that PI LB films on metals and ITO-coated glass slides are charged due to the excess charges displaced from metals and ITO-coated glass slides, and the amount excess charges increases as the number of deposited PI layer increases and saturates at the number of deposited layers of about 10.(9,10) Thus the electrostatic phenomena observed in LB films will also contribute to the anchoring 12 strength. However, the detail is still not clear.

In Fig.6, we have 4 valleys and 3 hills in the transmittance during the first UV irradiation where the relationship given by Eq. (11) is satisfied. Using the experimental capacitance, we estimated the angle θ_0^- which gives the director distribution $\theta(z) = \theta_0^-$ $(0 \leqq z \leqq L)$, and then plotted in the bottom of the figure. Similarly, using the hills and valleys observed in the transmittance, the angle $\theta(z) = \theta_0^-$ which satisfies the relationship given by Eq. (11) was estimated, and the results were plotted as open circles in the bottom of the figure. As we can see from Fig. 7, the angle θ_0^- estimated from the capacitance agrees well with that estimated from the transmittance in Figs. 6 (a) and (b), whereas these angles are not in good agreement for LC cellls without PI Lbfilms. This result again suggests that the deposition of PI LB films is an effective way to prepare LC cells working purposefully by cis-trans photo-isomerization.

4. CONCLUSION

The orientational transition process of 5CB liquid crystal monolayers by compression and the cis-trans photoisomerization process of surface azobenzene monolayer were detected by the MDC measurement. The orientational transition process of 5CB molecules in Liquid crystal (LC) cells triggered by the conformational change of the surface azobezene monolayer on PI LB films were also examined, and the results were discussed with taking into account the physico-chemical property of PI LB films.

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