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Variable liquid crystal pretilt angles generated by photoalignment in homeotropically aligned azo dye-doped liquid crystals

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This letter demonstrates the feasibility of producing variable liquid crystal (LC) pretilt angles using light-induced ripple structures (LIRSs) in homeotropically aligned azo dye-doped liquid crystals (ADDLCs). Illuminating homeotropically aligned ADDLCs with a linearly polarized light for a suitable period yields the LIRSs which provide LCs an anisotropic homogeneous anchoring force. Experimentally, the effective alignment force produced by the homeotropic alignment layer and the LIRSs determines the LC pretilt angle (24° to 63.5°), defined from the normal to the substrate. A no-bias pi cell for liquid crystal displays is demonstrated using this approach. © *2009 American Institute of Physics*. [doi:10.1063/1.3253413]

The use of liquid crystals (LCs) requires a surface LC alignment on the substrate. Most LC devices are based on either homogeneous or homeotropic alignment. Recently, controlling intermediate pretilt angles of LCs has attracted increasing interest because of its potential use in the no-bias pi cell for liquid crystal displays.[1–3] Accordingly, various approaches for generating LC pretilt angles, from about 0° to 90°, have been reported.[3–9] Some of these methods raise many technical disadvantages, such as the need for mechanical rubbing and complexity. By overcoming these shortcomings, the noncontact approach for photoalignment in LCs, therefore, has become increasingly important. Photoalignment by photoisomerization using the light-induced adsorption of azo dye molecules, such as methyl red (MR) doped in LCs, has been extensively discussed.[10–16] Notably, MR adsorption markedly depends on the substrate surface.[15]

Another photoalignment approach, which involves the light-induced ripple structures (LIRSs) on the adsorbed azo dyes in ADDLCs, has also been reported.[12–15] The previous letters reported by the authors have demonstrated that the morphologies of the LIRSs in ADDLCs depend markedly on the intensity and the wavelength of the impinging light, the period of illumination, and the ambient temperature.[12–15] These related works have focused on the periodicity, the amplitude, the orientation, and the homogeneously anchoring force exerted by the developed LIRSs.

This letter presents a method for fabricating LC cells with various LC pretilt angles by LIRSs in homeotropically aligned ADDLCs. The formed LC pretilt angle, defined in this case the angle made between the LC director and the surface normal of the substrate, can be controlled from about 24° to 63.5°. Experimentally, the amplitude of the formed LIRSs increases with the period of illumination. Based on the Berreman theory,[17–19] the unidirectional anchoring force exerted by the LIRSs is proportional to the amplitude of the LIRSs. Additionally, based on the dual easy axis model,[20] the combination of the constant homeotropic alignment force exerted by the initially homeotropic alignment layer and the variable homogeneous forces exerted by the LIRSs determines the final LC pretilt angles. Restated, a homeotropically aligned ADDLC sample becomes a hybrid one (inset in Fig. 3). Additionally, a no-bias pi cell is demonstrated using this approach.

The ADDLC composite used herein was prepared by mixing 99 wt % nematic LC (E7, from Merck, clearing temperature ~61 °C) with 1 wt % azo dye (MR, from Aldrich). The dichroic ratio of MR is approximately six for visible light.[14] To promote an empty cell with homeotropic alignment, a film, N, N-dimethyl-N-octadecyl-3-aminoprophyltrimethoxy-ailyl chloride (DMOAP), was coated onto two indium-tin-oxide-coated glass slides that were separated by two 27 μm-thick plastic spacers. The homogeneously mixed compound was then injected into an empty cell by capillary action. Additionally, to increase the absorbance of green light by MR in a homeotropically aligned ADDLC cell, the temperature of the sample was maintained at 55 °C during illumination.

Figures 1(a) and 1(b) present the experimental setups for forming the LIRSs by the photoalignment in homeotropically aligned ADDLCs and for measuring the T-V curve of the formed hybrid LC cell, respectively. As presented in Fig. 1(a), a linearly polarized green laser beam ($\boldsymbol{E_G}$ along $\boldsymbol{y}$-axis, from an Ar^+ laser, λ_G=514.5 nm), with an intensity of ~280 mW/cm^2, was adopted as a pump beam, which was normally incident (along z-axis) onto the sample from one substrate, referred as the command surface (S_C). The other was called reference surface (S_R). After ADDLCs had been pumped by the green laser beam for the specified periods, LIRSs with various amplitudes were formed on S_C. Finally, the generated LIRSs on S_C and the DMOAP alignment layer on both S_R and S_C yielded hybrid LC cells with various pretilt angles. As presented in Fig. 1(b), a red probe beam

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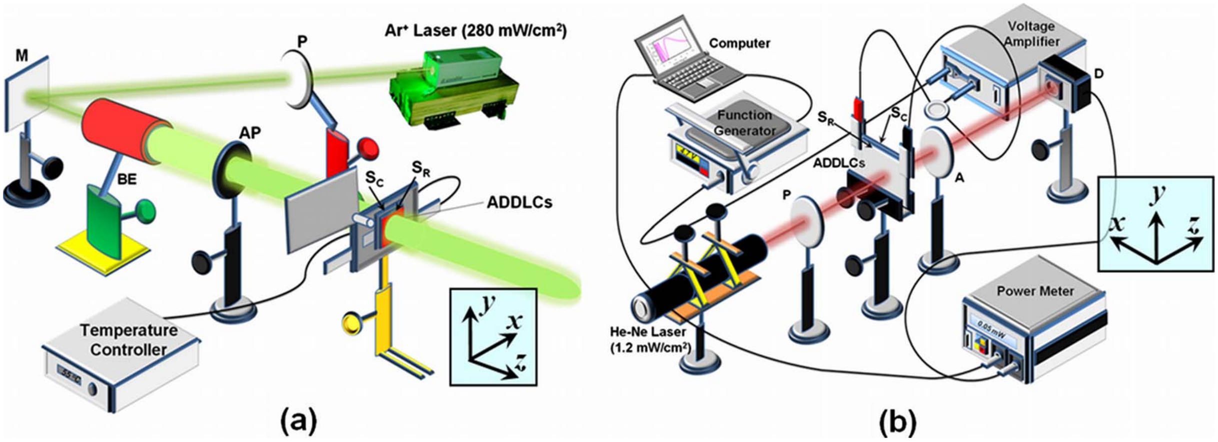

FIG. 1. (Color online) Experimental setups for (a) forming LIRSs in ADDLCs; (b) measuring the T-V curve of samples. P, A, M, BE, AP, and D represent polarizer, analyzer, mirror, beam expander, aperture, and detector, respectively.

from a He-Ne laser ($\lambda=632.8$ nm), with an intensity of 1.2 mW/cm^2, $\boldsymbol{E_R}$, linearly polarized at an angle 45° with respect to $\boldsymbol{E_G}$, was normally incident onto the ADDLC sample. The transmission of the sample under the application of an ac (1 KHz) voltage was measured and fitted with the simulated result by 1D-DIMOS software to obtain the pretilt angles.[8]

The approach for generating various LC pretilt angles presented herein involves setting the period of illumination at 15, 20, 25, 40, 60, 90, and 180 min. LIRSs are not observed experimentally when the period of illumination is less than 15 min in this case. Extending the period of illumination yields the LIRSs and produces an anisotropic homogeneous anchoring force that aligns LCs. The combination of the DMOAP film, which results in the vertical alignment, and the LIRSs, which are associated with homogeneous alignment, enables various pretilt angles to be achieved. Figure 2 plots the measured T-V curves of the ADDLC samples that were illuminated for 25 and 90 min, together with the simulated curves. Clearly, the experimental results qualitatively fit the simulated ones well, such that the pretilt angles of the 25- and 90-min-illumination samples are determined to be around 43.5° and 56°, respectively.

Figure 3 plots the measured pretilt angle as a function of the period of illumination with green laser beam. The LC pretilt angles, ranging from 24° to 63.5°, increase with the period of illumination. The results in Fig. 3 show that the rate of increase in the pretilt angles upon irradiation with green laser beam is initially large and almost saturates at 40 min. Because the interference between the impinging polarized light and the light scattered from the surface is the key to produce the LIRSs, and since the transmittance (scattering) of the impinging light, caused by the azo dye-adsorbed layer, is verified to increase (decrease) as the period of illumination in the late stage of the photoalignment process increases.[15] Notably, according to our previous study,[21] the adsorbed dyes are stable at room temperature, but can be thermally erased and optically rewritten.

The morphologies of the S_R, and the formed LIRSs on S_C of the 90-min-illumination sample are analyzed using an atomic force microscope (AFM). Figure 4(a) presents the two-dimensional (2D) AFM image of the S_R. Clearly, the LIRSs were not formed on S_R, because most of the azo dyes in the sample are diffused toward the impinging light and adsorbed on S_C. Figures 4(b) and 4(c) present the 2D and three-dimensional (3D) AFM images, respectively, of S_C. Refer to Ref. 14, the spacing (Λ) of the formed LIRSs in an ADDLC system is $\lambda/n \cos \theta$, where λ and θ are the wavelength in vacuum and the angle of incidence of the impinging light, respectively. n is the refractive index of the material. Substituting $\lambda=514.5$ nm, $n\sim1.6$, and $\theta=0°$ into the equation yields a spacing of ~320 nm, which agrees with that measured from Figs. 4(b) and 4(c). The average amplitudes of the LIRSs, generated by irradiation with an Ar$^+$ laser beam for 15, 20, 25, 40, 60, 90, and 180 min are 80, 90, 100, 110, 120, 140, and 150 nm, respectively. The average amplitude of the LIRSs is proportional to the period of illumina-

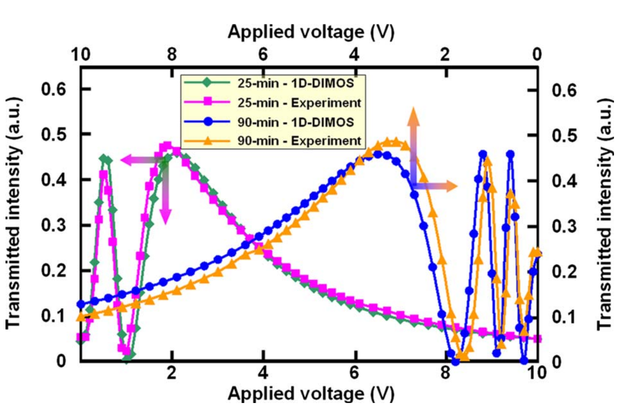

FIG. 2. (Color online) The simulated (green diamonds) [blue dots] and experimental (purple squares) [yellow triangles] T-V curves of ADDLC sample illuminated for 25 [90] mins.

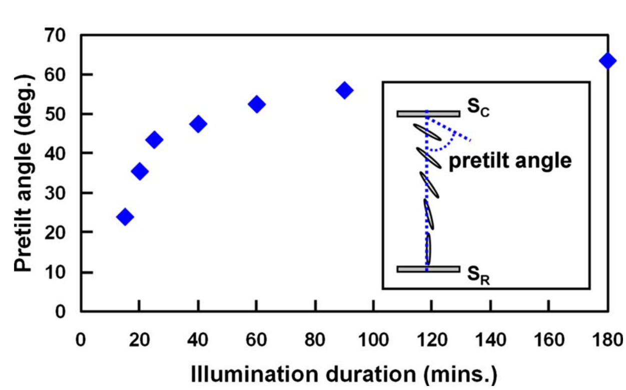

FIG. 3. (Color online) Pretilt angle as a function of the period of illumination. The inset defines the pretilt angle.

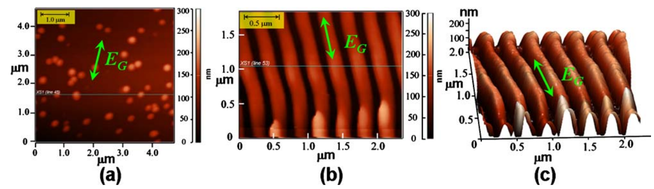

FIG. 4. (Color online) AFM images of the 90-min-illumination sample. (a) 2D AFM image of S_R, (b) 2D, and (c) 3D AFM images of formed LIRSs.

tion. Furthermore, according to Fig. 3, the achieved pretilt angle is proportional to the average amplitude of the LIRSs. The anchoring energy (W) provided by LIRSs is estimated to be,[17–19]

$$W = 2\pi^3 A^2 K_{33} \sin^4 \phi / \Lambda^3 \sqrt{\cos^2 \phi + (K_{33}/K_{11}) \sin^2 \phi}, \quad (1)$$

where A, K_{33} (K_{11}), and ϕ are, respectively, the amplitude, the bend (splay) elastic constant of the used LC, and the angle between the ripple direction and the director at the other substrate. Substituting A, given above (80–140 nm), $\Lambda \sim 320$ nm, $K_{33} \sim 19.5$ pN, $K_{11} \sim 12$ pN, and $\phi = 90°$ into Eq. (1) yields W of the order of 10^{-4} J/m^2, which is a typical value of anchoring energy for aligning LC.[22] The combination of the initial alignment layer and the variable LIRSs determines the final LC pretilt angles. Hence, the value of LC pretilt angle is proportional to the anchoring energy.

This approach was adopted to fabricate two substrates with a pretilt angle 40° (~50° from the surface) to show a no-bias pi cell. A pi cell is more stable in the bend state than in the splay state when the pretilt angle exceeds 47° from the surface of the substrate.[1–3] Experimentally, two ADDLC samples were separately irradiated for about 25 min using the experimental setup that is presented in Fig. 1(a), and then carefully separated into two S_C and two S_R substrates. A no-bias pi cell was fabricated using these two S_C substrates. Additionally, a homogeneous cell with pretilt angle 40° was constructed as well. The cell gap of these two cells was ~7 μm. Figure 5 shows both the experimental and the simulated T-V curves of the no-bias pi cell and the homogeneous cell. The experimental results are consistent with the simulated results obtained using 1D-DIMOS software.

In conclusion, this letter demonstrates the feasibility of obtaining variable LC pretilt angles by forming LIRSs in homeotropically aligned ADDLCs. The combination of the fixed alignment force due to the homeotropic alignment layer and the variable one induced by LIRSs offers an approach to achieve various LC pretilt angles from 24° to 63.5°. A no-bias pi cell is demonstrated using this approach. It is believed that, based on dual easy axis model, larger pretilt angles can be achieved using suitably weaker homeotropic treatments. An experiment is underway to verify this approach.

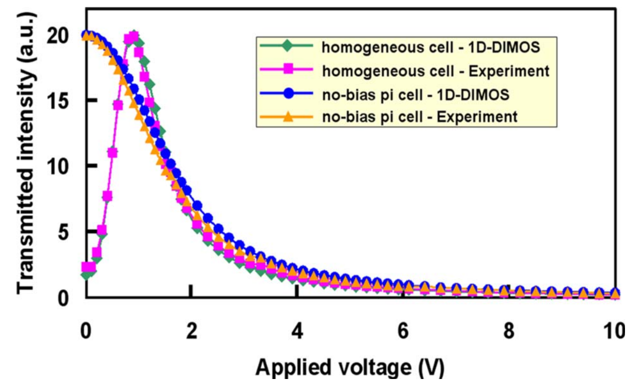

FIG. 5. (Color online) The simulated (green diamonds) and experimental (purple squares) T-V curves of homogeneous cell with pretilt angle of 40° and the simulated (blue dots) and experimental (yellow triangles) T-V curves of no-bias pi cell.

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