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A Polarizer-free Liquid Crystal Display using Dye-doped Liquid Crystal Gels

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1. Introduction

Liquid crystal (LC) based photonic devices are important in light modulated applications including amplitude modulation and phase modulation. [Yang *et al.* (2006)] The anisotropic properties of LC result in employing two polarizers in most of LC devices. The optical efficiency ($\sim 3\%$) and the viewing angle are limited. Therefore, it is highly desirable to develop polarizer-free LC devices. [Yang *et al.* (2006); Lin *et al.* (2008)] In polarizer-free liquid crystal displays (LCDs), two types are demonstrated. One is polarizer-free Guest-Host LCD which obtains dark state by doping small amount of dichroic dye molecules into LC host. [White *et al.* (1974); Cole *et al.* (1977); Bahadur (1992); Wu *et al.* (2001); Yang (2008)] However, the contrast ratio and reflectance are low due to the dichroic ratio ($\sim 10:1$) of dyes. The other is scattering-absorption type, which combines light scattering and absorption. The second type is to mimic the display shown in white paper. In a printed paper, the printed areas turn out dark because the ink absorbs light and light is scattered by the fibers in the white paper. In order to obtain scattering effect, the polymer and liquid crystal complex system is used, including polymer dispersed liquid crystals (PDLCs), polymer networks liquid crystals or liquid crystal gels. [Drzaic (1995)] The scattering mainly results from the mismatch of refractive indices of polymer networks and liquid crystal molecules. To further increase light absorption, dye molecules are doped into the polymer and liquid crystal complex system, for example, dye-doped PDLCs [Drzaic (1995); Lin *et al.* (2004)], and dye-doped LC gels. [Lin *et al.* (2005); Lin *et al.* (2006); Lin *et al.* (2008); Lin *et al.* (2009)] Contrast ratio (CR) of dye-doped PDLC is still not good enough because the dye solubility with polymer matrix, the order parameter of dye and dichroic ratio (typically $\sim 10:1$) of dye. [Drzaic (1995); Lin *et al.* (2004)] In 2005, we have developed a polarizer-free LCD using a dye-doped dualfrequency liquid crystal (DFLC) gel on the ITO-only glass substrates [Lin *et al.* (2005); Lin *et al.* (2006)]. Although its contrast ratio reaches $\sim 150:1$ and response time ~ 6 ms under frequency modulation and the laser-based measurement, the frequency driving scheme, high driving voltage ($\sim 30 V_{\text{rms}}$) and unavoidable dielectric heating effect. [Wen *et al.* (2005); Yin *et al.* (2006)] need to be overcome for TFT-LCDs and flexible displays applications.

To avoid the dielectric heating effect of DFLC, negative LC within vertical alignment layer is a good alternative and it is suitable for making a transflective LCD [Lin *et al.* (2006)]. The

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gel-like feature of materials, vertically aligned polymer network and low temperature processes drives us to realize a trim-able and bendable polarizer-free flexible display in reflective mode. In 2008, we demonstrated a polarizer-free flexible electro-optical switch using dye-doped LC gels which is polarizer-free, fast response, high contrast. [Lin *et al.* (2008)] Many parameters affect the phase separation process [Yang *et al.* (2006); Ren *et al.* (2008)] and then have influence on the electro-optical properties of dye-doped LC gels, such as curing temperature effect. The normally white gels exhibit ~55% reflectance, ~450:1 contrast ratio, ~6.4 ms response time, and ~30 V_{rms} at $f=1$ kHz driving voltage at curing temperature 10 °C. A single pixel flexible reflective display using such dye-doped LC gels are also demonstrated under bending and trimming. To further realize a display with multi-pixels, the substrate is patterned by pixilated indium-tin-oxide (ITO) in general. However, the involved fabrication and driving are complicated. It also causes problems especially in the fabrication process of flexible displays, such as chemical stability of plastic substrates, failure of ITO under tension and so on. [Crawford (2005)] In order to achieve a simple and easy process for flexible displays, we developed a multiple-step switch using distinct dye-doped LC gels without patterning ITO layers in 2009. [Lin *et al.* (2009)] Moreover, the switch should provide extra information states besides voltage-on and voltage-off states for the applications of the decorative displays within a simple driving and manufacturing process. The distinct dye-doped LC gels can display information by the spatial distribution of polymer network density without patterned ITO layers. The different regions of polymer network densities have different threshold voltages, but the similar bright and dark states. The distinct dye-doped LC gel is transparent (or in bright state) at 0 V_{rms} and opaque (or in dark state) at 30 V_{rms} . At 9 V_{rms} , it shows the colored pattern (or information) because of distinct polymer networks. The response time is ~10 ms and contrast ratio is ~200:1. We can also extend the concept for a polarizer-free four step switch. The potential applications are for decorative displays, electrically tunable diaphragm, and electrically tunable low pass or high pass filter.

In this book chapter, we introduce a polarizer-free LCD using dye-doped LC gels whose the physical mechanism is mainly the combination of both light scattering and absorption. In the beginning, we introduce the structure and mechanism of our dye-doped liquid crystal gels. Second, the experimental setups, results (morphologies, electro-optical properties, response time) and mathematical model are also discussed. Third, the performance of such a polarizer-free liquid crystal display is shown. Finally, we will introduce multiple step switches using distinct dye-doped LC gels for the application of decorative displays. The potential applications are flexible displays, electrically tunable light shutters, and decorative displays.

2. Polarizer-free dye-doped liquid crystal gels

2.1 Structures and operating principles

The structure and operation principles of the dye-doped LC gel are schematically depicted in Fig. 1. At $V=0$, the cell does not scatter light and the absorption is rather weak due to the vertically aligned polymer networks, liquid crystal directors and dye molecules. Therefore, the display has the highest reflectance. When the applied voltage (V_1) is larger than the threshold voltage (V_{th}), the LC directors with a negative dielectric anisotropy are tilted away from the electric field and LC molecules bring dye molecules to rotate with them, as shown in Fig. 1. The cell is then switched into a multi-domain structure. As a result, both of

scattering and absorption increase. Then the reflectance decreases. In addition, all the LC directors and dye molecules have the same tilt angle but random orientations. Therefore, the reflectance is polarization-independent. When we further increase the voltage ($V_2 \gg V_{th}$), the LC directors and dye molecules are aligned along x-y plane randomly, as Fig. 1 depicts. The scattering and absorption are maximal. Besides, all the polarizations of incident light experience the same averaged scattering and absorption. As a result, the dye-doped LC gel appears black and is still polarization independent. The appearance of color is mainly because of the light absorption of dye. The scattering and reflection assist the multiple absorption due to the elongate the paths of light propagation.

Fig. 1. Schematic operating principle of dye-doped liquid crystal display at $V=0$, $V_1 > V_{th}$, and $V_2 \gg V_{th}$. The alignment layer has no rubbing treatment.

2.2 Fabrications

The dye-doped LC gel we employed is a mixture of negative nematic liquid crystal ZLI-4788 (Merck, $n_e = 1.6567$, $\Delta n = 0.1647$ at $\lambda = 589$ nm; $\Delta\epsilon = -5.7$ at $f = 1$ kHz) and a diacrylate monomer (bisphenol-A-dimethacrylate) with a dichroic dye S428 (Mitsui, Japan) at 90:5:5 wt% ratios. The structure of the diacrylate monomer is shown as follows:

The dye-doped LC mixture was then injected into an empty cell consisting of two glass substrates whose inner surfaces were coated with a thin conductive layer, indium-tin-oxide (ITO) and polyimide (PI) layer *without* rubbing treatment. The PI layer provides vertical alignment for the LC directors. The cell gap was 5 μm . The filled cell was irradiated by a UV

light ($\lambda \sim 365$ nm, $I \sim 3$ mW/cm²). The cell was cured at a fixed temperature for 1.5 hr. Such a fixed temperature is called curing temperature. After photo-polymerization, the formed chainlike polymer networks are along the z direction because the LC directors are aligned perpendicular to the glass substrates during the UV curing process, as shown in Fig. 1.

2.3 Morphologies

Fig. 2 show the morphologies observing under an optical microscope with a single polarizer only. The top region of the two regions in Fig. 2 is the patterned ITO area. The bright region represents the state of $V=0$. The dark area represents the ITO electrodes applied $30 V_{\text{rms}}$ at $f=1$ kHz. At the voltage-off state, the cell shows good bright state because of the vertically aligned polymer networks, LC and dye molecules. At $30 V_{\text{rms}}$, it shows the fine domain textures of the polymer networks, and red color because of dye molecules, as shown in Fig. 2. Our LC cell shows good dark and bright states although the dark state up to now is redish, not truly black.

Fig. 2. The microscopic images of dye-doped LC gels at $0 V_{\text{rms}}$ and $30 V_{\text{rms}}$.

The photographs of Scanning Electron Microscopy (SEM) are shown in Fig. 3 (a)-(h). Fig. 3 (a), (c), (e) and (g) are the top views of the cells at different curing temperatures (T). Fig. 3 (b), (d), (f), and (h) are the side views of the cells at different curing temperatures. In Fig. 3 (b), (d), (f), and (h), the polymer networks are perpendicular to the glass substrates. The polymer networks of dye-doped LC gels consist of chain-linked polymer grains. The averaged sizes of polymer grains measured from Fig. 3 (a), (c), (e) and (g) are around 68 nm at $T=10^\circ\text{C}$, 94nm at $T=20^\circ\text{C}$, 125nm at $T=30^\circ\text{C}$, 132nm at $T=40^\circ\text{C}$. The averaged domain sizes of polymer networks measured from Fig. 3 (b), (d), (f), and (h) are around 3.25 μm at $T=10^\circ\text{C}$, 4.62 μm at $T=20^\circ\text{C}$, 4.78 μm at $T=30^\circ\text{C}$, 6.12 μm at $T=40^\circ\text{C}$. Both of the domain sizes and the size of polymer grains increase with curing temperatures. The scattering is mainly because of the domain sizes of polymer networks because the domain sizes of polymer networks are near wavelength of incident light while the sizes of polymer grains are smaller than the wavelength.

2.4 Electro-optical properties

We adopt the typical reflectance measurement to measure the electro-optical properties of dye-doped LC gels. Because the guest-host system we employed appears dark red rather than black, we used an unpolarized green He-Ne laser ($\lambda=543.5$ nm, Melles Griot, Model 05-LGR-173) instead of a white light source for characterizing the device performances. A dielectric mirror was placed behind the cell so that the laser beam passed through the cell twice. A large area photodiode detector (New Focus, Model 2031) was placed at ~ 25 cm (the

Fig. 3. SEM photographs of dye-doped LC gels at curing temperatures 10°C (a, b), 20°C (c, d), 30°C (e, f), and 40°C (g, h). The LC and dye were extracted. (a), (c), (e) and (g) are the top views of the cells. (b), (d), (f), and (h) are the side views of the cells. The white-indicated bars in (a), (c), (e) and (g) are 100 nm.

normal distance for viewing a mobile display) behind the sample which corresponds to $\sim 2^\circ$ collection angle. A computer controlled LabVIEW data acquisition system was used for driving the sample and recording the light reflectance. In order to prove our dye-doped LC gel is polarization independent, we placed a polarizer between the laser and the LC cell. The reflectance as a function of an angle of the polarizer at different applied voltages is shown in Fig. 4. The variation of reflectance is less than 5% when we rotated the polarizer. It indicates the dye-doped LC gels are indeed polarization independent at all applied voltages. The reflectance at 0 V_{rms} is around 50%.

Fig. 4. Reflectance as a function of angle of the polarizer at different applied voltages. The curing temperature was 20 °C.

2.4.1 Curing temperature effect

After we removed the polarizer, the incident light then was unpolarized green laser beam. Fig. 5 (a) is the measured voltage-dependant reflectance of the dye-doped LC gels at various curing temperatures. The reflectance was normalized to that of a pure LC cell with the same cell gap. The reflectance decreases gradually with the applied voltage $V > V_{\text{th}}$ due to the increases of the scattering and the absorption. As curing temperature decreases, the increases ($\sim 40\%$ to $\sim 55\%$) of maximum reflectance at $V=0$ and that is because of the better vertical alignment of LC directors, dye molecules and polymer networks at a low curing temperature. The contrast ratio (CR) is defined as a reflectance ratio of 0 V_{rms} to 30 V_{rms} . The CRs are $\sim 450:1$ at 10 °C, $\sim 250:1$ at 20 °C, $\sim 200:1$ at 30 °C, and $\sim 300:1$ at 40 °C. The contrast ratio decreases as $T < 30^\circ\text{C}$ and then increases as $T > 30^\circ\text{C}$. That is because the increase of a curing temperature results in larger polydomains; therefore, the CR and threshold voltage decrease. Moreover, the decay time increases, as shown in Fig. 5(b). When the temperature is higher than 30°C , we found the cell has dynamic scattering, a fluctuation of liquid crystal directors in polymer domains, to help rebooting the contrast ratio in spite of the larger domain size. To lower the driving voltage, a high birefringence and high absolute value of dielectric anisotropy ($\Delta\epsilon$) of a negative LC and slightly lower polymer concentration could be considered.

Response time is also an important parameter for Guest-Host LCDs. The response time of the dye-doped LC gels was measured using 30 V_{rms} squared pulses with time duration 500ms at $f=1$ kHz. The curing temperature-dependent rise times and decay times are shown in Fig. 5(b). The rise times are about 0.4 ms and the decay time decreases with decreasing curing temperatures. A typical response time of a guest-host display is around 50 ms. The response time of our dye-doped LC gel ($\sim 6.4\text{ms}$) is faster because polymer network helps LC directors to relax back. The rise time is ~ 0.4 ms and decay time is ~ 6 ms at 10 °C.

Fig. 5. (a) Voltage-dependent reflectance at various curing temperature. (b) Curing temperature-dependent response time

Besides the curing temperature, the UV curing intensities, monomer concentrations and dye concentrations also affect the performance of the dye-doped LC gels. We discuss in the following sections.

2.4.2 Curing UV intensity effect

To examine the effect of UV curing intensity (I), we prepared four samples with the same mixtures at the curing temperature $10^{\circ}C$, but at different UV curing intensities which were 2.6, 1.37, 0.733, 0.354 mW/cm^2 respectively. The cell gaps were still $5 \mu m$. The measured reflectance as a function of voltage is shown in Fig. 6(a). With the increases of UV curing intensity, threshold voltage (V_{th}) increases from $4.82 V_{rms}$ ($I \sim 0.354 \text{ mW/cm}^2$) to $5.92 V_{rms}$ ($I \sim 2.6 \text{ mW/cm}^2$). The maximal reflectance at $V=0$ increases from 44% ($I \sim 0.354 \text{ mW/cm}^2$) to $\sim 57\%$ ($I \sim 2.6 \text{ mW/cm}^2$). In Fig. 6(b), rise time is around 0.4 ms, but decay time decreases from 9 ms ($I \sim 0.354 \text{ mW/cm}^2$) to 6.88 ms ($I \sim 2.6 \text{ mW/cm}^2$). The larger UV curing intensity results in smaller domain size of polymer networks. That causes a stronger anchoring energy and then enlarges the threshold voltage. Therefore, it boosts the response. The reflectance at $V=0$ decreases under higher UV curing intensity owing to better vertically alignment at $V=0$. Generally speaking, UV curing intensity does not affect the electrooptical

properties of dye-doped LC gels dramatically. However, weak UV curing intensity can result in the longer curing time in order to complete the polymerization.

(a)

(b)

Fig. 6. (a) Voltage-dependent reflectance at various curing intensity and (b) response time as a function of UV curing intensity. The curing temperature was 10 °C.

2.4.3 Monomer concentration

In this section, the monomer concentration effect is discussed. We prepared three cells at different monomer concentrations which were 3 wt%, 5 wt%, and 7 wt%. The fabrication processes of the cells were at the same UV curing intensity 2.6 mW/cm² and also at the same curing temperature 20 °C. The cell gaps were still 5 μm. The voltage-dependent reflectance at different monomer concentrations is shown in Fig. 7(a). The threshold voltage increases from 2.1 V_{rms} (at 3 wt% M1) to 6.52 V_{rms} (at 7 wt% M1) due to the denser polymer networks. The reflectance at V=0 decreases slightly (from 51% at 3 wt% M1 to 46% at 7 wt% M1). That is because denser polymer networks affect the vertical alignment of LC directors and also increases the scattering slightly. CR increases from 222 : 1 (at 3 wt% M1) to 486 : 1 (at 7 wt% M1) owing to better scattering of higher monomer concentration at the high driving voltage. Fig. 7(b) shows the measured response time as a function of monomer concentration. Rise time is around 0.2 ms-0.4 ms, and decay time increases from 52 ms (at 3 wt% M1) to 7.3 ms (at 7 wt% M1).

The higher monomer concentration has smaller domain sizes; therefore, the LC directors are relaxed back faster due to stronger anchoring energy after turning off the applied voltage. To lower driving voltage, we can reduce the monomer concentration; however, the polymer network of dye-doped LC gels is not stable as the monomer concentration is too low.

Fig. 7. (a) Voltage-dependent reflectance at different monomer concentrations. (b) Measured response time as a function of monomer concentration.

2.4.4 Dye concentration

As to the effect of dye concentration, three cells with the different dye concentrations, 3 wt%, 5 wt%, and 7 wt%, were prepared at the same curing temperature 20 °C and at the same UV curing intensity 2.6 mW/cm². The cell gaps were 5 μm. Fig. 8(a) plots measured voltage-dependent reflectance at three dye concentrations. The reflectance at $V=0$ decreases from ~63% (at 3 wt% S428) to ~42.5% (7 wt% S428) as the dye concentration increases due to the increases of absorption, even at the same concentration of monomer. The threshold voltages of cells with 3 wt%, 5 wt%, and 7 wt% S428 are almost the same around 5 V_{rms}. That is because the monomer concentrations are the same. The CRs of the cells with 3 wt%, 5 wt%, and 7 wt% S428 are 668:1, 342:1, and 31:1, respectively. Higher dye monomer concentration reduces not only reflectance at $V=0$, but also degrades the dark state at $V=30$.

In Fig. 8(b), rise times are around 0.3 ms and the decay time increases 2 times from 9.8 ms at 3wt% S428 to 20.96 ms at 7wt% S428) with the dye concentration. That is because dye molecules hinder LC directors relax back at higher concentration of dye under same monomer concentration.

Fig. 8. (a) Voltage-dependent reflectance at different dye concentrations. (b) Measured response time as a function of dye concentration.

3. Mathematical models

By considering the scattering and absorption, the reflectance ($R(\theta)$) as a function of tilt angle (θ) of LC directors with respect to x-axis can be expressed as:

$$R(\theta) \approx e^{-\alpha_{ave}(\theta) \cdot 2d} \cdot e^{-\beta_{ave}(\theta) \cdot 2d} \quad (1)$$

where d is cell gap, $\alpha_{ave}(\theta)$ is the average absorption coefficient, and $\beta_{ave}(\theta)$ is the average scattering coefficient. $\alpha_{ave}(\theta)$ and $\beta_{ave}(\theta)$ satisfy the following equations.

$$\alpha_{ave}(\theta) = \rho_1 \cdot \frac{\alpha_{eff}(\theta) + \alpha_{\perp}}{2} \quad (2)$$

$$\beta_{ave}(\theta) = \rho_0 \cdot \frac{\sigma_{eff}(\theta)}{V} \quad (3)$$

$\alpha_{eff}(\theta)$ can be expressed as:

$$\alpha_{eff}(\theta) = \frac{\alpha_{||} \cdot \alpha_{\perp}}{\sqrt{\alpha_{||} \cdot \cos^2 \theta + \alpha_{\perp} \cdot \sin^2 \theta}} \quad (4)$$

where ρ_1 is the dye concentration, $\alpha_{||}$ and $\alpha_{\perp}$ are the absorption coefficients when the polarization of incident light is parallel or perpendicular to the principal axis of dye molecule. ρ_0 is the LC concentration, V is the average volume of a domain. σ_{eff} in Eq. (4) is the effective scattering cross section from all liquid crystal droplets and can be expressed as:

$$\sigma_{eff}(\theta) = \frac{1}{\pi} \int_0^{\pi} \sigma_s(\theta, \alpha_o) \cdot d\alpha_o \quad (5)$$

Based on anomalous diffraction approach [Drzaic (1995)], scattering cross section results from a single LC domain is:

$$\sigma_s(\theta, \delta) = 2\sigma_o [H_{ve}(\theta) \cdot \cos^2 \delta + H_{vo} \cdot \sin^2 \delta] \quad (6)$$

where σ_o is the geometrical optics cross section related to the domain size, δ is the polarization angle; $H_{ve}(\theta)$ and $H_{vo}(\theta)$ stand for phase shift induced by e-ray and o-ray respectively. The averaged domain sizes of polymer networks are measured around 1.5 μm ~3 μm . For numerical calculations we have chosen the following parameters according to the experiments: $\rho_1=0.05 \text{ g/cm}^3$, and $\rho_0=0.89 \text{ g/cm}^3$. $\alpha_{||}=11.83 \text{ }\mu\text{m}^{-1}$ and $\alpha_{\perp}=0.926 \text{ }\mu\text{m}^{-1}$. The simulation result is shown in Fig. 9. In Fig. 9, the reflectance decreases with the tilt angle. The R- θ curve shifts to right as the domain size is smaller. The simulation results agree with the experimental results in Fig. 5. The smaller domain size or larger density of polymer networks can result in the larger operating voltage and better dark state. We can adjust UV intensity, curing temperature, the controlled temperature under UV illumination, and concentration of LC, dye or monomer to change the domain sizes of polymer networks.

Fig. 9. Calculated reflectance as a function of tilt angle in dye-doped LC gels at different domain sizes.

4. Polarizer-free flexible LCDs using dye-doped LC gels

Roll-able, bendable, trim-able, and conformable paper-like flexible displays are useful for electronic paper, electronic tag, and decorative displays. [Crawford (2005)] Many liquid crystal (LC) technologies, such as polymer-dispersed liquid crystals (PDLC) [Mach *et al.* (2001); Sheraw *et al.* (2002); Hohnholz *et al.* (2005); Buyuktanir *et al.* (2006); Wang *et al.* (2007)], cholesteric liquid crystals [Wu *et al.* (2001); Yang *et al.* (1994); Chari *et al.* (2006); Khan *et al.* (2005); Khan *et al.* (2007)], and single-substrate LCDs using photoenforced stratification [Penterman *et al.* (2002); Raynes (2002); Vogels *et al.* (2004)] or using LC/polymer composites [Kim *et al.* (2002); Kim *et al.* (2004); Lin *et al.* (2006); Ren *et al.* (2007)], and non-liquid crystal technologies, such as electrophoretic imaging [Comiskey *et al.* (1998); Gelinck *et al.* (2004); Daniel *et al.* (2007)], Gyricon [Crowley *et al.* (2002)], and organic light-emitting diode (OLED) [Gu *et al.* (1997); Burrows *et al.* (1997); Krasnov (2002); Sugimoto *et al.* (2004); Zhou *et al.* (2006)], have been carried out to achieve transmissive type or reflective type flexible displays. In liquid crystal-based flexible displays, bistability and colors of cholesteric liquid crystals limits the application due to the complexity of driving and color shift at off angle. Instead of cholesteric liquid crystals, dye-doped LC gels can be used in flexible displays because the dye-doped LC gels is gel-like and the polymer networks of dye-doped LC gels are perpendicular to the glass substrates.

The images of a single pixel of the polarizer-free LCD using dye-doped LC gels at $V=0$ and $V=30 V_{rms}$ are shown in Fig. 10(a). By replacing glass substrates with flexible substrate, dye-doped LC gel is not only bendable but also trim-able because our material is gel-like, as shown in Fig. 10(b). The flexible substrates are provided by EOL/ITRI (Electronics& Optoelectronics Research Laboratories, Industrial Technology Research Institute, Taiwan). IZO was over coated on the top of flexible substrates made by polycarbonate with thickness 120 μm . The cross shaped microstructures made by photo-spacers, resins, were developed on the flexible substrates by photolithography process. The width of photo-spacers is 10 μm and the pitch of photo-spacers is 430 μm . The ambient white light was used to illuminate the cells. However, the CR is degraded. The CR is higher under laser-based measurement because of the collimation of the laser beam. Figure 10(c) is the transmission as a function of radius of curvature under bending at 0 and 30 V_{rms} . The measurement method is two-point bending technique. The transmission of dye-doped LC gels is almost the same as the radius of curvature larger than 21 mm. The dye-doped LC gel is trim-able as well because our material is gel-like, as shown in Fig. 10 (d) and (e). The flexible display performance remains almost the same after cutting by a scissor. Since no polarizer is needed, the residual birefringence of polycarbonate does not affect the performance of our flexible display.

5. Polarizer-free multiple step switches using distinct dye-doped LC gels

In order to realize a display with multi-pixels, we usually pattern the substrate by pixilated indium-tin-oxide (ITO). However, the involved fabrication and driving are complicated. Especially in the fabrication process of flexible displays, chemical stability of plastic substrates and failure of ITO under tension degrade the performance of flexible displays. In order to achieve a simple and easy process for flexible displays, we can develop a multiple step switch using dye-doped LC gels without patterning ITO layers. Moreover, the switch should provide extra information states besides voltage-on and voltage-off states for the applications of the decorative displays within a simple driving and manufacturing process. A polarizer-free multiple step switches using distinct dye-doped liquid crystal gels

Fig. 10. (a) A single pixel polarizer-free reflective LCD using the dye-doped LC gels in glass substrates and (b) in flexible substrates. (c) The transmission as a function of bending radius of curvature. (d)The dye-doped LC gel is trim-able. (e) The voltage dependent transmission before and after trimming. A piece of white paper was used as a diffusive reflector.

is demonstrated in reflective mode. By controlling the spatial distribution of the density of polymer networks, the distinct dye-doped LC gels can produce multiple states: bright, dark and information states without patterning ITO. The multiple states are generated by adapting different polymer network density in different display regions. The dark state is due to the combination of scattering and absorption. In this section, we demonstrated a three step switch using distinct dye-doped LC gels. [Lin *et al.* (2009); Du *et al.* (2003)] The potential applications are decorative displays, electrically tunable iris, and electrically tunable low pass or high pass filter.

5.1 Operating principles of a polarizer-free three step switch

Fig. 11 illustrates the operating principles of a polarizer-free three step switch using distinct dye-doped LC gels. The structure consists of ITO glass substrates, vertical alignment layer without rubbing treatment, a diffusive reflector, negative liquid crystals, dichroic dye molecules, and the distinct distribution of polymer networks which divide the cell into two parts, the low density of polymer networks (LDPN) and the high density of polymer networks (HDPN), as shown in Fig. 11(a). Without an applied voltage ($V=0$), polymer networks, LC and dye molecules are aligned vertically; hence, the cell does not scatter light and the absorption is weak. The cell has high polarization-independent reflectance. When V_1 is larger than the threshold voltage in LDPN (V_{th1}), the LC directors are reoriented first and then bring dye molecules to rotate accordingly due to weak anchoring energy provided by LDPN; meanwhile, the LC directors remain vertically aligned in HDPN, as shown in Fig. 11(b). The reflectance then reduces in LDPN region due to the increases the scattering and absorption while the reflectance remains high in HDPN region. Furthermore, all the LC directors and dye molecules in LDPN have the same tilt angle within random orientations which originate from the alignment layer without rubbing treatment. The reflectance in LDPN is polarization-independent. When the applied voltage is larger than the threshold voltage V_{th2} ($>V_1$) of HDPN, the LC directors in HDPN begin to reorient by the electric fields and then the reflectance in this region decreases as well. When the applied voltage V_2 is much larger than V_{th2} , the negative liquid crystals and dye molecules in both regions are reoriented randomly in the x-y plane, as shown in Fig. 11(c). Thus, the both regions appear polarization-independent dark state.

Fig. 11. Schematic structure and operating principles of distinct dye-doped LC gels at (a) $V=0$, (b) V_1 , and (c) V_2 ($V_2 > V_1$).

5.2 A polarizer-free three step switch using distinct dye-doped liquid crystal gels

The fabrication process is similar to dye-doped LC gels, but spatial distribution of polymer networks. To prepare a cell of distinct dye-doped LC gels, we mixed nematic LC ZLI-4788 with an UV-curable diacrylate monomer and a dichroic dye S428 (Mitsui, Japan) at 90: 5: 5 wt% ratios. In order to generate distinct distribution of polymer networks, two-step UV curing process was applied. First, one region of the cell with filled mixtures was exposed to a UV light ($\lambda \sim 365$ nm) with UV intensity 1.37 mW/cm^2 at 10°C for 1.5 hours to generate HDPN. After that, the other region of the cell was irradiated by the UV light with lower UV intensity 0.73 mW/cm^2 at 20°C for another 1.5 hours to develop LDPN. The higher curing intensity of UV light and lower curing temperature results in HDPN. [Lin *et al.* (2008); Du *et al.* (2003)] The cell gap was $5 \mu\text{m}$. The three step switch can also be a flexible switch by replacing substrates.

Fig. 12. (a) Voltage-dependent reflectance of the three step switch when voltage is ramped up (solid line) and down (dotted line) (b) Images of the three step switch. A white paper was used as a diffusive reflector.

Fig. 12(a) plots the measured voltage-dependent reflectance of the sample for HDPN and LDPN when the voltage is ramped up (solid line) and down (dotted line). The threshold voltage in HDPN ($5.42 \text{ V}_{\text{rms}}$) is larger than that in LDPN ($4.62 \text{ V}_{\text{rms}}$). At $V=0$, the reflectance in HDPN and LDPN are 50.2% and 49.2%, respectively. The CR is still defined as a reflectance ratio of 0 V_{rms} to $30 \text{ V}_{\text{rms}}$. The CRs in both regions are $\sim 200:1$ under laser-based measurement. When we use an integrating sphere (DMS803, product of the AUTRONICMELCHERS GmbH) under a white light source, the CRs in both regions are $\sim 3:1$. Typically CR for newspaper under the same measurement is $\sim 5:1$. Improving the scattering state and dichroic ratio of dye can further improve the CR. The response time (rise time plus decay time) is 11 ms in LDPN and ~ 9 ms in HDPN by using $30 \text{ V}_{\text{rms}}$ squared pulses with time duration 500 ms at $f=1 \text{ kHz}$. The HDPN results in the stronger anchoring

force between the LC and polymer networks; therefore, the threshold voltage is higher and response time is faster. In order to demonstrate the image of the 3-step switch, we patterned the distinct dyedoped LC gels with a photomask of a "C" pattern without patterning the ITO layer, as shown in Fig. 12(b). The cell can show three switching states: bright state at $V=0$, information state at $9 V_{rms}$, and dark state at $30 V_{rms}$.

The distinct dye-doped LC gels show the hysteresis when the voltage is ramped up and down. Without patterning ITO layer, the voltage is applied to different density regions simultaneously. Thus, the sample still can be operated as a three step or a four step switch as long as the electro-optical curves in different density regions are distinguishable no matter ramped-up voltages or ramped-down voltages.

The reflectance in HDPN (R_{high}) and in LDPN (R_{low}) can be expressed as a function of tilt angle θ of LC directors with respect to x-axis (in Fig. 11): [Drzaic (1995); Khoo *et al.* (1993)]

$$R_{high}(\theta) \approx e^{-2d \times (\alpha_1(\theta) + \beta_1(\theta))} \quad (7)$$

$$R_{low}(\theta) \approx e^{-2d \times (\alpha_2(\theta) + \beta_2(\theta))} \quad (8)$$

where d is the cell gap, $\alpha_1(\theta)$ and $\alpha_2(\theta)$ are the averaged absorption coefficients. $\beta_1(\theta)$ and $\beta_2(\theta)$ are the average scattering coefficients. The averaged absorption can be expressed in Eq. (9).

$$\alpha_i(\theta) = \rho \times \frac{\alpha_{eff}(\theta) + \alpha_{\perp}}{2}, i = 1, 2 \quad (9)$$

where ρ is dye concentration, and $\alpha_{\perp}$ is the absorption coefficient for the polarization of incident light perpendicular to the principal axis of the dye molecule. The effective absorption coefficient $\alpha_{eff}(\theta)$ of the dye molecules can be expressed as:

$$\alpha_{eff}(\theta) = \frac{\alpha_{//} \times \alpha_{\perp}}{\sqrt{\alpha_{//} \times \sin^2 \theta + \alpha_{\perp} \times \cos^2 \theta}} \quad (10)$$

Where $\alpha_{//}$ is the parallel absorption coefficient. At $V=0$, θ is near $\pi/2$; consequently, $\alpha_1 \approx \alpha_2$ approximately equals to $\rho \times \alpha_{\perp}$ and $\beta_1(\theta) \approx \beta_2(\theta)$ equals to 0 because of the vertically alignment of LC directors, dye molecules and polymer networks. Therefore, $R_{high}(\theta) \approx R_{low}(\theta) \sim 63\%$ after we calculated by $\rho=0.05$ and $\alpha_{\perp}=0.926 \text{ m}^{-1}$. However, the measured reflectance ($\sim 50\%$) is lower than 63%. That is because of the imperfection of vertical alignment of LC, dye and polymer networks which is limited by the order parameter of dye molecules and the structure of the monomer without a rod-like central core. At V_2 ($\gg V_{th1}$ and V_{th2}) in Fig. 11(c), $\alpha_1(\theta=0) \approx \alpha_2(\theta=0) \approx \rho \times (\alpha_{\perp} + \alpha_{//})$ is around 0.319 m^{-1} since measured $\alpha_{//}$ is 11.83 m^{-1} . Moreover, the scattering is stronger in HDPN due to smaller domains and the mismatch of refractive indices, i.e. $\beta_1(\theta=0) > \beta_2(\theta=0)$; therefore, theoretically $R_{high}(\theta=0)$ is lower than $R_{low}(\theta=0)$ which also means higher CR in HDPN. However, CRs in both regions are similar in our experiment. Owing to the strong anchoring energy in HDPN, LC directors and dye molecules are not totally aligned along x-y plane which means $\theta \neq 0^\circ$ at $30 V_{rms}$; hence, absorption and scattering is not as high as we expect in HDPN. At V_1 (in Fig. 11(b)), $\theta_1 > \theta_2$ due to the different anchoring strengths in two regions. Thus, $\alpha_1(\theta_1) < \alpha_2(\theta_2)$. So does $\beta_1(\theta_1) < \beta_2(\theta_2)$. Therefore, $R_{high}(\theta_1)$ is larger than $R_{low}(\theta_2)$ at V_1 .

6. Conclusion

In conclusion, we have introduced and demonstrated polarizer-free LCDs using dye-doped LC gels. These polarizer-free dye-doped LC gels exhibit high reflectance, high contrast ratio, wide viewing angle, and fast response time. Especially the low temperature process is favorable for flexible displays. The gel-like materials assist stabilizing the flexible display under trimming. Our dye-doped LC gels provide a stable LC mode and open a new window in paper-like flexible displays. A polarizer-free three step switch using distinct dye doped LC gels is also demonstrated. The distinct dye-doped LC gels can display information by the spatial distribution of polymer network densities without patterned ITO layer. The advantages of such polarizer-free switches are 1) polarizer-free, 2) simple fabrication without ITO patterning, and 3) simple driving. However, the issues we have to overcome are high driving voltages, low resolution, non-black colors and long term stability of dye molecules due to photobleach. This concept can also be extended for making a polarizer-free multiple-switch. The potential applications are decorative displays, electrically tunable iris, and electrically tunable low pass or high pass filter.

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