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## Molecular Crystals and Liquid Crystals

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### Polarizer-Free Gradient Dye-Doped Liquid Crystal Gels

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## Polarizer-Free Gradient Dye-Doped Liquid Crystal Gels

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*A multi-switch display using gradient dye-doped LC gels which is still polarizer-free, fast response (~10ms in general), and high contrast (~200:1 in general) has demonstrated. By controlling the spatial distribution of the density of polymer networks through fabrication process, gradient dye-doped LC gels can be a multi-switch. The gradient dye-doped LC gel is bright without applied voltage and is dark at a high voltage. It appears the colored pattern when LCs are partially reoriented due to the gradient density of polymer networks. The optical analysis of dye-doped LC gels is also discussed. The potential applications are flexible display and decorative displays.*

**Keywords:** gel; guest-host display; liquid crystal; polarization-independent; polarizer-free

### I. INTRODUCTION

Paper-like flexible displays attract many attentions for electronic papers, electronic tag, and decorative displays [1]. Several liquid crystal (LC) technologies, for instance, polymer-dispersed liquid crystals (PDLC) [1–6], cholesteric liquid crystals [1,7–11], and single-substrate LCDs using photoenforced stratification [1,12–14] or using LC/polymer composites [15–18] have been demonstrated for making

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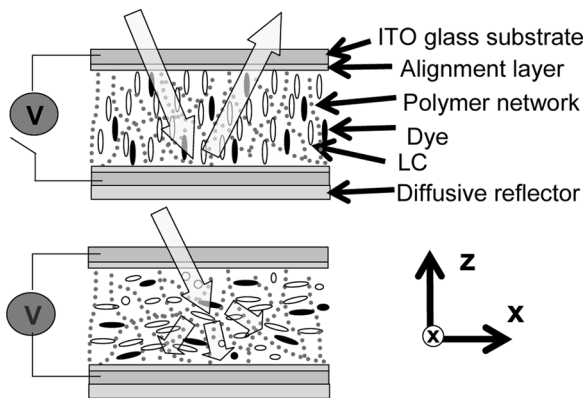
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flexible displays. Based on the combination of scattering and absorption we have recently developed polarizer-free LCDs using dye-doped dual frequency liquid crystal gels [19,20] and flexible dye-doped LC gels [21,22] with high contrast and fast response.

In this paper, we demonstrated a multi-switch display using gradient dye-doped LC gels which is still polarizer-free, fast response ( $\sim 10$  ms in general), and high contrast ( $\sim 200:1$  in general). The gradient dye-doped LC gel is patterned by controlling the fabrication process without patterned ITO layer. As a result, the density of polymer networks has spatial distribution. At  $0V_{\text{rms}}$ , the gradient dye-doped LC gel is bright and is dark at  $30V_{\text{rms}}$ . At  $9V_{\text{rms}}$ , it appears the colored pattern because of gradient polymer networks. The optical analysis of dye-doped LC gels is also discussed. The potential applications are flexible display and decorative displays.

## II. MECHANISM AND OPTICAL ANALYSIS OF POLARIZER-FREE DYE-DOPED LC GELS

The structure of polarizer-free dye-doped LC gels is schematically depicted in Figure 1. The structure consists of two ITO glass substrates whose inner surfaces were coated with a thin indium-tin-oxide (ITO) electrode and polyimide (PI) layer *without* rubbing treatment. The PI layer provides vertical alignment for the LC molecules. The materials we employed are negative liquid crystal (ZLI-4788, Merck), a diacrylate monomer with a dichroic dye S428 (Mitsui, Japan) at 90:5:5 wt% ratios. The sample preparation process is similar to the



**FIGURE 1** Schematic representation of the operating principle of dye-doped LC gels at voltage-off state and voltage-on state.

previous one [22]. The cell gap was  $5\ \mu\text{m}$ . After photo-polymerization, the formed chain-like polymer networks are mainly along the  $z$  direction because the LC directors are aligned perpendicular to the glass substrates during the UV curing process, as Figure 1 shows. At  $V=0$ , the cell does not scatter light and the absorption is rather weak. Therefore, the display has the highest reflectance. When we apply a high voltage at  $f=1\ \text{kHz}$  in the dye-doped LC gel, the liquid crystals and dye molecules are reoriented in the  $x$ - $y$  plane. The polymer network scatters light strongly. Since the alignment layer has no rubbing treatment, the absorption has no preferred direction; therefore, the light scattering and dye absorption efficiency reaches their maxima. As a result, the display appears black. The appearance of color is mainly because of the multiple light absorption of dye.

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

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

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

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

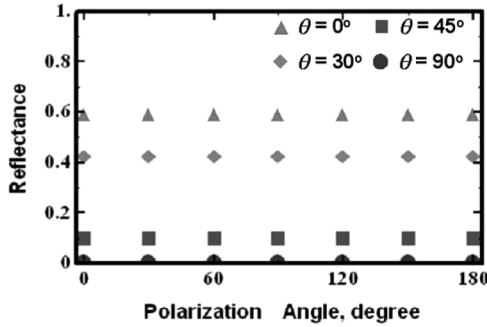
$$\beta_{\text{ave}}(\theta) = \rho_0 \cdot \frac{\sigma_{\text{eff}}(\theta)}{V} \quad (3)$$

where  $\rho_1$  is the dye concentration,  $\alpha_{\parallel}$  and  $\alpha_{\perp}$  are the absorption coefficients when the polarization of incident light is parallel or perpendicular to the principal axis of dye molecule.  $\rho_0$  is the LC concentration,  $V$  is the average volume of a droplet.  $\alpha_{\text{eff}}(\theta)$  can be expressed as:

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

We can estimate  $\alpha_{\parallel} = 11.83\ \mu\text{m}^{-1}$  and  $\alpha_{\perp} = 0.962\ \mu\text{m}^{-1}$  based on the experimental results.  $\sigma_{\text{eff}}$  in Eq. (3) is the effective scattering cross section from all liquid crystal droplets and can be expressed as:

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

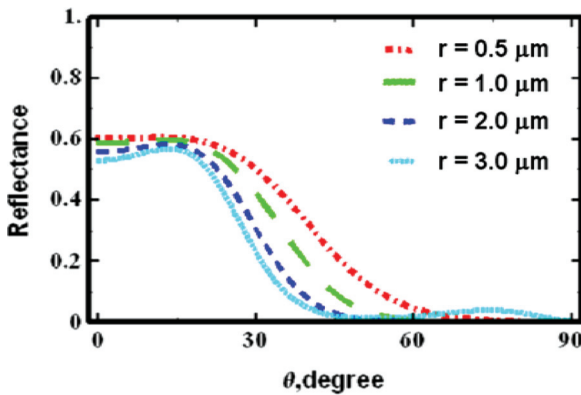


**FIGURE 2** Calculated reflectance as a function of polarization angle in dye-doped LC gels.

Based on anomalous diffraction approach [23,24], scattering cross section results from a single LC droplet is;

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

where  $\sigma_o$  is the geometrical optics cross section related to the domain size,  $\delta$  is the polarization angle;  $H_{ve}(\theta)$  and  $H_{vo}(\theta)$  stand for phase shift induced by e-ray and o-ray respectively. The simulation results are shown in Figures 2 and 3. In Figure 3, the reflectance decreases with the tilt angle. The R- $\theta$  curve shifts to right as the domain size is smaller. In Figure 2, the simulation results show our dye-doped LC gel is polarization independent. The simulation results agree with the experimental results [23]. The smaller domain size or larger



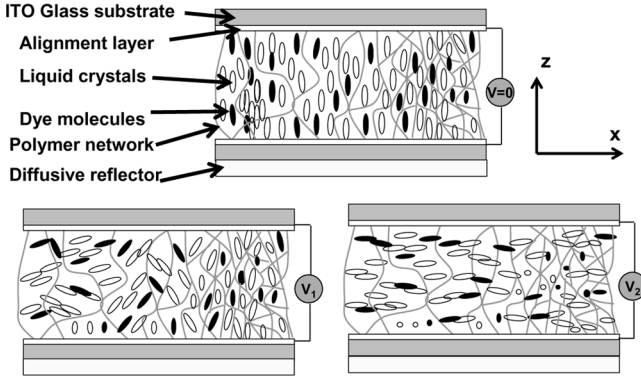
**FIGURE 3** Calculated reflectance as a function of tilt angle in dye-doped LC gels at different domain sizes.

density of polymer networks results in the larger operating voltage and better dark state. We can control UV intensity, curing temperature, the controlled temperature under UV illumination, and concentration of LC, dye or monomer to adjust the domain size [23].

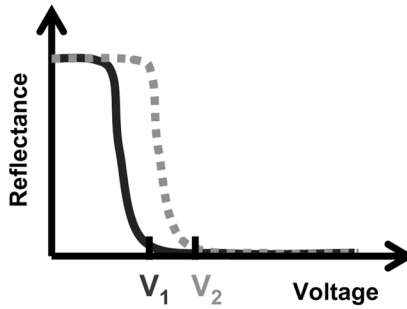
### III. SAMPLE PREPARATION AND MECHANISM OF POLARIZER-FREE GRADIENT DYE-DOPED LC GELS

The structure and mechanism of gradient dye-doped LC gels are schematically depicted in Figures 4(a) and 4(b). Similar to dye-doped LC gels, the structure consists of ITO glass substrates, vertical alignment layer without rubbing treatment, a diffusive reflector, negative liquid crystals, dye, and the gradient distribution of polymer networks. The corresponding voltage-dependent reflectance in low density and high density of polymer networks is also depicted in Figure 4(b). At  $V = 0$ , polymer networks, LC and dye are aligned vertically in all regions. The cell has high reflectance because the cell does not scatter light and the absorption is weak. When the applied voltage  $V_1$  is larger than the threshold voltage (at  $f = 1$  kHz) in the low density of the polymer networks of gradient dye-doped LC gels, the LC directors are reoriented by the electric field due to weaker anchoring energy while the LC directors remain vertically aligned in the region of a high density of polymer networks. As a result, reflectance decreases in the region of low density of polymer networks (solid line in Fig. 4b) due to the increases of both multi-domain scattering and dye absorption. Besides, it is polarization-independent in low density region because all the LC directors and dye molecules have the same tilt angle with random orientations which result from the alignment layer without rubbing treatment. The reflectance in low density region decreases while the reflectance is still high in high density region, as shown in Figure 4(b). When the applied voltage  $V_2 (> V_1)$  is high enough, the negative liquid crystals and dye molecules are reoriented randomly in the x-y plane in both low density and high density of polymer networks, as Figure 4(a) depicts. The polymer network scatters light strongly; therefore, both regions appear black because of the strong light scattering and dye absorption. It is still polarization-independent.

The fabrication process is similar to dye-doped LC gels. We use a photomask to control the UV intensity in UV illumination process and also control the temperature during photo-polymerization process in order to obtain the voltage-dependent reflectance, as shown in Figure 4(b). The UV curing intensity was  $1.37 \text{ mW/cm}^2$  at the temperature  $10^\circ\text{C}$  in high density region of polymer networks, and the UV curing intensity was  $0.73 \text{ mW/cm}^2$  at the temperature  $20^\circ\text{C}$



(a)



(b)

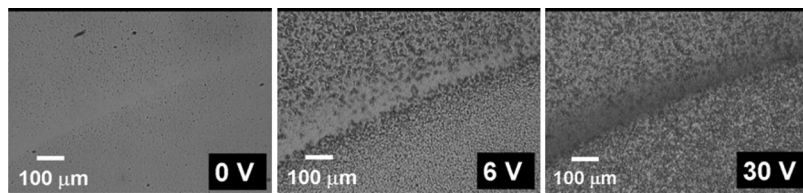
**FIGURE 4** Schematic structure of gradient dye-doped LC gels (a) at voltage-off state and voltage-on state. (b) The corresponding voltage-dependent reflectance at the low density of polymer networks (solid line) and the high density of polymer networks (dotted line).

in low density region of polymer networks. The domain size is small or the density of polymer networks is high under low curing intensity and high UV curing intensity. By arranging the density distribution of polymer networks, the threshold voltages are different in different density of polymer networks.

#### IV. EXPERIMENTAL RESULTS AND DISCUSSION OF POLARIZER-FREE GRADIENT DYE-DOPED LC GELS

Figures 5(a)–(c) show the morphologies observing under an optical microscope with a single polarizer only. The upper regions in



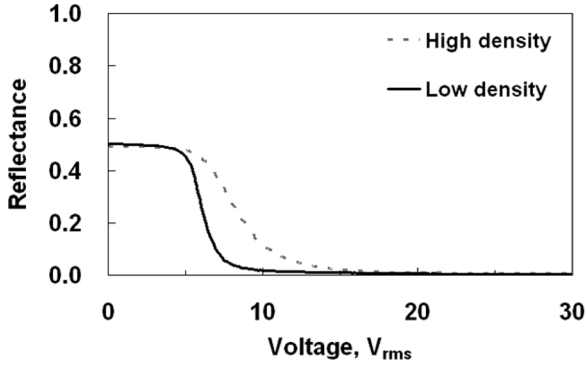


**FIGURE 5** The morphologies of gradient dye-doped LC gels at (a) 0, (b) 10, and (c)  $30 V_{\text{rms}}$ .

Figure 5(a)–(c) are the low density of polymer networks and the lower regions are the high density of polymer networks. At  $0 V_{\text{rms}}$ , the gradient dye-doped LC gel is bright due to the vertically aligned LC directors, dye and polymer networks. At  $6 V_{\text{rms}}$ , the upper region turns out darker than the lower region due to the lower threshold voltage in low density region. At  $30 V_{\text{rms}}$ , both regions show the dark red color and different domain textures of the polymer networks because of dye molecules.

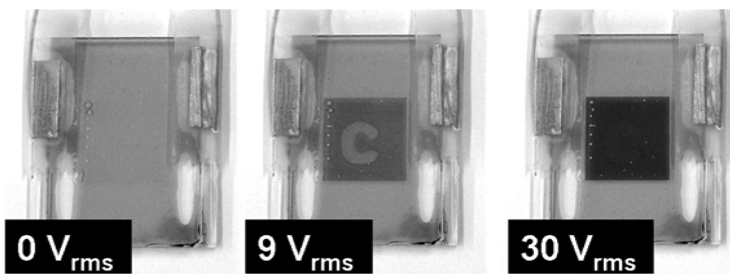
We use the laser-based reflectance measurement in order to measure the electro-optical properties of dye-doped LC gels. Because the guest-host system we employed appears dark red rather than black, we used an unpolarized green He-Ne laser ( $\lambda = 543.5 \text{ nm}$ , Melles Griot, Model 05-LGR-173) instead of a white light source. 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  $\sim 25 \text{ cm}$  (the 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.

Figure 6 is the voltage-dependant reflectance of the gradient dye-doped LC gels in the low density of polymer networks (solid line) and in the high density of polymer networks (dotted line). The reflectance was normalized to that of a pure LC cell with the same cell gap. The reflectance in both regions decreases with the applied voltage  $V > V_{\text{th}}$  due to the increase of the scattering and the absorption. The reflectance at  $0 V_{\text{rms}}$  is  $\sim 50.2\%$  in the low density region and is  $\sim 49.2\%$  in the high density region. The threshold voltage in low density region is around  $4.62 V_{\text{rms}}$  and is around  $5.42 V_{\text{rms}}$  in high density region. The contrast ratio (CR) is defined as a reflectance ratio of  $0 V_{\text{rms}}$  to  $30 V_{\text{rms}}$ . Then the CRs in low and high density regions are  $\sim 219:1$  and  $166:1$ , respectively. The response times in low and high density regions are 10.6 and 8.6 ms, respectively. The higher density



**FIGURE 6** Voltage-dependent reflectance in the low density region (solid line) and high density region (dotted line).

of polymer networks results in larger threshold voltage and faster response because of the strong anchoring energy of polymer networks. In order to operate the gradient dye-doped LC gels as a multiple switch, we demonstrated a character “C” by using a photomask with a “C” pattern as an example, as shown in Figure 7. “C” is in the region of the high density of polymer networks and out of the “C” is low density of polymer networks. The cell gap was still  $5\ \mu\text{m}$ . To avoid specular reflection, we laminated a diffusive reflector, a white paper, on the backside of the bottom glass substrate. The ambient white light was used to illuminate the samples. The gradient dye-doped LC gel is bright without an applied voltage because of the vertically aligned polymer networks, LC and dye. At  $9V_{rms}$ , outside “C” region is dark while inside region is still bright. That is because the reorientation of LC directors and dye in the low density of polymer networks. The whole cell appears dark at  $30V_{rms}$  when the all LC directors



**FIGURE 7** A multi-switch of the  $5\text{-}\mu\text{m}$  gradient dye-doped LC gels at  $0V_{rms}$ ,  $9V_{rms}$  and  $30V_{rms}$ .

and dye are along x-y plane in Figure 4(a). It does not require any polarizer. Therefore, the polarizer-free gradient dye-doped LC gel can be used as a multi-switch while maintaining the patterned information. The multi-switch using polarizer-free gradient dye-doped LC gels has several advantages. Fabrication process is simple without patterned ITO and the driving method is simple without complex IC. It can be colorful by changing the dye molecules. However, the drawbacks are high driving voltage, low contrast ratio. This technique can be further applied for making a flexible segmented multi-switch.

## V. CONCLUSION

We have demonstrated a multi-switch display using polarizer-free gradient dye-doped LC gels. By arranging the density of polymer networks, the switch can show three operating states. The CR is around 200:1 and the response time is  $\sim 10$  ms in general under laser-based measurement. The reflectance reaches  $\sim 50\%$ . This approach can be further extended to a multi-switch with more than 3 states. The potential applications of the polarizer-free gradient dye-doped LC gel are for decorative displays and flexible displays.

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