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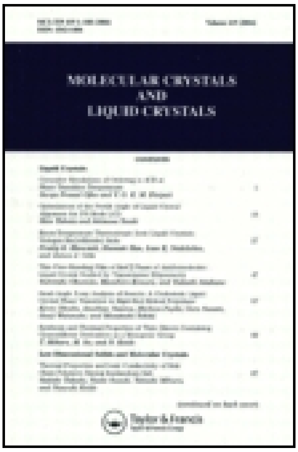

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Electrically Induced Multicolored Hyper-Reflection and Bistable Switching from a Polymer-Dispersed Cholesteric Liquid Crystal and a Templated Helical Polymer

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Electrically Induced Multicolored Hyper-Reflection and Bistable Switching from a Polymer-Dispersed Cholesteric Liquid Crystal and a Templated Helical Polymer

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In this study, a special bistable polymer-dispersed cholesteric liquid crystal (PDCLC) with two different reflection modes was fabricated by refilling a PDCLCs composites into a template helical polymer with the same or opposite helical structure, in which the templated helical polymer with a half of the cell thickness was fabricated by nonuniform photopolymerization of monomers induced by ultraviolet light absorbing dye in the cholesteric liquid crystal (Ch-LCs) mixture. Modulation of a hyper-reflective state (>50% reflection) and normal-reflective state (≤50% reflection) at a fixed wavelength, even switching between bicolor reflection with different handedness can be achieved. Additionally, a patterned multicolored switchable Ch-LCs film can be achieved by infiltrating the above PDCLCs into a patterned templated helical polymer, and the switching behavior of the patterned film was exhibited. These special optical properties make the novel Ch-LCs composites an interest for great potential application in many fields such as flexible display, optical components, LCs lasing, etc.

Keywords: Bistable switching; multicolored hyper-reflection; polymer-dispersed cholesteric liquid crystal; templated helical polymer

Introduction

Cholesteric liquid crystals (Ch-LCs) are well known for vivid colors due to a macroscopic helical structure, which makes them promising in reflective display and switchable optical elements. The reflected wavelength of Ch-LCs is generated by the photonic band gap (PBG) from the periodical cholesteric helical orientation, in which the pitch p is defined as the distance in which Ch-LCs molecules rotate 360° along its helical, and for light propagating along the helical axes, $\lambda_0 = p \times n$, where λ_0 is the wavelength of the maximum reflection or the middle of selective reflection band and n is the average refractive index $n = (ne + no)/2$. The extraordinary and ordinary indices of refraction are denoted by ne and no, respectively. The maximum reflection of unpolarized light is 50% because only circularly

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*Address correspondence to Jinbao Guo and Jie Wei, College of Materials Science and Engineering, Beijing University of Chemical and Technology, Beijing 100029, P. R. China. E-mail: guojb@mail.buct.edu.cn; weij@mail.buct.edu.cn polarized light with the same handedness as that of the Ch-LCs will be reflected, while the component with opposite handedness will be transmitted [1,2].

A high brightness color or higher contrast is needed in display and photonic fields, many approaches to achieve a high contrast in Ch-LCs have been investigated until now [3–29]. A simple method is to associate two Ch-LCs cells with the same mean reflection wavelength but opposite helicity senses to increase the reflected light intensity [5–7]. Additionally, a structure comprising a half-wave plate sandwiched between polymeric Ch-LCs films with the same handedness was demonstrated to realize the reflectance in PBG exceeds 50% [9–13]. However, these approaches have some limitations such as complicated structure design, control of optical defects and losses at the interfaces. In order to overcome these drawbacks, some new approaches have been reported to exceed 50% reflectance limit in a single Ch-LCs through the use of polymer stabilization or polymer template. Mitov et al. demonstrated a method of fabricating a single-layer Ch-LCs gel exceeding 50% reflectance limit [14–18]. In the work, photopolymerizable monomers were introduced into a Ch-LCs with a thermally induced inversion of the helicity sense, and the composites were polymerized by UV light when the helix is right-handed, resulting that a polymer stabilized Ch-LCs (PSCLCs) or a Ch-LCs gel. The reflectance exceeds 50% when measured at the temperature assigned for a cholesteric helix with the same pitch but a left-handed sense before photopolymerization. We utilized a helical polymer structure to fabricate a double handedness structure Ch-LCs composites exceeding 50% reflectance limit [21–25], in which the helical polymer network was originated from memory effect of the initial PSCLCs [19,20]. Recently, Bunning's group has developed a method to achieve near 100% reflectivity in a single Ch-LCs film, in which a spatial segregation of the right-handed and left-handed (LH and RH) domains occurs in the z-direction by surface-bounded polymer stabilization (through the thickness of the cell). Photo-induced or thermally induced hyper-reflective Ch-LCs can be achieved by backfilling the above templated helical polymer with a photoresponsive Ch-LCs mixture or a thermally tunable SmA* mixture [27,28].

As is well known that bistable-reflective Ch-LCs display is an attractive technology due to its wide range applications, many researchers have focused on increasing reflective brightness and achieving a multicolor even a full-color display through stacking technique [30–34]. However, this approach has a number of issues that it needs to overcome, such as structure design and pixel addressing. In this study, we use the template helical polymer to achieve a special bistable polymer-dispersed Ch-LCs (PDCLCs) with hyper-reflection state or multicolor reflection state. Herein, the templated helical polymer was derived from the nonuniform distribution of the polymer network from photo-polymerization of LC diacrylates in Ch-LCs, which, induced by an ultraviolet light absorbing dye (ULAD), absorbs UV light and produces a gradient in the intensity of UV light in the cell. Tuning of bicolor reflection with different handedness, and of between a hyper-reflective state (>50% reflection) and normal-reflective state ($\leq$50% reflection) at a fixed wavelength can be realized. Additionally, a patterned multicolor switchable film can be achieved by infiltrating the PDCLCs into a patterned templated helical polymer, and the switching behavior of the patterned film was exhibited.

Experimental

Materials

In this study, nonreactive LCs, SLC-1717 (20°C, 589 nm, $\Delta n = 0.201$; Slichem Liquid Crystal Material Co. Ltd., Shijiazhuang, China); Nematic diacrylate monomer, C6M,

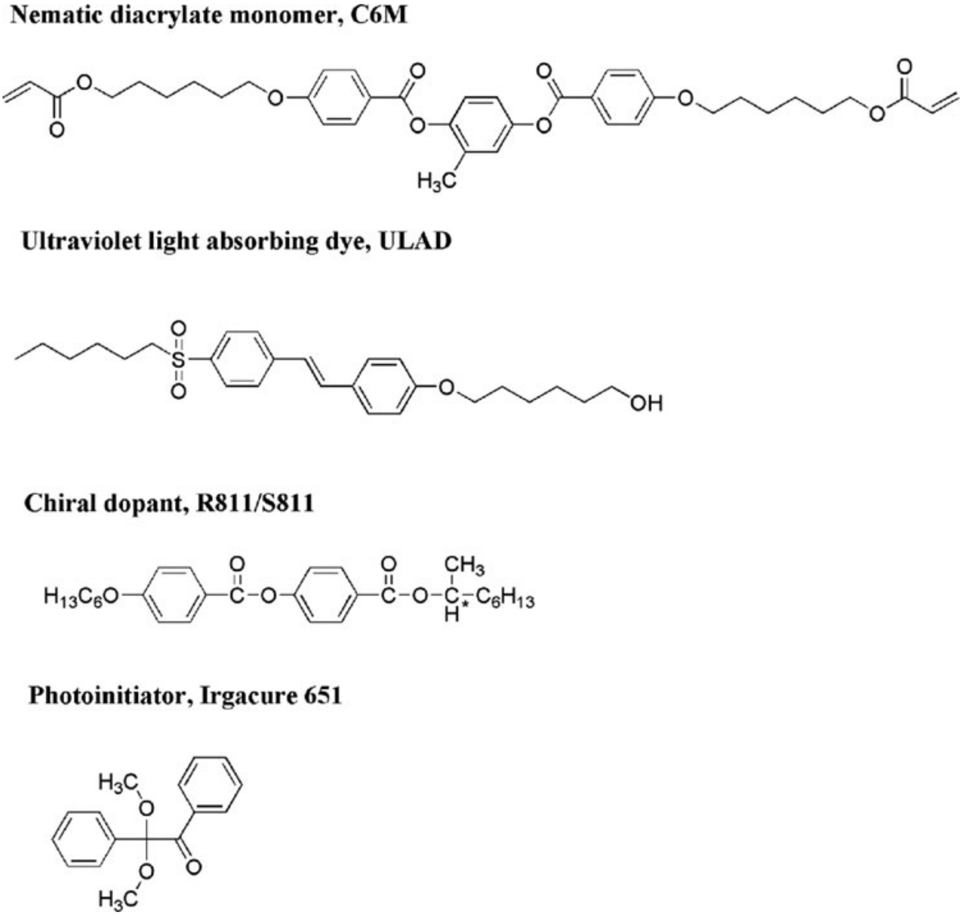

Figure 1. The chemical structures of the materials used in this study.

LC242 and LC1057; Photocurable monomer (Norland Optical Adhesives Inc., Cranbury, NJ, USA) NOA-65; Chiral dopant, S811/R811 (Merck Co. Ltd., Whitehouse Station, NJ, USA) and photoinitiator, 2, 2-dimethoxy-2-phenyl-acetophenone (Irgacure 651, TCI Co. Ltd., Nei Hu,Taipei, Taiwan); and ULAD, 2-{2, 6-bis-[2-(4-dimethylamino-phenyl)-vinyl]-pyran-4-ylidene}-malononitrile were used. Nematic diacrylate monomer, C6M was synthesized according to the method suggested by D. J. Broer et al. [35], LC242, and LC1057 were purchased from BASF Co. Ltd. (Mumbai, Maharashtra, India), ULAD was lab-synthesized according to the earlier paper [36]. Figure 1 shows the chemical structures of C6M, S811/R811, ULAD and Irgacure 651.

Experimental Cells

In order to induce a planar orientation of LC molecules, the inner surfaces of indium tin oxide coated (ITO) glass cells were coated with a 3.0 wt% polyvinyl alcohol (PVA) aqueous solution. The deposited film was dried at 80°C for 30 min and subsequently rubbed with a textile cloth under a pressure of 2.0 g/cm^2 along one direction. PET (polyethylene terephthalate) film of 12 μm thickness was used as the cell spacers. The samples were

Table 1. Compositions of Ch-LCs mixture and reflection band of the cells obtained

Mixture	Weight ratio/wt%[1]	λ_M/nm[2] Before	λ_M/nm[2] After
1	SLC-1717 (66.8)/C6M (15.0)/R811 (17.0)/ULAD (1.2)	718	742
2	SLC-1717 (62.3)/C6M(15.0)/R811 (21.5)/ ULAD (1.2)	578	552
3	SLC-1717 (58.8)/C6M(15.0)/R811 (25.0)/ ULAD (1.2)	514	476
4	SLC-1717 (66.25)/S811 (18.75)/NOA65 (15.0)	—	548
5	SLC-1717 (66.8)/C6M (15.0)/S811 (17.0) /ULAD (1.2)	728	756
Cell A[3]	mixture 1 + (4)		
Cell B	mixture 2 + (4)		
Cell C	mixture 5 + (4)		
Cell D	mixture 2 + 3 + (4)		

Notes: [1] 2% by the weight of monomers of Irgacure 651 is added.

[2] Reflection band before and after UV irradiation.

[3] Cell A was obtained by infiltrating mixture 4 with LHHS into the helical polymer template from mixture 1 followed by UV irradiation.

filled into the cells by capillary action at appropriate temperature. The compositions of the samples are listed in Table 1.

Templated Helical Polymer

The procedure for fabricating polymer template was carried out as follows: first, the cell containing the sample was irradiated with 365 nm UV light (0.15 mW/cm^2) for 30 min to initiate polymerization as shown in Fig. 2(a), in which the sample was composed of C6M/SLC-1717/Chiral dopant/ULAD/photo-initiator with the periodicity or pitch (*P*) defined by the Ch-LCs. Following that, the cell was immersed in cyclohexane for about 48 h and later in tetrahydrofuran for 20 min to remove the nonreactive LCs. After that the cell was kept in vacuum chamber at 60°C for about 3 h. Thus the polymer template was obtained as shown in Fig. 2(b). And then, the cell containing the polymer template was refilled with samples with different kind of Ch-LCs by capillary filling process and followed by other process such as second irradiation as shown in Figs. 2(c) and (c').

Patterned Multicolored Switchable Ch-LCs Films

The patterned multicolored switchable Ch-LCs films were prepared by carrying out the following procedure. For one-side patterned film, a photomask composed of simple grid structure was positioned on top of a filled sample cell. Herein, the samples also include C6M/SLC-1717/Chiral dopant/ULAD/photo-initiator. Then, the cell was illuminated for 30 min at certain temperature with a UV light (365 nm, 0.15 mW/cm^2). After this period, the cell was immersed in solvent to remove all components of unexposured regions and nonreactive components of exposured regions, resulting that the polymer scaffold only existed in the exposured areas. Following that, the cell with the patterned polymer scaffold was refilled with PDCLCs sample by capillary filling process and followed by a second irradiation with a UV light (365 nm, 5.0 mW/cm^2) for 30 min. To create a two-side patterned film, two cells containing C6M/SLC-1717/Chiral dopant/ULAD/photo-initiator

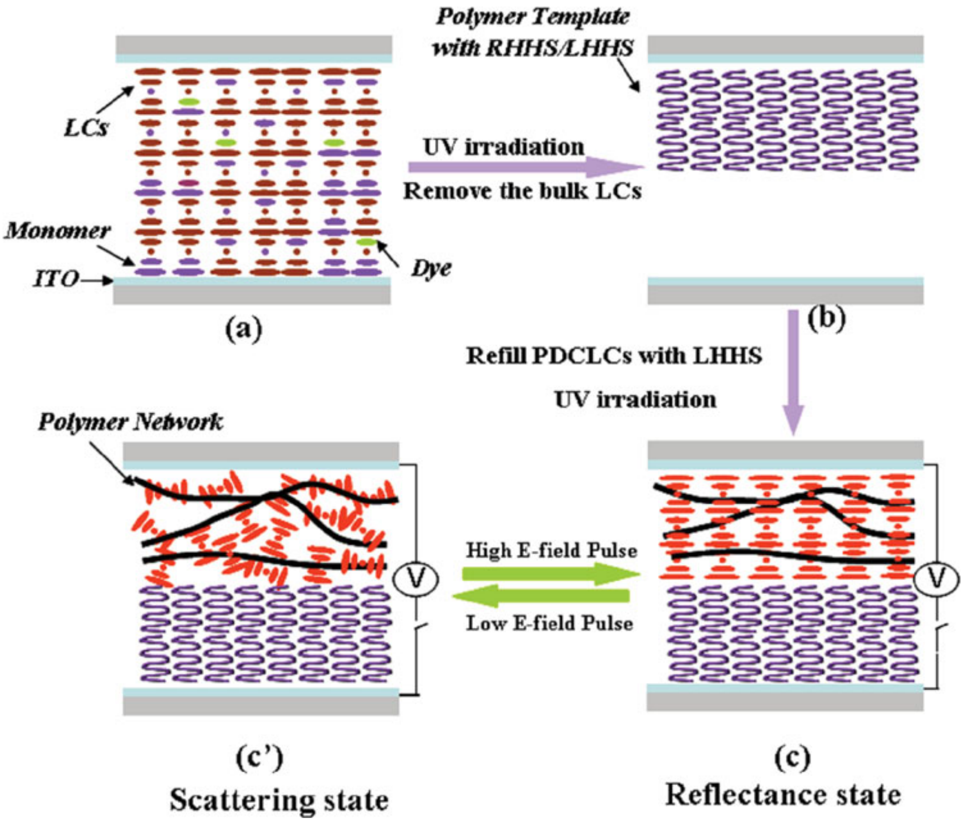

Figure 2. Schematic representation of the fabrication process of Ch-LCs film.

with different pitch were irradiated by a UV light (365 nm, 0.15 mW/cm^2) under a mask composed of simple grid structure. After the above wash-out process, the patterned cells were split open and glued together with staggered stacking structure. And the cell was subsequently capillary filled with PDCLCs sample and the latter procedure was similar as above.

Measurements

The samples were observed using a polarizing light microscope (POM) (DM2500P; Leica Microsystems CMS GmbH, Wetzlar, Hesse, Germany) with a heating stage (LTS 420). The optical images were recorded using Linksys 2.43 software. The transmission spectra were obtained by UV/vis spectrophotometer (U-3010; Hitachi Ltd., Chiyoda, Tokyo, Japan,) at normal incidence. The transmittance of a blank cell was normalized to 100%. In general, λ_M and $\Delta\lambda$ are defined as the minimum wavelength of the transmitted light and the bandwidth at half-height of the peak, respectively. The reflection spectra were obtained by Fiber spectrometer (AvaSpec-2048; Avantes BV, Apeldoorn, Gelderland, The Netherlands). The electric field is applied to the cells using a signal generator (HT102V4; Halation Technology Co. Ltd., Beijing, China). The morphology of the polymer network was observed by scanning electron microscopy (SEM) (Hitachi S-4700). The samples for SEM studies were prepared according to the method described in the previous study [37,38].

Results and Discussion

A detailed schematic of the preparation and subsequent mechanism for tuning reflection color is shown in Fig. 2. Due to the addition of a small amount of ULAD (1.2 wt.%)—which has an absorption maximum at 334 nm, close to that of the photoinitiator (Irgacure 651, Ciba, 345 nm), and an extinction coefficient two orders larger than that of Irgacure 651—a gradient in UV intensity could be achieved over the thickness of the film [39,40]. The polymerization rate of C6M will be fastest at the top of the film (lamp-side) resulting in a faster consumption of C6M. The depletion of this monomer starts a diffusion process, in which the upper part of the layer is enriched with polymer network originated from photopolymerization of C6M, and the lower part with nonreactive LCs, as also suggested by Mitov et al. [16]. After the wash-out process, a polymer template with right-handed handedness structure (RHHS) tethered to the upper surface of the cell was achieved as shown in Fig. 2(b). Following that, we introduce the precuring PDCLCs into the above template helical polymer, resulting that a special bistable PDCLCs with two different reflection modes is achieved (Figs. 2(c) and (c')). Figure 3 shows SEM image of the cross-section of the cell A. It is obvious that the cell possesses two distinct regions, it means that

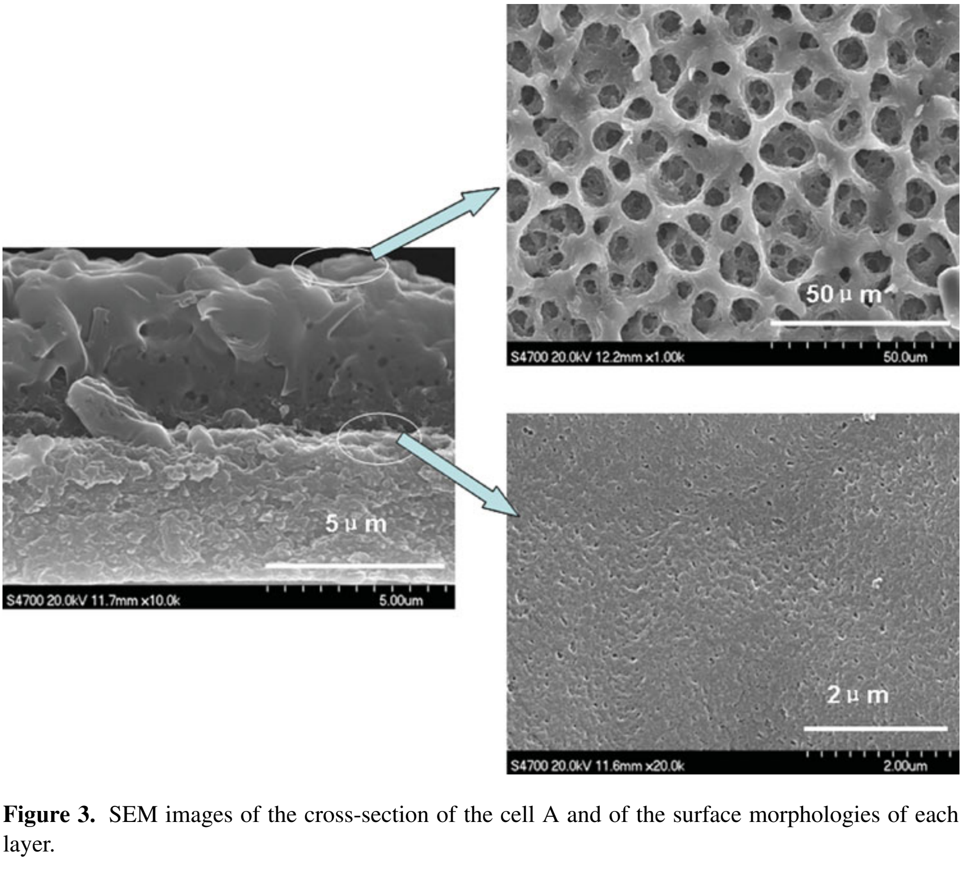

Figure 3. SEM images of the cross-section of the cell A and of the surface morphologies of each layer.

a spatial segregation structure of RH and LH domains occurring through the thickness of the cell was fabricated. Herein, the bottom layer is the RH helical polymer scaffold, the thickness of which was measured to be about 6 μm with a half of the cell thickness of 12 μm. While the upper layer is PDCLCs section with LH helical structure formed from the mixture 4. Moreover, the surface morphologies of the two layer in the cell A were also given, the upper layer has a typical morphology of PDCLCs, in which the spherical droplet with 10 μm in diameter on the thickness of the cell can be seen. While the helical superstructure of the polymer template formed in Ch-LCs can clearly be imaged from the surface morphology of the bottom layer [37,38].

It is well known that the bistable Ch-LCs display has two stable optical contrasting states at zero field: one state reflects colored light and the other state is weakly scattering. The reflecting and scattering states correspond to the planar and focal conic textures, respectively [34]. The mixture 4—composed of SLC-1717, S811, NOA65, and IRG 651, as listed in Table 1—was used to fabricate PDCLCs cell; the reflection spectrum of the planar reflecting state and the scattering state is shown in Fig. 4(a). It can be seen that the reflection band is at about 550 nm, and the reflectivity intensity can about approach 30% as compared to a standard white diffuser. The photos of the corresponding bistable state of PDCLCs were recorded as shown in the insets of Fig. 4(a), high contrast of the reflecting state and weak scattering state can be seen. Meanwhile, the response of the PDCLC to voltage pulses is shown in Fig. 3(b). The pulse width was 60 ms and the reflectance was measured 2s after the pulse. If the cell was initially in the planar texture, it remained in the planar texture for voltage pulses with amplitude less than the threshold V1 = 5 V, and completely switched into focal conic texture by the pulse if its amplitude was above V2 = 13 V. If the cell was initially in the focal conic texture, it remained there until the applied voltage was increased above V3 = 40 V. When the voltage was above V4 = 46.5 V, the material was switched to the homeotropic texture during the pulse and relaxed to the planar texture afterward.

Figure 5(a) shows the optical response of the cell A to E-field pulses. We can observe the cell A has two reflection bands at the initial state, in which red-colored R-CPL and green-colored L-CPL from the upper PDCLCs were reflected by the cell A. When the E-field pulse reaches $V2_A = 13.5$ V, the reflection band at 550 nm of the cell A corresponding to green-colored L-CPL disappears and the reflection band at 650 nm of the cell A corresponding to red-colored R-CPL still maintains. While another E-field pulse of $V4_A = 47$ V is applied to the cell A, the reflection band at 550 nm reappeared. Moreover, these two states can also keep even if the E-fields are removed. This is because that transition of the bistable states between reflection state and weak scattering state of the upper PDCLCs layer happens in the cell A when an E-field pulse is applied. The insets are the corresponding POM micrographs of the cell A at the initial reflection state and the finial scattering state, and we can see the planar texture and the focal conic texture of the cell A at these two states are obvious. Additionally, switching of a hyper-reflective state (>50% reflection) and normal-reflective state ($\leq$50% reflection) at a fixed wavelength can be realized. Herein, the polymer template was prepared from the mixture 2 with RLLS, in which the reflection band was at about 552 nm. Figure 5(b) shows the optical response of the cell B to E-field pulse, it is obvious that the cell B at the initial state can exceed 50% reflectance at 550 nm, which means that both green-colored L-CPL and R-CPL within the reflection band are reflected. When a pulse of $V2_B = 13.4$ V was applied to the cell B, the reflectance intensity of the reflection band at 550 nm decreased, which is due to that the upper PDCLC of the cell B was driven to the weak scattering state during the pulse, resulting that only green-colored R-CPL of the reflection band at 550 nm was reflected by the cell B. From the above discussions,

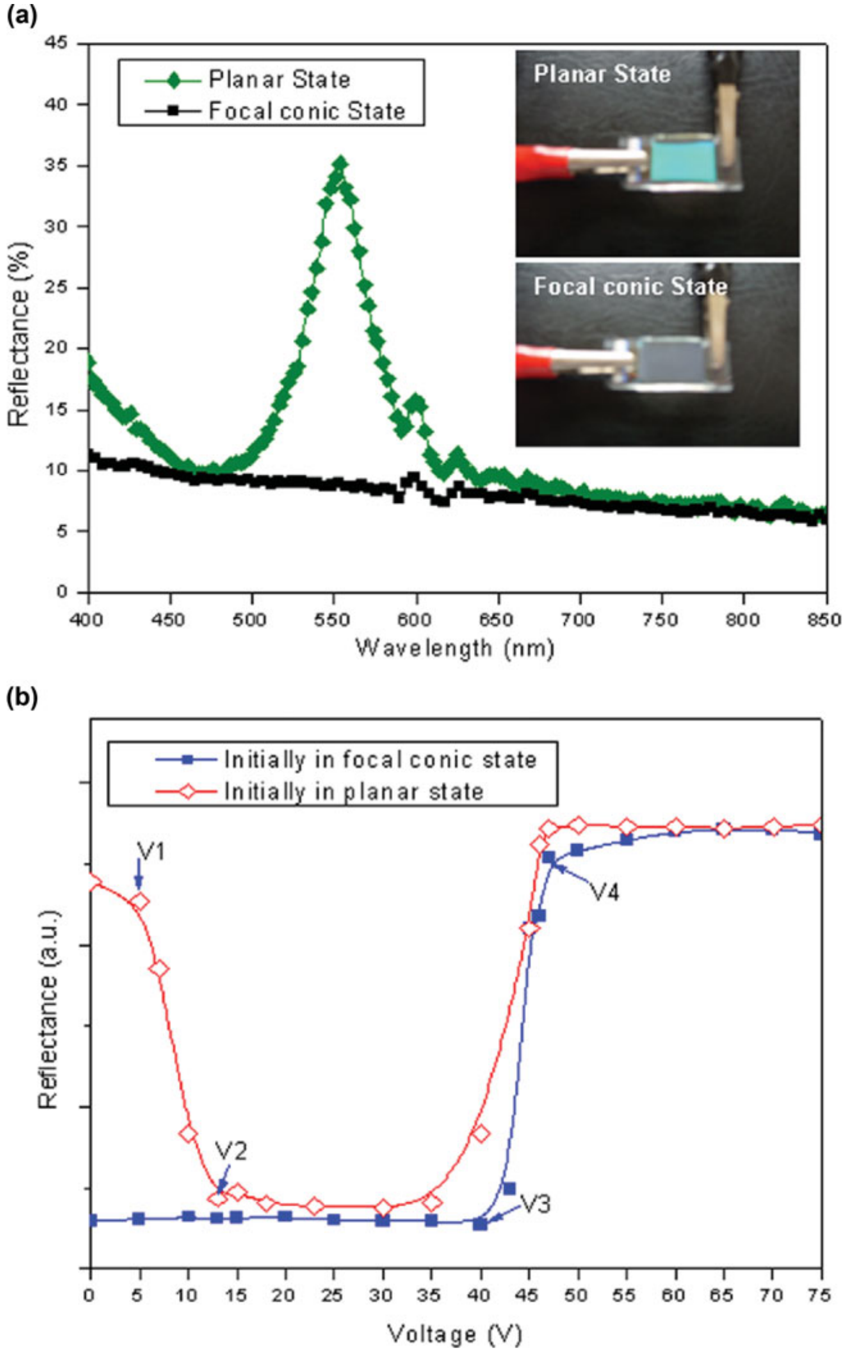

Figure 4. (a) Reflection spectra of PDCLCs at the reflecting state and at the scattering state; the insets are the photographs of the PDCLCs taken at these two states. (b) Response of the PDCLC to voltage pulses.

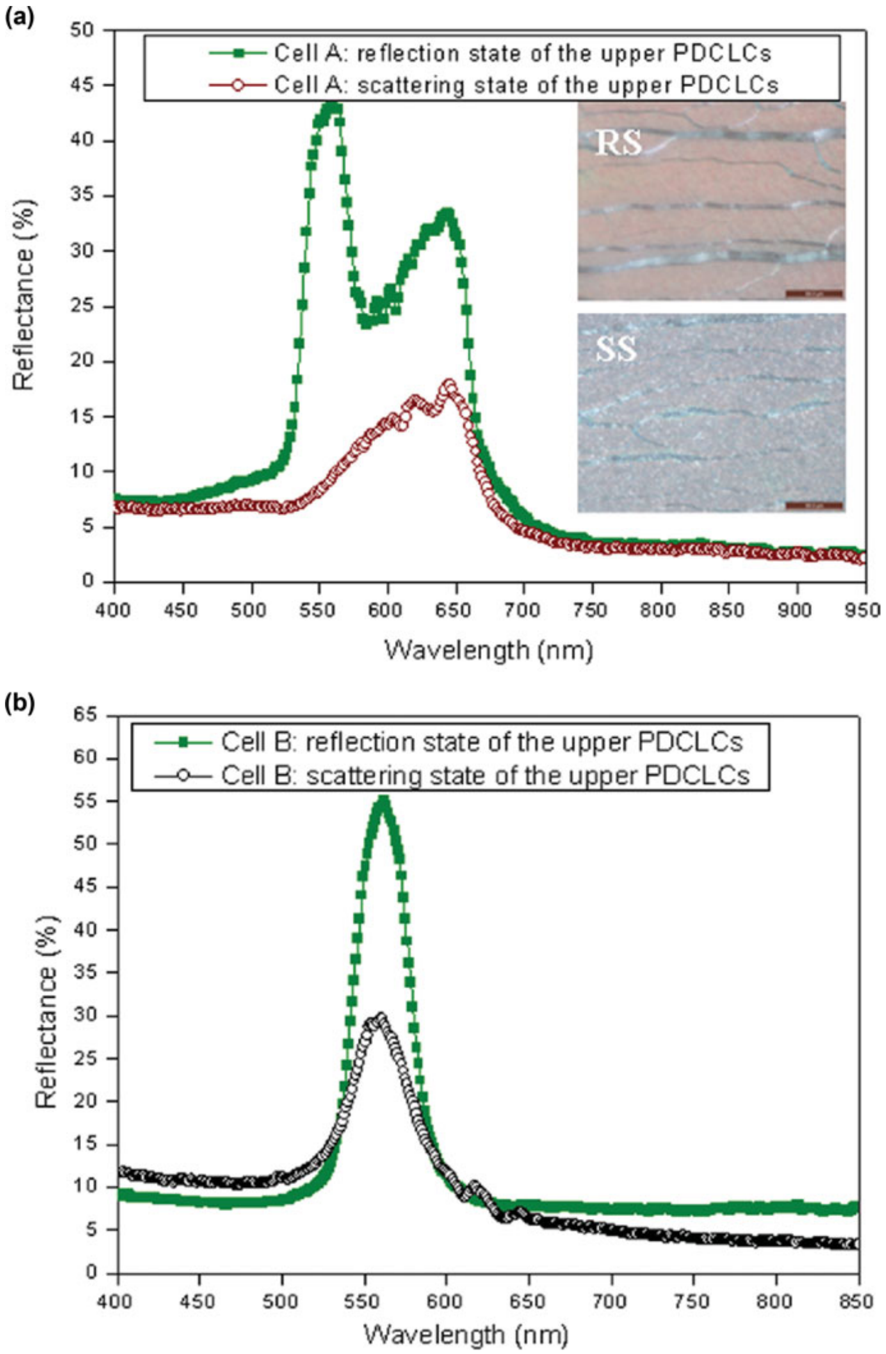

Figure 5. (a) Reflection spectra of cell A in the reflecting state and in the scattering state; the insets are the POM micrographs of the textures in the reflecting and scattering state of the cell A. (b) Reflection spectra of cell B in the reflecting state and in the scattering state.

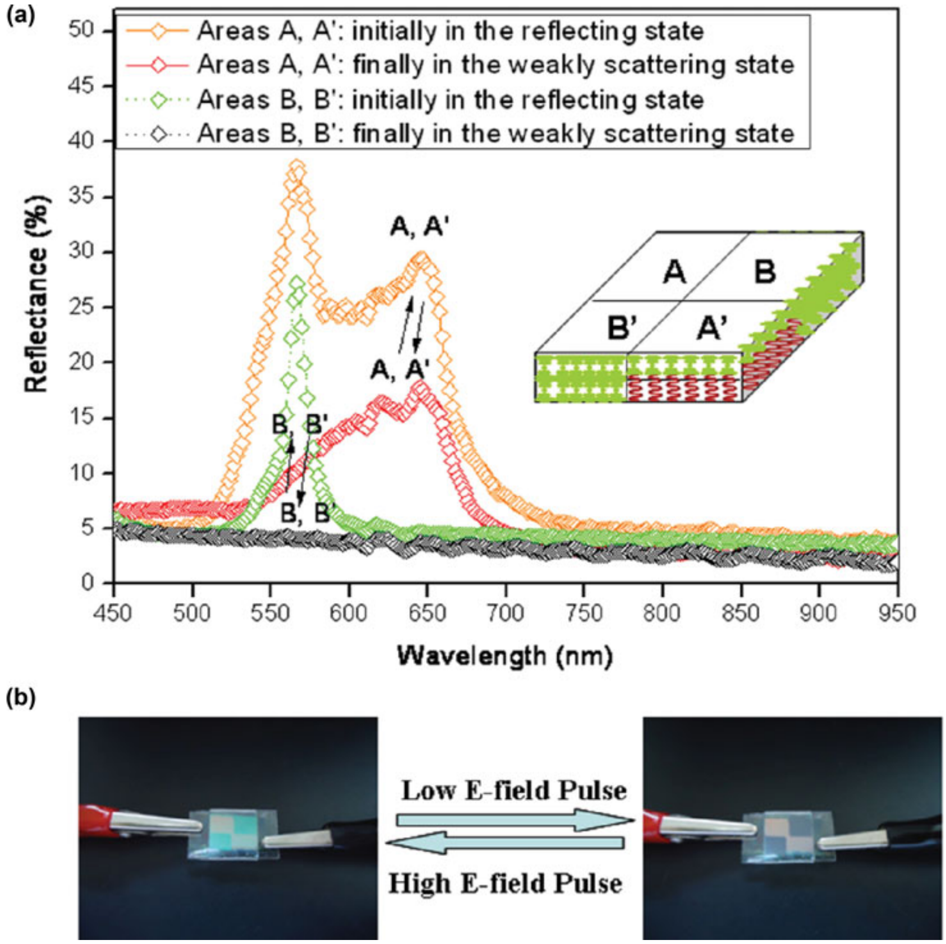

Figure 6. (a) Reflection spectrum of cell C obtained by the photomask, the inset is illustration of the three-dimensionally simplified mode of the cell C. (b) The images of the cell C at bistable states.

it is clear that the reflection color and reflection intensity of the special PDCLCs can be switched under E-field pulses based on the templated polymer scaffold.

In this study, we extend this technological method to the fabrication of a polymer template with a simple patterned square of exposured and unexposured regions by photolithographic techniques. To achieve this purpose, mixtures 4 and 5, as shown in Table 1, were used to fabricate cell C with a multicolored reflection mode, in which the helical structure of Ch-LCs mixtures 4 and 5 were both left-handed, and mixture 4 was refilled into the patterned polymer template from the mixture 5 as described above. Figure 6(a) shows the optical response of the cell C to E-field pulses. For areas A and A’ of the coexisted regions of polymer template and PDCLCs, the cell was found to exhibit a yellow Bragg reflection, which is attributed the overlapping of red-colored reflection of the polymer template and green-colored reflection of the PDCLCs. In contrast, for the areas B and B’ of nonpolymer templated areas, which consisted of only the PDCLCs, they only did exhibit green-colored Bragg reflection. When a pulse of $V2_C = 13.6$ V was applied to the cell C, the PDCLCs is switched to the focal conic texture after the pulse, areas A and A’ exhibit a green Bragg

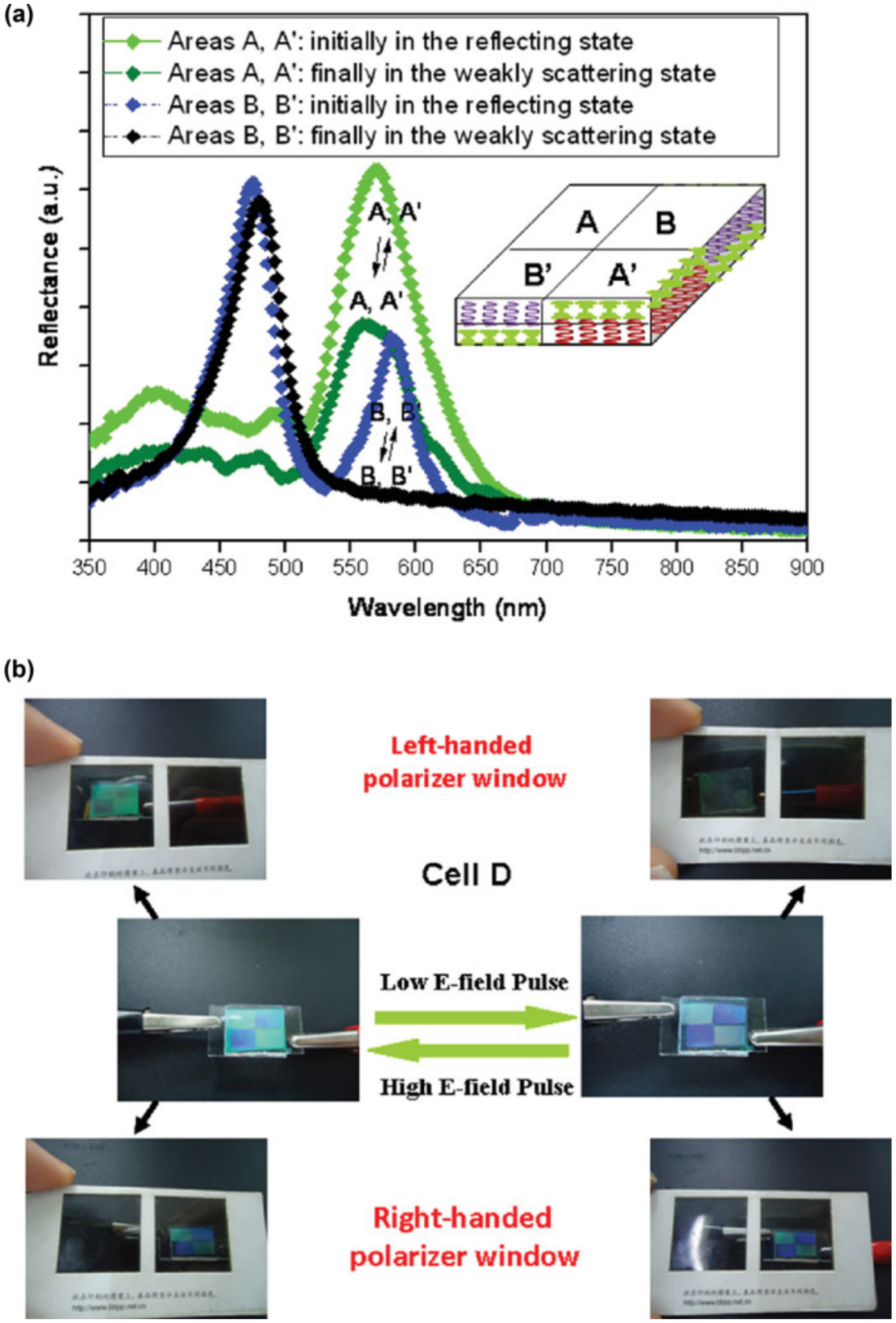

Figure 7. (a) Reflection spectrum of cell D obtained by the photomask, the inset is illustration of the three-dimensionally simplified mode of the cell D. (b) The images of the cell D observed at bistable states under a polarizer.

reflection, while areas B and B' did not exhibit any Bragg reflection. The inset in Fig. 6(a) gives the three-dimensionally simplified mode of the cell C at the initial reflecting planar state, and we can see the layer structure and Ch-LCs molecule arrangement of the cell C at this state. Figure 6(b) shows the photos of the corresponding bistable states of the cell C, a switching mode between different states can be seen clearly.

The versatility of the polymer template approach is further demonstrated in cell D, fabricated with patterned grid film on both sides, as schematically shown in Fig. 7. This cell was fabricated by preparing two-patterned cells in the same photo-lithographic technique as described above, but the polymer template exhibited different helical structure, which derives from mixtures 2 and 3. The cell D with two polymer template regions in a staggered stacking fashion was obtained (inset in Fig. 7(a), regions depicted with red and purple helical network) and was backfilled with the PDCLCs of an untemplated region of the half thickness of the cell (inset in Fig. 7(a), region depicted with green helical pitch) in a manner similar to that described above. Figure 7(a) shows the optical response of different areas of the cell D to E-field pulses. It is obvious that, one mode of a hyper-reflective state (>50% reflection) and normal-reflective state ($\leq$50% reflection) in 565 nm at areas A and A', and another mode of bicolor reflection with different handedness (blue-colored R-CPL and green-colored L-CPL) at areas B and B', can be achieved in the cell D. The images of the cell D, both at the initial state and the finial state, under a polarizer were shown in Fig. 7(b). The polarizer used here has two windows for both left-handed polarized light and right-handed polarized light, which is prepared by stacking multilayer PVA films on TAC substrate and followed by dyeing and uniaxial tension process [22]. It can be seen that the cell D exhibits different reflection modes within left-handed polarizer and right-handed polarizer window at the initial state and the finial state, respectively, which is in accordance with the above spectrum characteristics. It should be pointed out that full-color Ch-LCs display can be realized by changing the photomask pattern and designing the double-electrode, especially regarding to the influence of handedness chirality on the reflection intensity.

Conclusions

In summary, a template helical polymer with a half the cell thickness was employed to tune reflection intensity/color from a hybrid structure of a PDCLCs within it, resulting that a special bistable PDCLCs with two different reflection modes, modulation of bicolor reflection with different handedness, and switching between a hyper-reflective state (>50% reflection) and normal-reflective state ($\leq$50% reflection) at a certain wavelength can be achieved. Additionally, patterned multicolored switchable films can be fabricated and tuning mode of the patterned film was exhibited. These results are of promise for a number of possible LCs device applications such as flexible flat display, tuneable lasers, and optical components.

Acknowledgments

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