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Electrically Switchable, Hyper-Reflective Blue Phase Liquid Crystals Films

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Blue phase liquid crystals (BPLCs) with three-dimensional periodicity have attracted significant attention due to their fast response time in the submillisecond region, which is critical in sequential liquid crystal displays and fast switchable photonic devices. However, the reflectivity of single-layer BPLC films in the visible range, which is generally used for reflective display, is still low and limited to 50%, which hinders its use in a wide range of applications. Herein, hyper-reflective, electrically switchable, fast responsive, and colorful reflective displays are demonstrated, which are based on multi-layer BPLC films consisting of two single-layer BPLC templates with opposite handedness. Hyper-reflectivities of 89%, 82%, and 68% in the red, green, and blue color regions are achieved by refilling multi-layer BPLC templates with achiral nematic liquid crystals. A reflectance switching of the film is achieved by unwinding the helical structure of the blue phase in an electric field, where the highest reflectance achieved in the red, green, and blue color regions is 94%, 86%, and 72%, respectively, in the presence of an electric field of 1 V μm^{-1}. Compared to previously demonstrated BPLC films, this newly developed multi-layer BPLC film substantially improves the overall reflection efficiency by up to 3.6 times. Our study on hyper-reflective BPLC film provides an attractive platform for future development including sequential colorful reflective displays and switchable optoelectronic devices.

1. Introduction

Blue phases represent self-assembled liquid crystal states with a three-dimensional (3D) nanostructure existing between the isotropic phase and the chiral nematic phase.[1,2] There are three well-known blue phases, so called BPIII, BPII, and BPI, which can be observed during the process of cooling from the isotropic phase to the chiral nematic phase. BPIII is an amorphous phase, whereas BPII and BPI possess double-twisted cylinders arranged in a cubic crystalline structure with simple cubic and body-centered cubic symmetry, respectively.[3,4] BPLCs have attracted people's interest for many potential applications, such as field-sequential displays,[5,6] full-color reflective displays,[7,8] electrically switching devices,[9–12] tunable photonic crystals,[13,14] phase modulator optical devices,[15–17] as well as soft templates for 3D colloidal crystals.[18] The intrinsic features of blue phases include a submillisecond response time, optically isotropic dark state, and periodic helical structure on the order of the visible wavelength.

Because only circularly polarized incident light with the same handedness as that of the BPLC will be reflected, the reflectance of a single BPLC film for unpolarized or linearly polarized incident light is limited to 50%. Recent reports have shown that it is possible to increase the reflectance by assembling one BPLC template with another reflective layer, such as a BPLC or cholesteric liquid crystal (CLC), with opposite handedness. The increased reflectance of BPLCs is critical to applications such as reflective displays, color filters, lasers, and mirrors. The BPLC template, which was firstly proposed by Castles et al.,[19,20] was a free-standing porous material fabricated by reactive mesogens and liquid crystals in BP. It was used to form a stretchable and flexible BPLC film by refilling liquid crystal.[19,20] H. C. Jau et al. studied the electro-optical properties of templated blue phase liquid crystals as well as the effects of the helical twisting power of the filling mesogen on electro-optical properties.[21] The transfer of the orientational order of the blue phase to the surface of the polymer matrix and the resulting surface anchoring were numerically modeled.[22] Based on the BPLC template, Guo et al. reported a double-layer BPLC film with a segregation of the left-handed (LH) and right-handed (RH) domains across the LC film to reflect both left- and right-circularly polarized light.[23,24] In Guo's paper, two kinds of double-layer structures were demonstrated: one was fabricated using a R-BPLC template and a L-BPLC template, with a reflectance of 25%; another was fabricated using a R-BPLC template and a pure L-CLC, with a reflectance of 36%. The low reflectances are partially due to the low reflectance of the single BPLC film formed in BPI and partially

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due to the large scattering induced by the refractive index mismatch of the BPLC film and BPLC/CLC during the assembling process. Therefore, the currently reported reflectance of BPLC is still quite low and far from satisfaction and a super-high reflectance in visible light range would be highly desirable for practical applications.

In this paper, we experimentally demonstrate for the first time the fabrication of hyper-reflective, electrically switchable, fast responsive, and colorful reflective displays based on a multi-layer BPLC film consisting of two BPLC templates with opposite handedness formed in BPII. Hyper-reflectivities of 89%, 82%, and 68% were achieved in red, green, and blue color by refilling the multi-layer BPLC film with achiral nematic liquid crystals, achieving improvements of 3.6, 3.3, and 2.7 times that of a traditional multi-layer BPLC film, respectively. The reflectance switching of the film was achieved by unwinding the helical structure of the blue phase in an electric field, where the highest reflectance achieved in the red, green, and blue color region was 94%, 86%, and 72%, respectively, in an electric field of 1 V μm^{-1}. The proposed electrically switchable and hyper-reflective multi-layer BPLC film can largely improve the performance of sequential colorful reflective displays. It can also be applied in other photonic applications such as switchable optoelectronic components, lasers, mirrors, etc.

2. Results and Discussions

Figure 1a shows the schematic structure of LH and RH BPII liquid crystals with a simple-cubic lattice, constructed by LH and RH chiral dopant liquid crystals (in green), as well as the shape of the double-twisted cylinders and disclination lines (in red). For unpolarized incident light, the LH and RH BPLCs will selectively reflect the LH and RH circularly polarized light, respectively. The local refractive index variation leads to a selective Bragg reflection in the spectral range that depends on the chiral dopant concentration. The reflection wavelength is determined by

$$\lambda = 2na/\sqrt{h^2+k^2+l^2} \quad (1)$$

where n and a represent the average refractive index and lattice constant of BP, respectively, and h, k, and l are the Miller indices of a crystal plane.[7] In our experiment, a left-handed blue phase liquid crystal (L-BPLC) and a right-handed blue phase liquid crystal (R-BPLC) layers were fabricated separately. Then, the L-BPLC and R-BPLC templates, consisting of a polymer network and pores first occupied by LC molecules, were obtained after washing out the LC molecules and all remaining unpolymerized monomers/RMs (step I), as shown in Figure 1b. The BPLC templates retained the BPLC lattice structure and could reform BPII by refilling with an achiral nematic liquid crystal. The observed Kossel diagram of the fabricated BPLC film is shown in Figure S1 (Supporting Information), which is consistent with the reported Kossel diagram of BPII.[4,25,26] Figure 1c shows the proposed multi-layer film, which consists of three layers including the L-BPLC, R-BPLC, and an interlayer in between the two. Such multi-layer BPLC film was achieved by assembling (step II) two BPLC films with opposite handedness together and then refilling (step III) with an achiral nematic liquid crystal (HTG135200-100, Hecheng Display, P. R. China). For unpolarized light, the fabricated multi-layer film can reflect LH and RH circularly polarized light simultaneously if the reflection notches of L-BPLC and R-BPLC overlap, which can increase the reflectivity to a point higher than the limit of 50% in single-layer BPLCs.

Figure 2a depicts the surface morphology of the single-layer L-BPLC template observed by scanning electron microscopy (SEM). The cross section of a single-layer L-BPLC template is also demonstrated in the inset. The polymer network is porous. Figure 2b shows the cross-sectional SEM image of the multi-layer BPLC film assembled in step II, where no LCs have been refilled yet. The polymer structure has collapsed and no porous structure can be observed directly in the SEM. Two polymer layers, including a R-BPLC layer with a thickness of 8.7 μm and a L-BPLC layer with a thickness of 7.7 μm, can be observed. The thicknesses of the L-BPLC and R-BPLC layers were designed to be equal to each other and the difference may be due to the washout process and fabrication errors. In addition, a gap with a thickness of 1 μm is also found between the R-BPLC and L-BPLC layers, representing an undesirable interlayer, which is minimized to reduce its effect on the multi-layer BPLC film in our experiment. After step III, the refilling with an achiral nematic LC, the gap as well as the pores in the BPLC template will be occupied by the achiral nematic LC. After refilling with the achiral liquid crystal, the polymer network will gradually become a BPII. Figure 2c–f demonstrates the photos of the multi-layer BPLC film with a central wavelength of 545 nm captured during the refilling process after 1 second, 5 minutes, 30 minutes, and 1 hour, respectively. It can be seen that the multi-layer BPLC template is transparent at the beginning, then gradually shows more and more reflective green color with increasing refilling of the achiral nematic LC, and finally illustrates a bright reflective green color.

Figure 3a–c demonstrates the reflection spectrum of a multi-layer BPLC film refilled with an achiral nematic LC corresponding to either a red, green, or blue reflective color, respectively. All reflection spectra were measured using unpolarized light. In order to obtain multi-layer films with a hyper-reflectivity for different colors, the concentration of the chiral dopant in the LH and RH BPLC films was carefully adjusted to ensure the central wavelengths of the reflection spectra of the two films with opposite handedness were superimposed. The blue and red curve represent the reflection spectra of the LH and RH single-layer BPLC film, respectively. At the central wavelength, the maximal reflectance of L_R, L_G, L_B, (R_R, R_G, R_B) was 45%, 43%, and 38% (48%, 47%, and 36%), respectively. An integrating sphere system (from Ocean Optics) was used to calibrate the absolute value of the reflectance, and a spectrometer (from Ocean Optics) was applied to measure the reflectance of the sample. After assembling the two films together, the reflection spectrum of the multi-layer BPLC film was measured and is demonstrated by the black curve in Figure 3a–c. Compared to a previously reported BPLC film consisting of a R-BPLC template and L-BPLC with a maximal total reflectance of 25% or a blue color BPLC achieved in BPI,[7,23] the proposed multi-layer BPLC film consisting of two BPLC templates with opposite handedness in BPII demonstrates a distinguished advantage

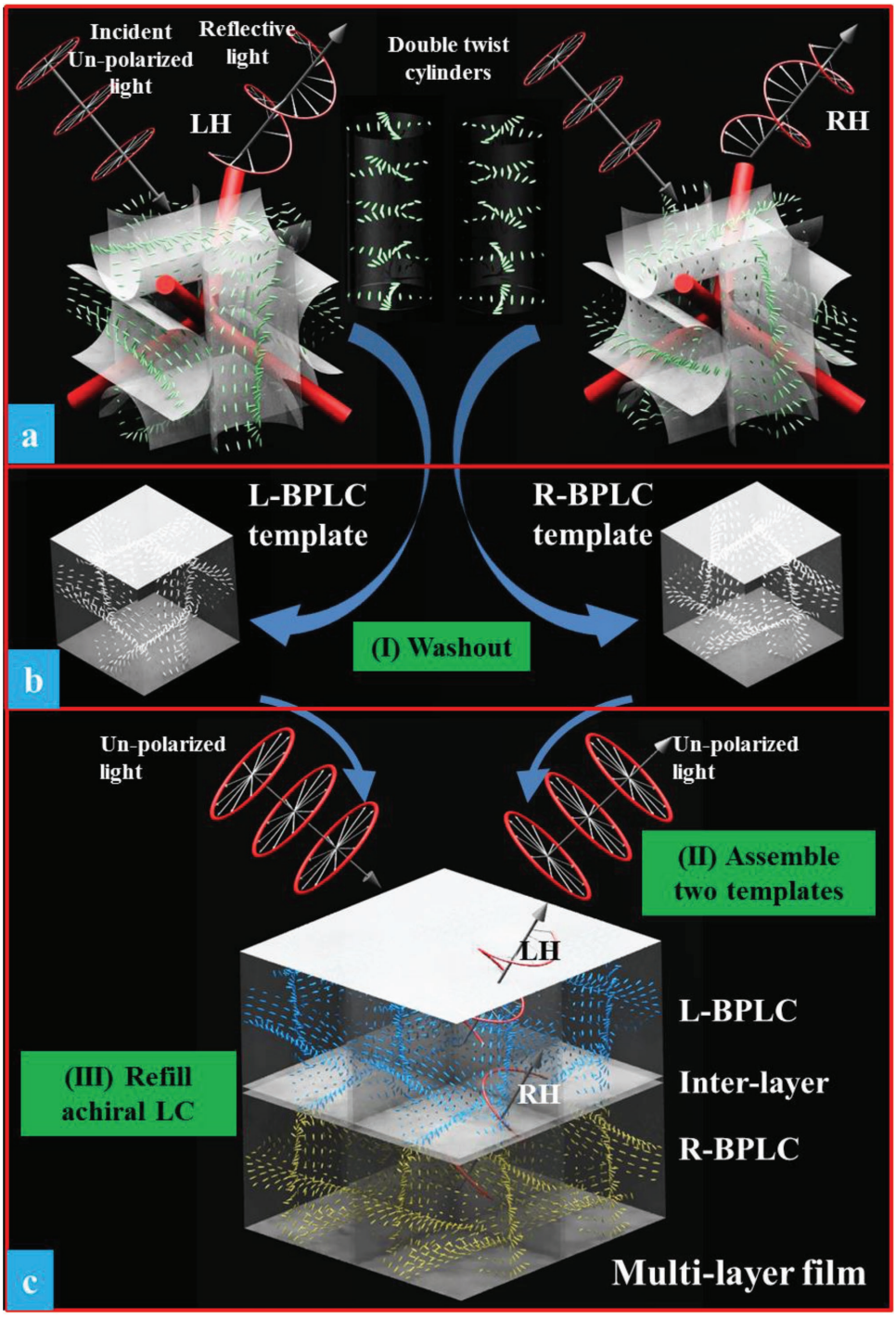

Figure 1. a) Schematic of left-handedness (LH) and right-handedness (RH) BPII liquid crystal with simple-cubic lattice structure. The LC molecules self-assemble as double-twisted cylinders in BP. The LH- and RH-BPLCs selectively reflect LH and RH circularly polarized light, respectively. b) L-BPLC and R-BPLC templates are obtained through immersing the BPLC sample in toluene, where the LC molecules will be washed out (step I). c) The proposed multi-layer film consists of three layers including the L-BPLC, R-BPLC, and an inter-layer in-between the two. The multi-layer BPLC film is achieved by assembling (step II) the two BPLC films with opposite handedness together and then refilling (step III) with an achiral nematic liquid crystal.

of hyper-reflectivity in the visible range. It can be seen that the reflection spectrum of multi-layer BPLC films refilled by achiral nematic LC HTG135200-100 is centered at 650 nm, 550 nm, and 440 nm, with a maximal reflectance of 89%, 82%, and 68% (3.6 times, 3.3 times, and 2.7 times compared to recently demonstrated results),[23] respectively. The insets of Figure 3a–c show photos of multi-layer BPLC films taken under a fluorescent lamp, indicating the full-color, hyper-reflective film

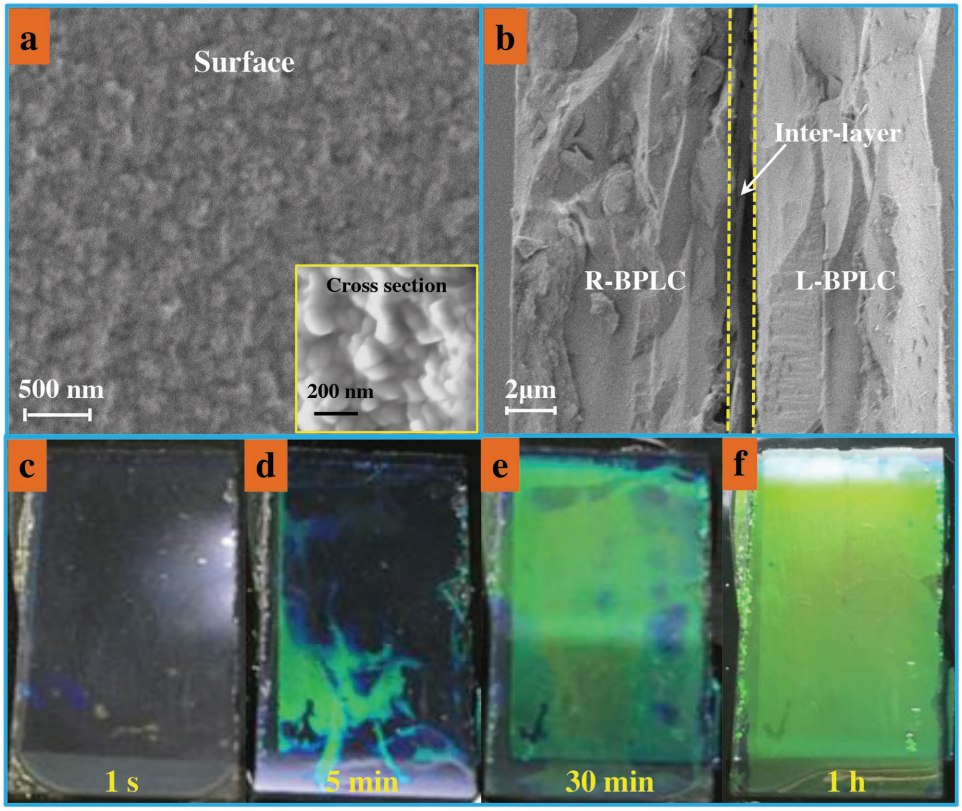

Figure 2. a) Surface morphology of the single-layer L-BPLC template observed by scanning electron microscopy (SEM). The cross section of the single-layer L-BPLC template is shown in the inset. b) Cross-sectional SEM image of the multi-layer BPLC film, before refilling with LCs. The thickness of the R-BPLC, L-BPLC, and inter-layer is 8.7 μm, 7.7 μm, and 1 μm, respectively. c–f) Photos captured during the refilling process of a multi-layer BPLC film with central wavelength of 545 nm after c) 1 s, d) 5 min, e) 30 min, and f) 1 hour, respectively.

fabricated by our method. As a proof-of-concept, we successfully demonstrated that the multi-layer BPLC films can dramatically improve the reflectance in the red, green, and blue colors by about 2.7–3.6 times.

The optoelectronic properties of the multi-layer BPLC films were explored in an electric field by applying an external alternating current (AC) voltage. The configuration of the BPLC film under the electric field is depicted in **Figure 4**a. The BPLC films consisted of a L-BPLC, an inter-layer of the achiral nematic liquid crystal, and an R-BPLC. The optical performance of the multi-layer BPLC film under an external electric field can be discussed in three parts: i) When no electric field was applied, the L-BPLC and R-BPLC were both in a bright state. However, the orientation of the achiral NLC in the inter-layer was random, which partially reduced the total reflectance of the BPLC due to increased disordering and scattering. Therefore, although the reflectance was high (68%, 82%, and 89% at 450 nm, 545 nm, and 654 nm, as shown in Figure 4c–e), it was not the highest at 0 V μm^{-1}, as shown in Figure 4b. ii) When an electric field higher than the threshold of the NLC in the inter-layer but lower than the threshold of the LC in the L- and R-BPLC was applied, the NLC molecules in the inter-layer would align along the electric field, which acted as the transparent layer with low scattering of the reflective light,

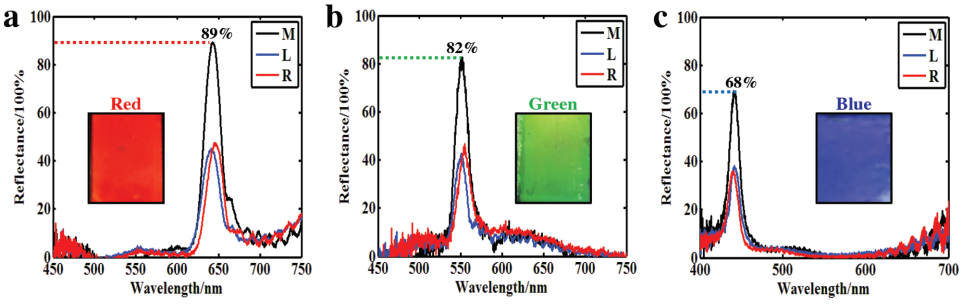

Figure 3. a–c) Reflection spectra of multi-layer BPLC films refilled with NLC corresponding to the reflective red (a), green (b), and blue (c) color, respectively. The insets show photos of the BPLC films taken under a fluorescent lamp, corresponding to the red, green, and blue color, respectively.

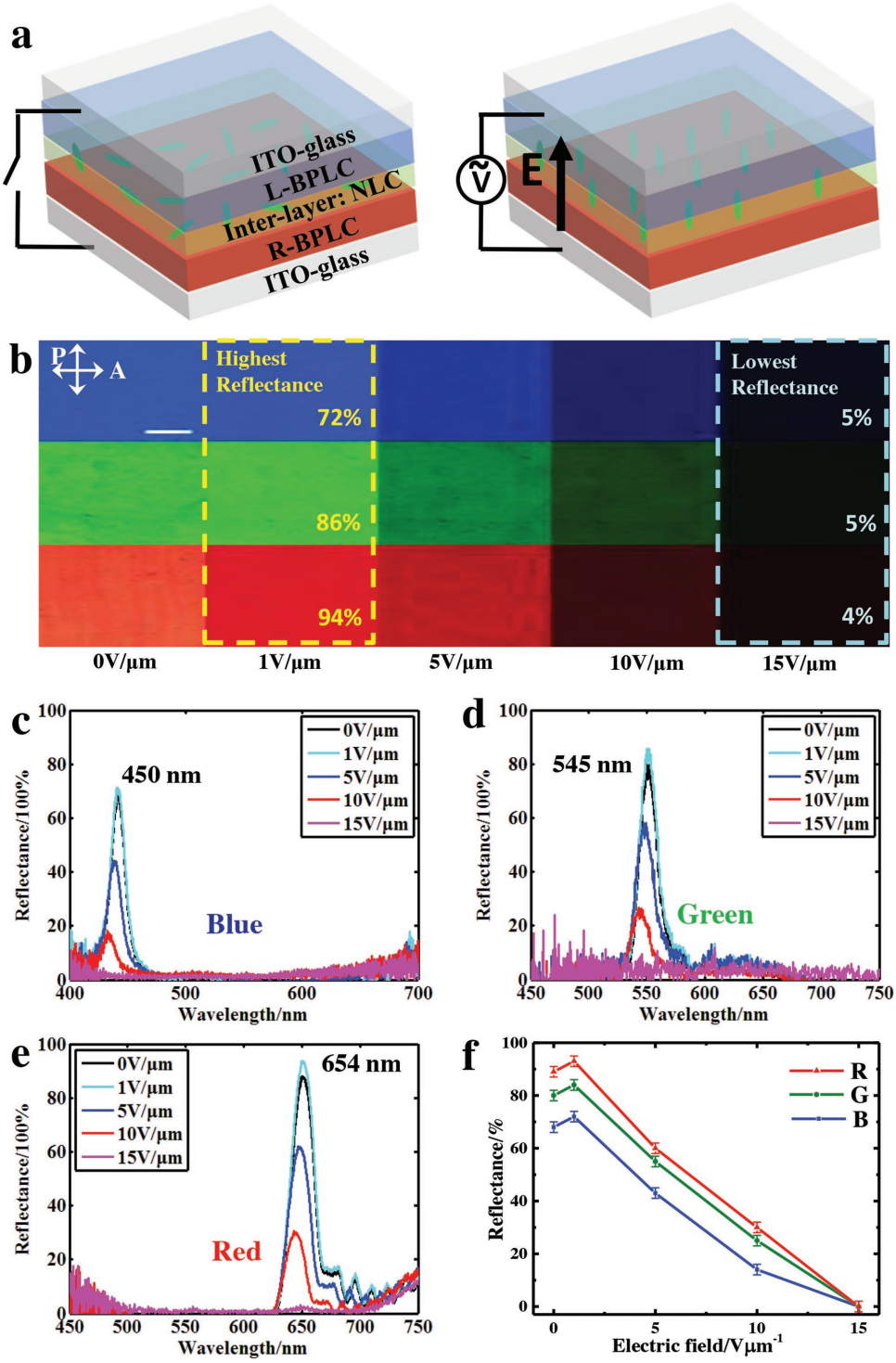

Figure 4. a) Configurations of the BPLC film with and without an electric field. b) Polarized optical microscopy images of the multi-layer BPLC film in blue, green, and red color regions under increasing electric field (up to 15 V μm^{-1}), the scale bar is 100 μm. c–e) Reflection spectra of the multi-layer BPLC film under an electric field of 0 V μm^{-1}, 1 V μm^{-1}, 5 V μm^{-1}, 10 V μm^{-1}, and 15 V μm^{-1} for the blue (c), green (d), and red (e) color regions, respectively. f) Relationship of reflectance versus electric field for 450 nm (B), 545 nm (G), and 654 nm (R).

resulting in a slight increase of the reflectance. It should be noted that the threshold of the NLC in the inter-layer was lower than that of the LC in the BPLC because the polymer network in BPLC itself increased the threshold of the LC molecules. Therefore, the highest reflectances of 72%, 86%, and 94% at 450 nm, 545 nm, and 654 nm were obtained at an electric field of 1 V μm^{-1} (as shown by the yellow dashed rectangle in Figure 4b). iii) When the applied electric field was increased

further to a value higher than the threshold of the LC in the BPLC but within the low-field region (which was the case in our experiment), the electric field aligned the LC molecules but did not deform the lattice structure of the blue phase, which usually happens in the high-field region. Therefore, the aligned LC molecules led to a reduced refractive index difference between the LC and the polymers in the BPLC and thus a gradual decrease of the reflectance under increasing electric field.[7] The polarized optical microscopy images under an applied electric field in the blue, green, and red reflection regions are demonstrated in Figure 4b. It can be seen that when the electric field was increased from 1 V μm^{-1} to 15 V μm^{-1}, the reflectance gradually decreased from its highest value (72%, 86%, 94% for blue, green, and red, respectively) to its lowest value (5%, 5%, 4% for blue, green, and red, respectively). For our multi-layer BPLC film, the reversing of the Bragg reflection was not perfect and it led to a hysteresis effect, which was similar to that of traditional BPLCs.[16] The corresponding reflection spectra in blue, green, and red, demonstrated in Figure 4c–d, are summarized in Figure 4f. It is worth mentioning that if the inter-layer can be further reduced the highest reflectance will appear at a lower electric field (less than 1 V μm^{-1}), maybe even at zero electric field. A photodetector (New Focus, Model 2031) was used to measure the response time of our multi-layer BPLC film. The rise and decay times of the multi-layer BPLC film in the green color region (defined as 10% to 90% and 90% to 10% of the reflectance change with the AC electric field between 0 V μm^{-1} and 15 V μm^{-1}, and at a frequency of 1 kHz) were 0.36 ms and 0.53 ms, respectively (Figure S2, Supporting Information). The fast response time of the BPLC film enables potential applications in sub-sequence reflective displays and fast switchable photonic devices.

3. Conclusion

Hyper-reflective, electrically switchable, multi-layer BPLC films have been fabricated based on two BPII templates with opposite handedness. The fast response, and colorful reflective displays based on proposed BPLC films were demonstrated experimentally. Hyper-reflectivities of 89%, 82%, and 68% in the red, green, and blue reflection color regions were achieved by refilling the multi-layer blue phase liquid crystal films with free-standing porous polymer network with an achiral nematic liquid crystal. The newly developed multi-layer BPLC film substantially improved the overall reflection efficiency by as much as 3.6, 3.3, and 2.7 times (in red, green, and blue, respectively), compared to previous results.[23] The reflectance switching of the film was achieved by unwinding the helical structure of the blue phase in an electric field, where highest reflectances of 94%, 86%, and 72% were achieved in the red, green, and blue color regions, respectively, under an electric field of 1 V μm^{-1}. The underlying mechanism was analyzed and can be attributed to the existence of the inter-layer NLC. This kind of electrically switchable and hyper-reflective BPLC film with submillisecond response time brings significant improvements on the performance of colorful reflective displays with high reflectivity. This technology can also be used in switchable optoelectronic devices, lasers, mirrors, etc.

4. Experimental Section

Materials: In our experiment, achiral nematic liquid crystal HTG135200-100, R5011, and S5011 (from Hecheng Display, China), reactive mesogen A1, A2, and A3 (from Shi Jia Zhuang Sdyano Fine Chemical, China), monomer B1, B2 (from Yantai Wanrun, China), and B3 (from Sigma-Aldrich), and photo-initiator IRG184 (from Hecheng Display, China) were used to fabricate the BPLC film. The chemical structures are shown in Figure S3 (Supporting Information). The concentration of monomers should be high enough to fill up the spaces of the double twisted cylinder structure of BPLC; if not, the film cannot retain its blue phase after refilling. Therefore, 19 wt% of monomer/polymers were used in our experiment. Herein, we used three polymerizable mesogens, i.e., A1, A2, A3, with a similar molecular structure but different length of the terminal groups to modulate the strength of the formed polymer network and the anchoring between the polymer and the LCs; the mesogenic structure was to obtain a better solubility in the LC system. The monomers B1, B2, and B3 were used to modulate the photo-crosslinking density. The monomer B1 with two longer terminal hydrocarbon chain was adopted to avoid an excessive density of polymer, thereby maintaining a better driving performances; while B2 and B3 with the short terminal chains were used to prevent a more loose polymer network, thus enhancing the structural strength of the polymer template; in addition, another purpose for mixing B2 and B3 was decreasing the viscosity of the pre-polymerized mixture, thus promoting a more sufficient photoreaction. The material composition corresponding to red, green, and blue colors is shown in **Table 1**.

Fabrication of the BPLC Film: Firstly, the BPLC/prepolymer mixture was filled into a LC cell assembled by two glass substrates coated with an indium tin oxide (ITO) electrode. The thickness of the LC cell was 10 μm. Taking the BPLC template in red reflection color as an example, the BPLC was heated up to 80 °C to the isotropic phase. The BPII, BPI, and chiral nematic phase appeared at 75 °C, 71 °C, and 67.5 °C, sequentially, when the temperature decreased gradually. After that, the BPLC sample in BPII was exposed to ultraviolet (UV) light with a wavelength of 360 nm and an intensity of 10 mW cm^{-2} for 30 minutes at a temperature of 71.5 °C. During the UV exposure, the reactive mesogens and monomers would photo-polymerize to stabilize the BPLC. The working temperature range of BPII was broadened from 4 °C (71 °C–75 °C) before polymerization to 105 °C (0 °C–105 °C) after polymerization. To obtain the BPLC template, the polymer-stabilized BPLC was immersed in toluene for about 12 hours to wash out the LCs, leading to a BPLC template with polymer network structure. Next, the LC cell was opened by cutting the

Table 1. Materials composition.

	LC host [wt%]	Chiral dopant [wt%]	Reactive mesogens [wt%]			Monomers [wt%]			Photoinitiator [wt%]
	HTG135200-100	R5011/S5011	A1	A2	A3	B1	B2	B3	IRG184
Red	77.36	2.64	5.7	2.85	5.7	1.9	1.9	0.95	1
Green	76.96	3.04	5.7	2.85	5.7	1.9	1.9	0.95	1
Blue	76.48	3.52	5.7	2.85	5.7	1.9	1.9	0.95	1

sealing glue and the BPLC template shrank and separated naturally from the glass substrate while drying in air. The LH and RH BPLC templates were prepared by the same procedure. Two BPLC templates with opposite handedness were assembled together and then transferred to the surface of a glass substrate. Another glass substrate was applied to form a new LC cell with a thickness of 20 µm. Finally, the multi-layer BPLC film was fabricated by refilling with the achiral nematic LCs.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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