

Piecewise Phototuning of Self-Organized Helical Superstructures

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Cholesteric liquid crystals (CLCs) exhibit selective reflection that can be tuned owing to the dynamic control of inherent self-organized helical superstructures. Although phototunable reflection is reported, these systems hitherto suffer from a limitation in that the tuning range is restricted to one narrow period and the optically addressed images have to sacrifice one color in the visible spectrum to serve as the background, resulting from the insufficient variation in helical twisting power of existing photoresponsive chiral switches that are all bistable. Here, delicate patterns of three primary red, green, and blue (RGB) colors with a black background are presented, which is realized based on piecewise reflection tuning of the CLC induced by a newly designed photoresponsive tristable chiral switch. Three stable configurations of the chiral switch endow the CLC with two continuous and adjacent tuning periods of the reflection, covering not only entire visible spectrum, but also one more wide period within near-infrared region. Therefore, the concept of piecewise tuning in CLC system demonstrates a new strategy for phototunable RGB and black reflective display.

Reflecting light, which is often based on periodic structures rather than pigments, is a fundamental strategy for most of the creatures to transmit optical signals in nature.^[1,2] Moreover, intimidation, camouflage, display, and communication among animals are frequently realized by voluntary color changes in response to external stimuli. Natural periodic structure observed in animals, plants, and even minerals generates a photonic bandgap, where the light propagation of a specific wavelength is prohibited. Taking inspiration from nature, scientists strived to use various responsive materials to fabricate periodic structures, not only obtaining the desired reflection,^[3] but also tuning the reflection by light,^[4] electricity,^[5,6] stress,^[7] temperature,^[8] pH,^[9] solvent,^[10] and humidity.^[11]

In the family of liquid crystals (LCs), cholesteric LCs (CLCs) possess periodic structures, i.e., self-organized helical superstructures, in which the orientation of the molecules varies in a helical fashion and thus selectively reflects light according to Bragg's law.^[12] The selective reflection is determined by the pitch length of the helices in CLC and can be tuned thanks to the dynamic control of the self-organized helical superstructure by external stimuli. Currently, light-driven CLCs have attracted

extensive interest because light, compared to other stimuli, has unique advantages of remote, temporal, local, and spatial manipulation.^[13–19] Such elegant systems are generally formulated by doping photoresponsive chiral switches, mainly consisting of chiral centers and photo-switches, into a nematic LC material. The chiral centers have the capability to twist the nematic LC phase into helical superstructure, which is represented by helical twisting power (HTP). The photoswitches undergo configurational changes upon photoisomerization, leading to the variation in HTP and the pitch length of the helices, and consequently tune the selective reflection of the CLC. These light-driven CLCs provide opportunities for various photonic applications such as tunable LC lasers,^[20,21] tunable color filters,^[22,23] and optically addressed display.^[24–26]

Scientists have devoted themselves to developing photoresponsive chiral switches that have high HTP and large HTP variation upon isomerization. Azobenzenes are well-known photoswitches that possess two isomers of *trans* and *cis*. Chiral switches based on azobenzenes^[26–34] usually have large variation in HTP due to the extraordinary configurational changes between rod-like *trans* isomers and bent *cis* isomers, and enable the tuning range of the resultant CLC to cover visible spectrum. Besides, chiral switches based on dithienylethenes^[35–39] have also been synthesized and utilized to tune the reflection of the CLC across red, green, and blue (RGB) colors. One should emphasize that the phototunable reflection remains unchanged in darkness even after one week, resulting from the excellent thermal stability of dithienylethenes.^[35,36]

With these systems mentioned above, optically addressed images including three primary RGB colors were demonstrated in a single thin film because of the reversible color tuning across entire visible spectrum. However, these systems described to date suffer from a limitation in that the optically addressed images have to sacrifice one color in visible spectrum to serve as background, which are thus unable to show the patterns of a certain color similar to the background and undesirable for full-color reflective display. This limitation results from the inadequate variation in HTP caused by the existing photoresponsive chiral switches that are all bistable^[40,41] because each of them only contains one kind of photoswitch with two isomeric configurations in chemical structure (such as *trans* and *cis* isomers of azobenzene or ring-opened and ring-closed forms of dithienylethene). The real challenge is to expand the tuning period

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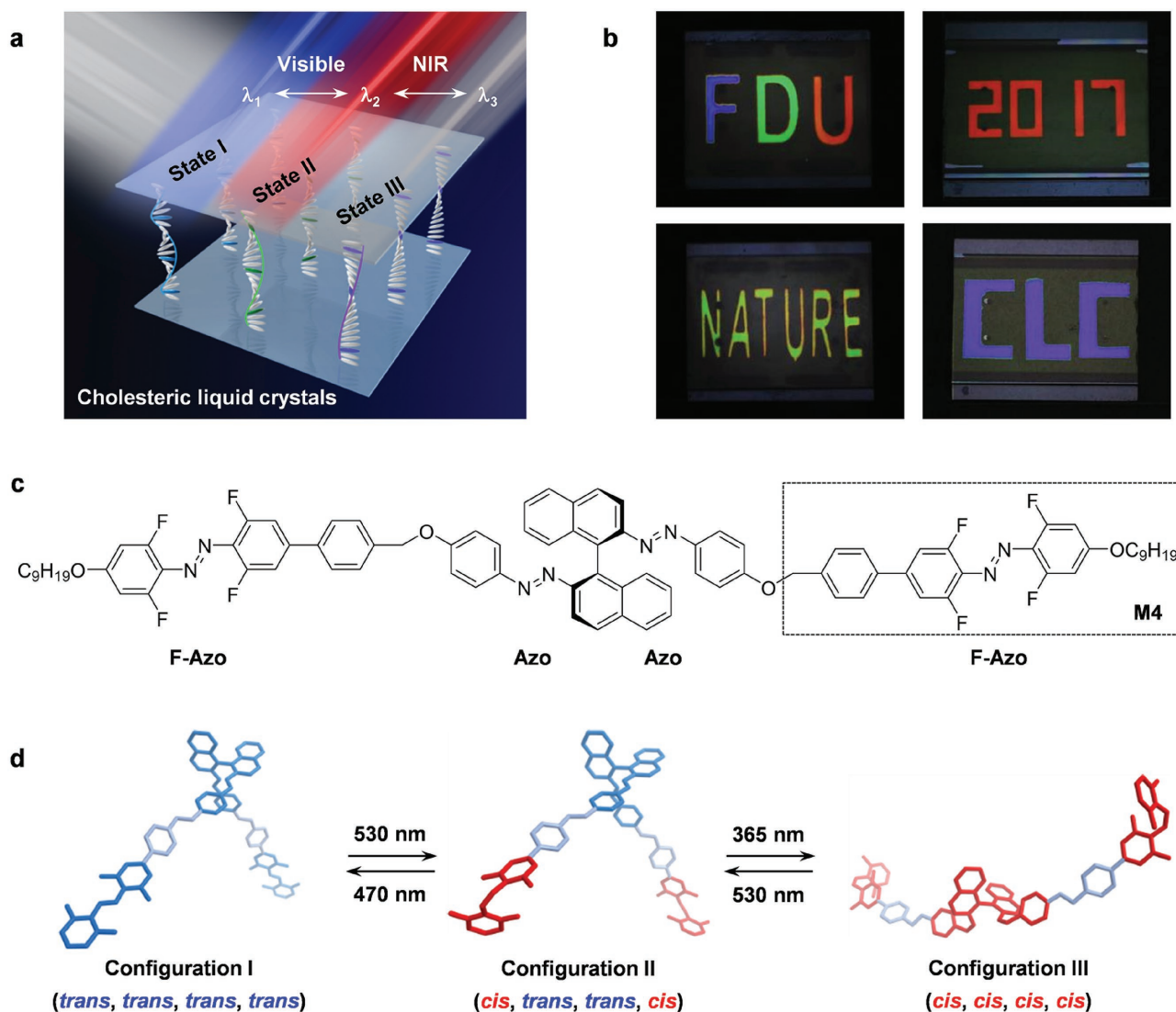


Figure 1. a) Schematic representation showing the piecewise control of self-organized helical superstructures in the CLC that exhibits two tuning periods of the selective reflection, including visible spectrum and near-infrared region. The selective reflection λ is determined by the pitch length (p) of the helices, $\lambda = np$, where n is the average refractive index of the LC host. b) Real cell images created by piecewise control of the helices in the CLC to show different patterns of RGB colors with a black background in 5 μm thick antiparallel aligned cells (2.0 cm \times 2.5 cm). c) Chemical structure of the photoresponsive tristable chiral switch containing Azo near the chiral center and F-Azo far away from the chiral center. The moiety in the dashed box is an important intermediate named M4. d) Schematic illustration showing the optimized molecular structures of three configurations of the tristable chiral switch, including Configuration I (*trans, trans, trans, trans*), Configuration II (*cis, trans, trans, cis*), and Configuration III (*cis, cis, cis, cis*). *Trans* and *cis* isomers are in blue and red. Hydrogen atoms and end groups $-\text{OC}_9\text{H}_{19}$ are omitted for clarity.

to near-infrared region and still remain the tuning of reflection wavelength covering visible spectrum at the same time.

Herein, a newly designed photoresponsive tristable chiral switch is constructed by incorporating two different photo-switches into one chiral structure. Remarkable variation in HTP among three configurations of our tristable chiral switch endows the CLC with two adjacent and continuous tuning periods of reflection, which are independently controlled by light of different wavelengths (Figure 1a). We preserve the RGB reflection colors of the images in visible spectrum, and further tune the reflection within near-infrared region to create a black background (Figure 1b). This unprecedented piecewise reflection tuning of self-organized helical superstructures in the CLC

induced by the tristable chiral switch provides a new concept for RGB and black (R_GB_B) reflective display.

Incorporating two kinds of different azobenzenes into one chiral structure would offer a manner to synthesize photoresponsive tristable chiral switches. In light of this idea, we designed the photoresponsive tristable chiral switch based on parent azobenzene (Azo) and *o*-fluoroazobenzene (F-Azo) (Figure 1c; Figure S1, Supporting Information). The binaphthalene moiety as an axially chiral center endows the tristable chiral switch with high HTP. Azo is one kind of photoswitch near the chiral center, which isomerizes from *trans* to *cis* upon exposure to UV light and restores from *cis* to *trans* upon exposure to visible light. F-Azo is another photoswitch far away

from the chiral center, which has been reported to undergo the *trans*–*cis* isomerization induced by UV light, as well as visible light thanks to the well separation of n – π^* bands of *trans* and *cis* isomers.^[42]

In order to confirm that our designed F-Azo moiety actually exhibits *trans*–*cis* isomerization induced by both UV light and visible light, photoisomerization of an important intermediate M4 containing F-Azo has been carefully studied (Figures S2 and S3, Supporting Information). The n – π^* bands of *trans* and *cis* isomers are separated by 22 nm characterized by UV–vis spectroscopy. Both 530 and 365 nm light resulted in high *trans*–*cis* photoconversions of 74% and 83%, respectively, and 60% *cis*–*trans* photoconversion occurred when irradiated with 470 nm light.

Our tristable chiral switch containing Azos and F-Azos would possess Configuration I (*trans, trans, trans, trans*) and Configuration III (*cis, cis, cis, cis*) because of the basic isomerization induced by 365 and 470 nm light. It is worth noting that 530 nm light triggers the *trans*–*cis* isomerization of F-Azo and the *cis*–*trans* process of Azo at the same time. As a result, the tristable chiral switch will show Configuration II (*cis, trans, trans, cis*) upon irradiation of 530 nm light (Figure 1d). When doped into an LC host, the tristable chiral switch is able to immediately induce an optically tunable helical superstructure and endows the system with two tuning periods of the reflection. We anticipated that one period is tuned in visible spectrum to demonstrate rich colors and another period is tuned within near-infrared region to create the black background because near-infrared light is not perceived by human eyes (Figure 1a). In this scenario, we can realize optically addressed images of **RGBB** colors in a single thin film.

As Figure 2b shows, our chiral switch exhibits three dominant configurations at three photostationary states (PSSs) as expected. Irradiated with 530 nm light, F-Azo underwent the *trans*–*cis* isomerization and the intensity of the π – π^* band around 360 nm decreased, while Azo remained unchanged in the *trans* form. Thus, the tristable chiral switch transformed to Configuration II (*cis, trans, trans, cis*) at the PSS₅₃₀. Moreover, both Azo and F-Azo isomerized from *trans* to *cis* upon exposure to 365 nm light, leading to the further intensity decrease of the π – π^* bands of both Azo and F-Azo, and giving rise to Configuration III (*cis, cis, cis, cis*) at the PSS₃₆₅. Irradiation of 470 nm light induced the *cis*–*trans* recover process of both Azo and F-Azo, and restored the intensity of the π – π^* bands, resulting in Configuration I (*trans, trans, trans, trans*) at the PSS₄₇₀.

When a small amount of the tristable chiral switch was doped into commercially available nematic LC host E7, a right-handed CLC mixture immediately formed. The characteristic fingerprint texture was observed by a polarized optical microscope even at a low concentration of 0.09 mol%, indicating very high HTP of our chiral switch at the initial state (Figure S4, Supporting Information). We used wedge cells to measure the pitch length of the CLC mixture and then calculated the HTP values (Figure S5, Supporting Information). The tristable chiral switch exhibits high HTP of 271 μm^{-1} (mol%) at the initial state and considerable HTP difference among three PSSs in E7 (Figure 2a), which is ascribed to the configurational changes driven by light with different wavelengths.

As Figure 2a shows, upon 530 nm light irradiation, the HTP changed to 112 μm^{-1} (mol%) at the PSS₅₃₀ (Configuration II-rich state) because of the increase in distance between the disclination lines compared to that at the initial state. When irradiated with 365 nm light, the disclination lines further moved outward, showing that the HTP decreased to 36 μm^{-1} (mol%) at the PSS₃₆₅ (Configuration III-rich state). Upon exposure to 470 nm light, the HTP increased up to 160 μm^{-1} (mol%) at the PSS₄₇₀ (Configuration I-rich state) since the distance between the disclination lines became shorter. After that, 530 nm light irradiation decreased the HTP to 112 μm^{-1} (mol%) at the PSS₅₃₀ again (cycle noted by blue arrows). The HTP was reversibly manipulated among three PSSs according to the cycle noted by dashed green arrows.

The tristable chiral switch remained original HTP value at each PSS without obvious fatigue upon sequential exposure to light stimuli of three different wavelengths after 50 cycles (Figure 2c). In addition, the thermal relaxation of the HTP from 36 μm^{-1} (mol%) to 198 μm^{-1} (mol%) took 80 h in darkness at room temperature, which was ascribed to the *cis*–*trans* isomerization of Azo (Figure S6, Supporting Information). After that, the dark relaxation was extremely slow thanks to the thermally stable *cis* isomer of F-Azo with a remarkable long half-life.^[42]

To gain further insight into the relationship between structure and property, we simulated the optimized structures of three configurations of the tristable chiral switch at the B3LYP/6-31G level (Figure S7, Supporting Information), which help to explain the effect of molecular structures on photochemical HTP variation. The Configuration I (*trans, trans, trans, trans*) possesses a stretched shape. The Configuration II (*cis, trans, trans, cis*) shows a bent shape due to the bent structure of F-Azos at *cis* state. All bent structures of Azos and F-Azos in Configuration III (*cis, cis, cis, cis*) lead to a zigzag shape. According to the HTP and the optimized structures, we conclude that the stretched structure of the *trans* form is beneficial for our chiral switch to transfer its chirality into LC host (high HTP), while the bent structure of the *cis* form has poor ability to twist a nematic LC phase (low HTP).

In addition to the HTP values obtained at three PSSs by monochromatic light, we can manipulate the HTP in a detailed way by using composite light. It has been found that the HTP is determined by the equilibrium ratio of different configurations of the chiral switches.^[26] In light of this theory, irradiation of composite light can give rise to different equilibrium ratios of the configurations and thus manipulate the HTP. Indeed, the HTP was manipulated between 160 μm^{-1} (mol%) and 112 μm^{-1} (mol%) upon exposure to composite light of 470 and 530 nm with different intensities (Figure 2d; Figure S8, Supporting Information). 530 and 470 nm light intensities were competitive to each other and triggered the opposite isomerization process of F-Azo,^[43] controlling the ratio of Configuration I and Configuration II when two light intensities were changed. Based on the same mechanism, exposure to composite light of 530 and 365 nm with different intensities manipulated the HTP between 112 μm^{-1} (mol%) and 36 μm^{-1} (mol%) because composite light determined the ratio of Configuration II and Configuration III (Figure 2e; Figure S9, Supporting Information). Therefore, according to the simulative 3D curve surfaces, we can obtain the desired HTP in a controllable way by using

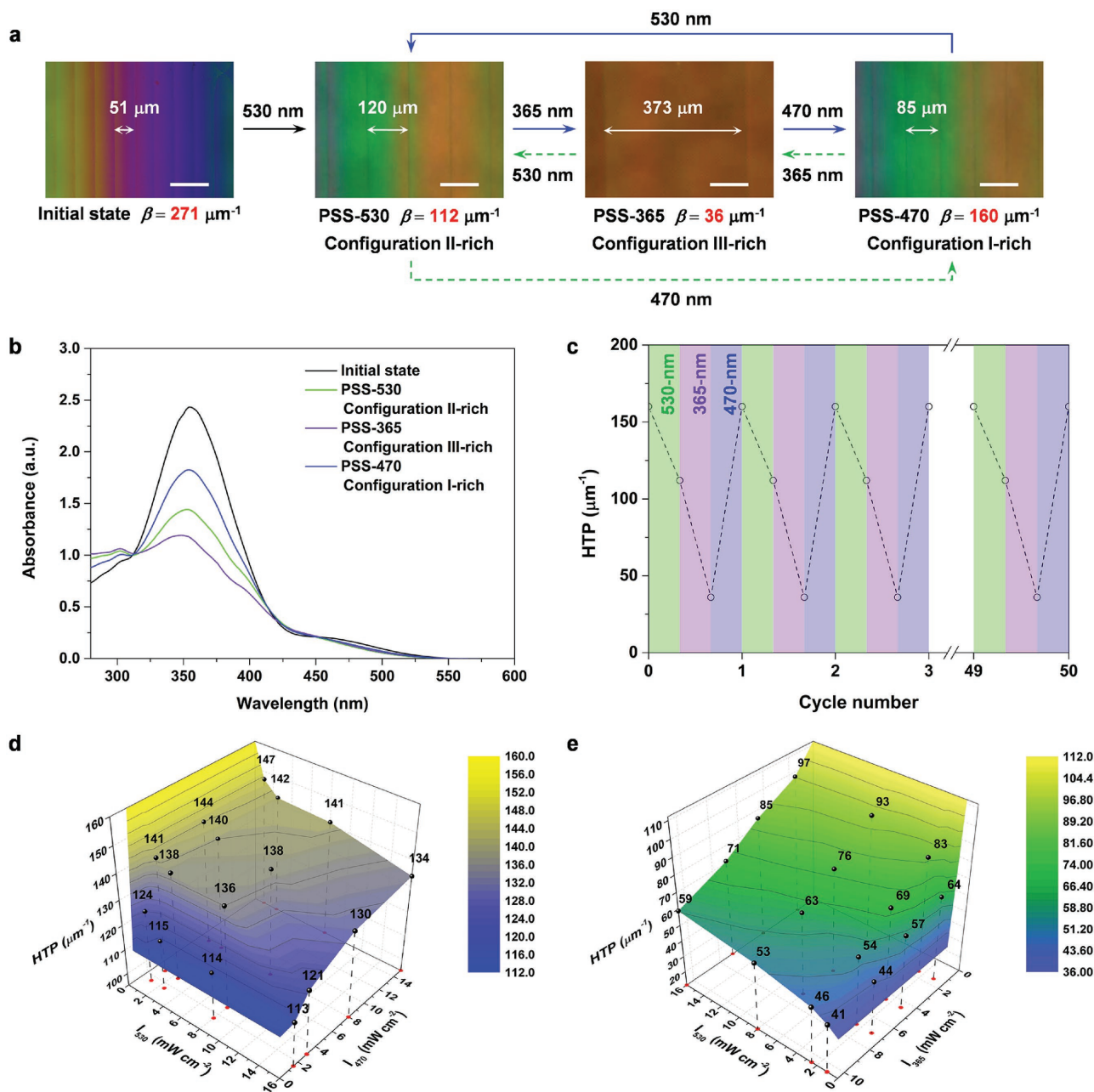


Figure 2. a) Optical images showing the disclination lines of 0.19 mol% tristable chiral switch in LC host E7 at different states. The HTP values of the tristable chiral switch at the initial state, PSS₅₃₀, PSS₃₆₅, and PSS₄₇₀ are 271, 112, 36, and 160 μm^{-1} (mol%), respectively. The intensities of 470, 530, and 365 nm light are 40, 80, and 10 mW cm^{-2} , respectively. Irradiation times are all 30 s to reach the PSSs. Scale bar: 100 μm . b) UV-vis spectra of the tristable chiral switch in CH_2Cl_2 ($c = 2 \times 10^{-5}$ M) at the initial state and three PSSs. c) Repetition experiment for measuring the HTP of the tristable chiral switch upon sequential irradiation with 530, 365, and 470 nm light in 50 cycles. d, e) 3D curve surfaces showing the HTP induced by composite light of 470 and 530 nm (d), or composite light of 530 and 365 nm (e) with different light intensities: I_{470} , intensity of 470 nm light; I_{530} , intensity of 530 nm light; I_{365} , intensity of 365 nm light.

composite light. Recently, Lee and co-workers have also demonstrated CLC manipulation by using composite light.^[44,45] However, they paid more attention to the changes on LC states, and we focused on exact HTP value at different PSSs.

Encouraged by the appropriate HTPs as well as the remarkable variation among three PSSs, we demonstrated piecewise tuning of the selective reflection in both visible spectrum and

near-infrared region. A mixture of the tristable chiral switch in E7 was capillary filled into a 5 μm thick antiparallel aligned cell that was painted black on one side to create a contrast with a bright iridescent Bragg reflective color from the CLC when viewed from the other side of the cell. The reflection color of 2.0 mol% tristable chiral switch in E7 was dark blue at the PSS₄₇₀ and was tuned from blue to green and red upon 530 nm

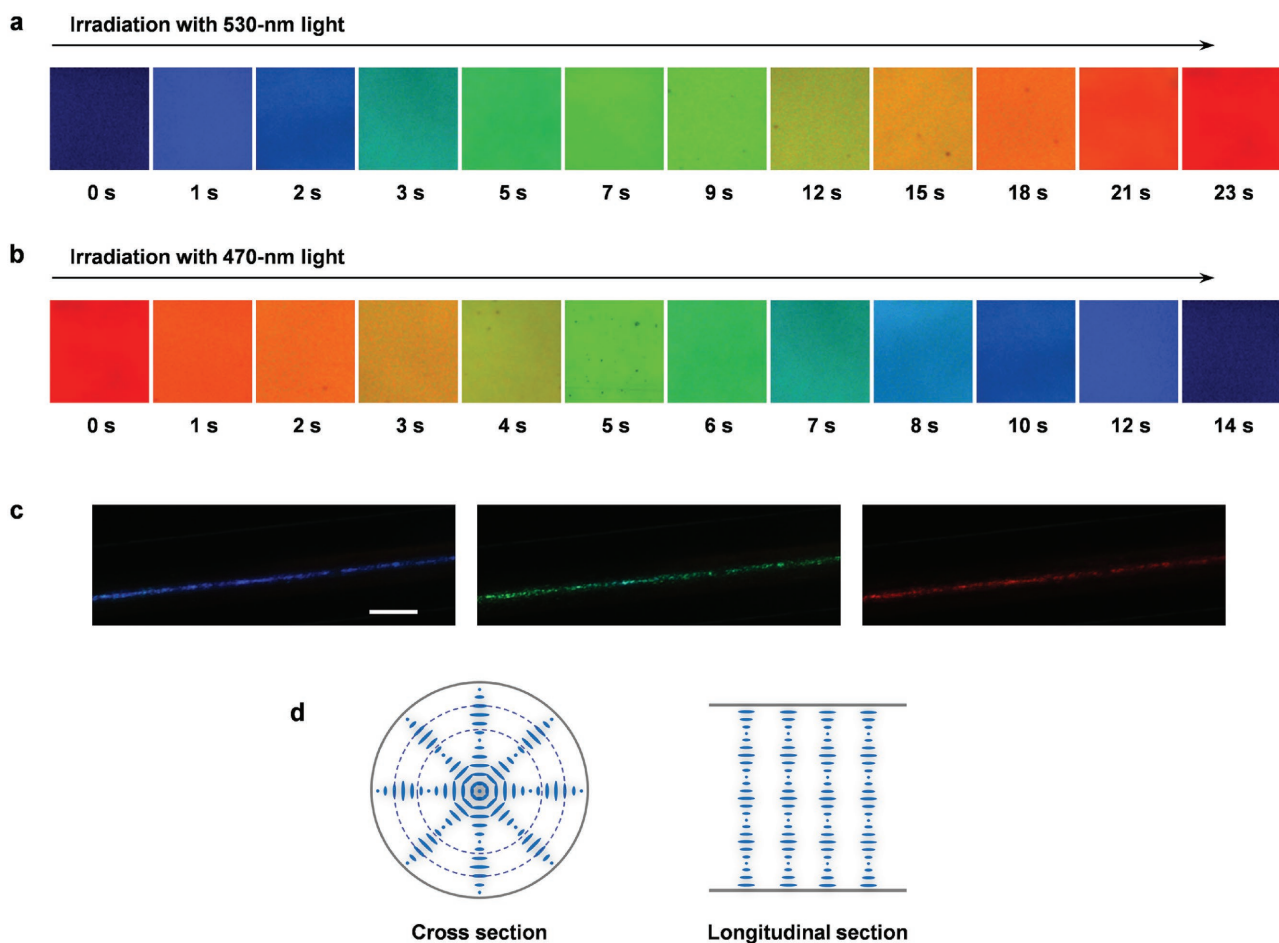


Figure 3. Reflection color images of 2.0 mol% tristable chiral switch in LC host E7 in a 5 μm thick antiparallel aligned cell upon exposure to: a) 530 nm light (80 mW cm^{-2}) and b) 470 nm light (40 mW cm^{-2}) with different times. The reflection color images were taken from a polarized optical microscope in reflective mode between the PSS₄₇₀ and the PSS₅₃₀. c) Reflection color images of 2.0 mol% tristable chiral switch in LC host E7 in a capillary taken from a polarized optical microscope in reflective mode. The inner diameter of the capillary is 60 μm . Scale bar: 100 μm . d) Schematic illustration showing the LC alignment in the capillary viewed from cross section and longitudinal section.

light irradiation (80 mW cm^{-2}) in 23 s (Figure 3a; Figure S10, Supporting Information), whereas irradiation of 470 nm light (40 mW cm^{-2}) induced the reverse process from red to green and blue in 14 s (Figure 3b; Figure S11, Supporting Information). The decrease of the HTP caused by 530 nm light irradiation led to the increase of the pitch length as well as the redshift of the reflection and vice versa. By taking advantage of visible light irradiation, we achieved reversible reflection tuning in the visible spectrum including rich and bright colors with fast response. Interestingly, when the CLC mixture was filled into a capillary, the reflection color was still observed and tuned with 530 and 470 nm light (Figure 3c). The CLC mixture confined in the capillary has helical superstructures with axes radially aligned at cross section, as the LC molecules form planar layers along the inner surface of the capillary, constructing a 2D periodic structure (Figure 3d).

Moreover, thanks to the significant decrease of HTP from $112 \mu\text{m}^{-1}$ (mol%) at the PSS₅₃₀ to $36 \mu\text{m}^{-1}$ (mol%) at the PSS₃₆₅, the above-mentioned CLC mixture was found to exhibit another tuning period in the near-infrared region as well. Upon exposure to 365 nm light (3 mW cm^{-2}), the reflection

wavelength was tuned from 670 to 2100 nm with a wide range of $\approx 1430 \text{ nm}$ in the near-infrared region because of the further decrease of the HTP (Figure 4a). The reflection wavelength at the PSS₃₆₅ was able to be switched back by 530 nm light irradiation (20 mW cm^{-2}) due to the recovery of the HTP (Figure 4b). Based on the tristable performance of our chiral switch, the CLC possessed two adjacent and continuous tuning periods of the reflection, which provides piecewise tuning in the visible spectrum by 470 and 530 nm light irradiation and in the near-infrared region by 530 and 365 nm light irradiation, respectively. This piecewise reflection tuning was repeated many times without obvious degradation.

In addition, continuous 365 nm light irradiation causes the reflection of the CLC mixture (1 mol% tristable chiral switch in E7) to tune more than 2000 nm owing to the high HTP at the initial state ($271 \mu\text{m}^{-1}$, mol%) and remarkably large variation in HTP between the initial state and the PSS₃₆₅ ($\Delta\beta/\beta_{\text{ini}} = 87\%$) (Figure 4c). Most of the previous investigations on photo-tunable CLC have only demonstrated reflection variation of $\approx 500 \text{ nm}$ ²⁶. The maximum shift of our CLC mixture ($\approx 3760 \text{ nm}$ in theory) is not experimentally quantifiable by our reflection

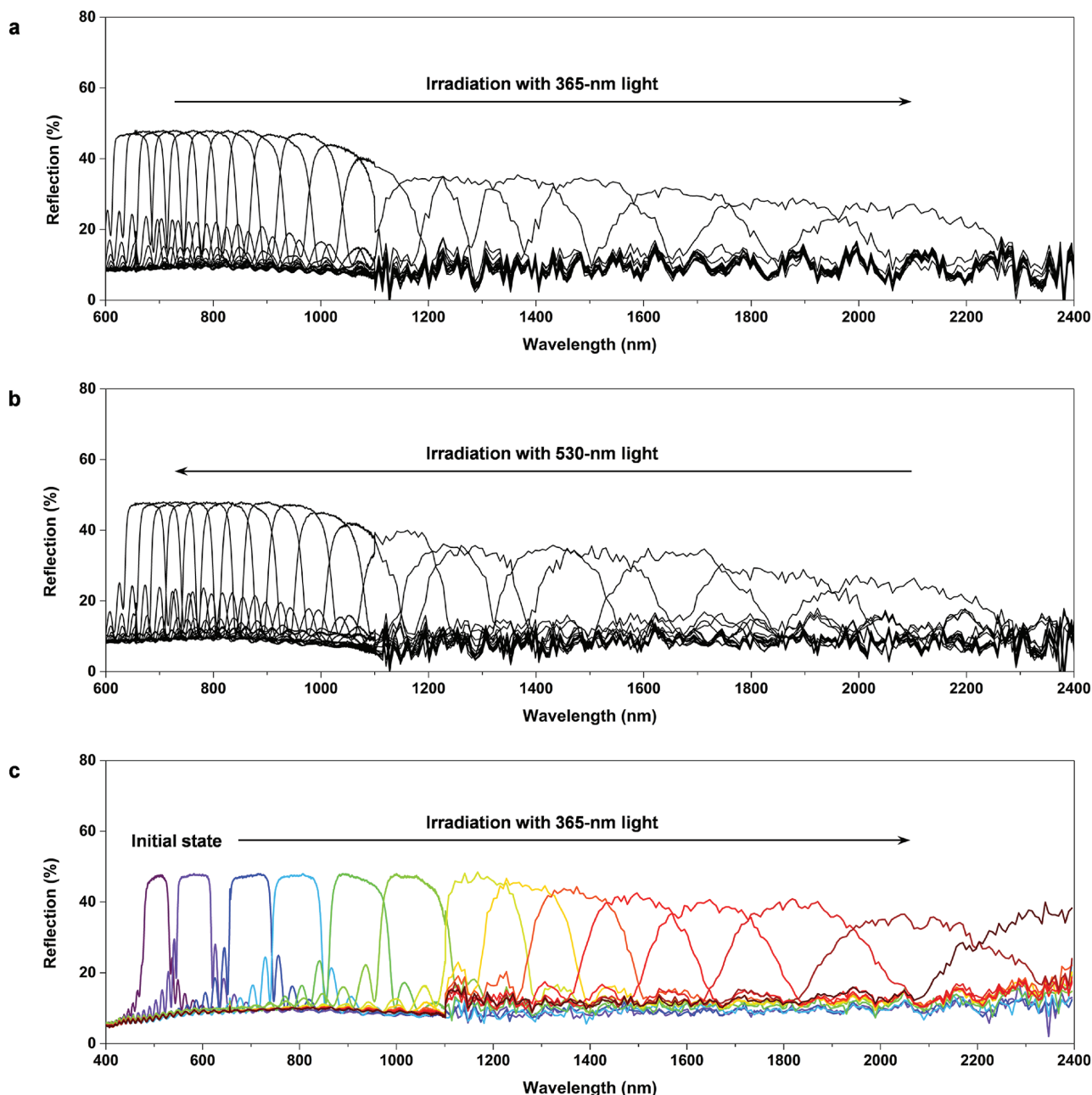


Figure 4. Reflective spectra of 2.0 mol% tristable chiral switch in LC host E7 in a 5 μm thick antiparallel aligned cell upon exposure to: a) 365 nm light (3 mW cm^{-2}) from the PSS₅₃₀ to the PSS₃₆₅ and b) followed by exposure to 530 nm light (20 mW cm^{-2}) from the PSS₃₆₅ to the PSS₅₃₀. c) Reflective spectra of 1.0 mol% tristable chiral switch in LC host E7 in a 5 μm thick antiparallel cell upon exposure to 365 nm light from the initial state.

spectroscopy (200–2500 nm), but the broad tuning range over 2000 nm encompasses visible, near-infrared, and a portion of short-wave infrared spectra.

Having established that the light-induced configurational changes of the tristable chiral switch result in the piecewise control of the reflection wavelength of the resultant CLC, we are able to create colorful patterns by irradiating the cell with different light through masks. The three primary RGB reflection colors were obtained upon irradiation of 530 nm light for different times (Figure 5a–c). When the reflection color was tuned to the desired one, 365 nm light was used to irradiate the cell

with a mask placed on the cell for creating patterns. The reflection wavelength of the exposed areas was further tuned to near-infrared region, driving the background color to become black, while the unexposed areas remained original color. Therefore, delicate images with the black background, such as patterns of Chinese mahjong (Figure 5d–f), numbers and English letters (Figure 1b), were addressed on the cells. These images could be seen clearly after 4 h, demonstrating acceptable color stability (Figure S12, Supporting Information).

Furthermore, letters of RGB colors could be observed simultaneously in a single cell (Figure 1b), which was prepared first

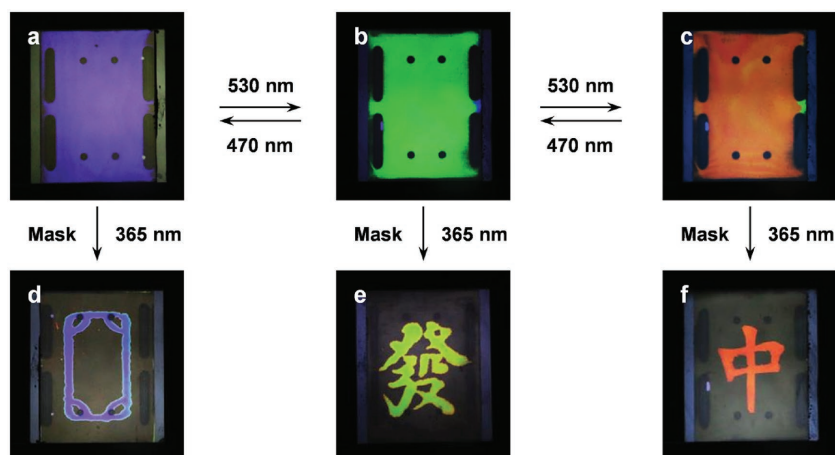


Figure 5. Real cell images of 2.0 mol% tristable chiral switch in LC host E7 in 5 μm thick antiparallel aligned cells (2.0 cm \times 2.5 cm) showing a–c) primary RGB colors and d–f) patterns of Chinese mahjong with the black background.

based on 530 nm light irradiation for different times to generate three areas of RGB colors and followed by 365 nm light irradiation with an “FDU”-shaped (FDU) mask. These experiments demonstrate that our light-driven CLC capable of creating **RGBB** images has promise for application in full-color reflective display.

In summary, the photoresponsive tristable chiral switch was constructed by incorporating Azo and F-Azo moieties into one chiral structure, to the best of our knowledge for the first time. It is worth noting that 530 nm light triggered the *cis*–*trans* isomerization of Azo and the *trans*–*cis* process of F-Azo at the same time. Different photoisomerization of Azo and F-Azo contributed to Configuration I (*trans, trans, trans, trans*), Configuration II (*cis, trans, trans, cis*), and Configuration III (*cis, cis, cis, cis*) facilitated by irradiation of 470, 530, and 365 nm light, respectively. When doped into LC host E7, our chiral switch immediately induced the optically tunable helical superstructures and endowed the resultant CLC system with two adjacent and continuous tuning periods of the reflection, which exhibited piecewise control of the reflection wavelength. By irradiation of 470 and 530 nm light, the selective reflection was tuned in the visible spectrum including three primary RGB reflection colors. Moreover, another wide tuning range of ≈ 1430 nm within the near-infrared region is realized in the same thin film upon irradiation of 530 and 365 nm light. Based on this mechanism, these novel light-driven CLCs are capable of creating images of RGB colors with a black background, which breaks the limitation caused by the bistable chiral switches and thereby demonstrates a new strategy to develop phototunable **RGBB** reflective display. However, as an intrinsic property of azobenzene derivatives, the thermal relaxation will drive the optically addressed images to slowly fade in time if they are not refreshed by light. Many efforts are still needed to conquer this problem and promote the phototunable CLC systems for future applications.

Experimental Section

Synthesis and characterization of the tristable chiral switch, photoisomerization of M4, and measurement of HTP are described in the Supporting Information.

General: All reagent chemicals were purchased from Adamas-beta, Sigma Aldrich, Alfa Aesar, and J&K Scientific and were used as supplied without further purification unless otherwise noted. Tetrahydrofuran (THF) was dried and distilled from sodium prior to use. Column chromatography was performed on silica gel (200–300 meshes). Deuterated solvents were purchased from Cambridge Isotope Laboratory. ^1H NMR, ^{13}C NMR, and ^{19}F NMR spectra were recorded on a Bruker AVANCE III spectrometer at 400 MHz with use of CDCl_3 as the lock. Chemical shift data are reported as δ in units of parts per million relative to residual solvent and are referenced to tetramethylsilane (TMS) as the internal reference. Data are represented as follows: chemical shift, multiplicity (s = singlet, d = doublet, t = triplet, q = quartet, and m = multiplet), coupling constants in Hertz (Hz), and integration. Mass spectra were recorded on an AB SCIEX 5800 mass spectrometer. Melting points were measured on a TA Q2000 differential scanning calorimeter. UV–vis absorption spectra were recorded on a PerkinElmer Lambda 650 spectrometer. Textures and disclination lines were observed on a Leica DM2500P polarized optical microscope (POM), with a Mettler FP-90 and FP-82 temperature controller at 25 $^\circ\text{C}$. Reflective spectra were recorded on an IdeoOptics Instruments PG2000-Pro-EX reflection spectrometer (200–1100 nm) and an IdeoOptics Instruments NIR2500 reflection spectrometer (900–2500 nm) at room temperature. Wedge cells (KCRK-07) and antiparallel aligned cells were purchased from EHC Co., Ltd. The commercially available eutectic liquid crystal E7 ($\Delta n = 0.223$, $M = 271.74$ g mol^{-1}) and standard dopant S811 and R811 were purchased from HCCH Co., Ltd. 365 nm UV light was generated by an Omron ZUV-H30MC light source with a ZUV-C30H controller. 530 nm green light was generated by a CCS HLV-24GR-3W light source with a PJ-1505-2CA controller. 470 nm blue light was generated by a CCS HLV-24BL-3W light source with a PJ-1505-2CA controller.

Preparation of CLC Mixtures: In order to prepare the CLC mixture, a dichloromethane solution of the tristable chiral switch and LC host E7 was evaporated to dryness at 45 $^\circ\text{C}$ in an oven. The residue was annealed at 55 $^\circ\text{C}$ until it became homogenous. The resultant mixture was filled into an antiparallel aligned cell by capillary action at 50 $^\circ\text{C}$, and then slowly cooled to ambient temperature and stored in the dark for 5 h.

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

liquid crystals, photoresponsive chiral switches, self-organized helices, tristable performance, tunable selective reflection

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