## **Supporting Information**

# Efficient Visible-Light Full-Color Tuning of Self-Organized Helical Superstructures Enabled by Fluorinated Chiral Switches

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### 1. UV-vis absorption spectra



**Figure S1.** Changes of UV-vis absorption spectrum of **S-3-5** in chloroform  $(2 \times 10^{-5} \text{ M})$  (a) upon irradiation of 530-nm light (27 mW cm<sup>-2</sup>) from the initial state to the PSS<sub>530</sub> and (b) followed by 445-nm light irradiation (0.4 mW cm<sup>-2</sup>) from the PSS<sub>530</sub> to the PSS<sub>445</sub>.



**Figure S2.** Changes of UV-vis absorption spectrum of **S-1-3** in chloroform  $(2 \times 10^{-5} \text{ M})$  (a) upon irradiation of 530-nm light (27 mW cm<sup>-2</sup>) from the initial state to the PSS<sub>530</sub> and (b) followed by 445-nm light irradiation (0.4 mW cm<sup>-2</sup>) from the PSS<sub>530</sub> to the PSS<sub>445</sub>.

## 2. Fingerprint texture



Figure S3. Fingerprint textures of 0.4 wt% (a) S-3-5 and (b) S-1-3 in E7 observed through POM. Scale bar, 100  $\mu$ m.

#### 3. Miscibility test

The standard left-handed and right-handed samples were prepared by doping commercially available non-responsive chiral dopants S811 and R811 at the concentration of 9.5 wt% into nematic liquid crystal E7, respectively. To determine the screw senses of the CLCs that the chiral switches induced, CLCs and left-handed or right-handed standard samples were inserted into the cells. As **Figure S4** shows, discontinuous area formed between the CLC mixtures and the right-handed standard LC, while the contact areas were continuous between the CLC mixtures and the left-handed standard LC. Therefore, all chiral switches induced the formation of left-handed helical structures of the CLCs.



Figure S4. POM images of the contact areas between CLC mixtures doped with (a) S-3-5, (b) S-1-3, and (c) S-1-5 and standard LC respectively.

#### 4. Measurement of helical twisting power (HTP)



**Figure S5.** Schematic representation showing the arrangement of the liquid crystals in the wedge cell.

The pitch was measured according to Grandjean-Cano wedge method. Wedge cells with an inclination angle  $\theta$  are made by applying spacers of different size at each end of the cell. Liquid crystals arrange themselves in a way as **Figure S5** shows due to the fixed *p* of the CLCs and antiparallel alignment direction of the substrate. Therefore, disclination lines form between areas with different layers. Both each layer and the thickness difference between adjacent domains are p/2 to satisfy the alignment boundary condition. The disclination lines can be observed through polarized microscope. The pitch is determined by the equation  $p = 2R\tan\theta$ , where *R* represents the distance between the disclination lines and  $\theta$  is the inclination angle of the wedge well (EHC, KCRK-07, tan  $\theta$ = 0.0196). The HTP value is determined according to the equation  $\beta = 1/(pc)$ , where *p* is the pitch of the cholesteric liquid crystal and *c* represents the concentration of the chiral dopants.

The cholesteric liquid crystals were prepared by doping appropriate amount of the chiral dopants into the nematic liquid crystal E7 and mixing them with dichloromethane. After the evaporation of the solvent, the mixtures were inserted into the wedge cell by capillary action. The HTP was determined by plotting  $p^{-1}$  (µm<sup>-1</sup>) against the concentration of the chiral dopants *c* (mol%) and finding the slope of the resulting straight line.



**Figure S6.** POM images of the wedge cells containing 1.0 wt% (a) **S-3-5** (b) **S-1-3** in achiral liquid crystal E7 before and after successional visible light irradiation at 530 nm (middle) and 445 nm to the photostationary states.



Figure S7. Reciprocal helical pitch as a function of concentration of chiral switches (a) S-3-5, (b) S-1-3, and (c) S-1-5 in E7.

### 5. Fatigue resistance



**Figure S8.** Cycles of the changes of HTP of 1.0 wt% (a) **S-3-5** and (b) **S-1-3** in E7 upon alternate irradiation with 530-nm and 445-nm light.





 Meas.m/z
 # Ion Formula
 Score
 m/z
 err [ppm]
 Mean err [ppm]
 mSigma
 rdb
 e<sup>-</sup> Conf
 N-Rule

 983.391735
 1
 C64H51F4N4O2
 100.00
 983.394266
 2.6
 1.4
 59.8
 40.0
 even
 ok



