



Scheme S1 Photochromic interconversion of a BTE unit

Table S1 Solid-state emission data of 1 and 2 before and after irradiation with 365 nm

light at room temperature

Compound	$\lambda_{\max}$ (nm)
1	433, 448, 482, 531, 570
1 after irradiation	428, 447, 482, 531
2	460, 489, 535, 593
2 after irradiation	460, 486, 535



Fig. S1 Experimental and simulated XRD patterns of 1



Fig. S2 <sup>1</sup>H NMR spectrum of 1 (500 MHz, CDCl<sub>3</sub>).



Fig. S3 Experimental and simulated XRD patterns of 2.



Fig. S4 The supramolecular chain structure in 1.



Fig. S5 The packing structure of 1.



Fig. S6 The packing structure of 2.



Fig. S7 Plots a and b: irradiating ( $\lambda = 365$  nm) 1 for 0, 30 minutes, respectively; plots c and d: using 574 nm light to irradiate the sample corresponding to plot b for 10 and 20 minutes, respectively; plot e: placing the sample corresponding to plot b in the dark for 45 minutes.



Fig. S8 Raman spectra of 1 before and after irradiation ( $\lambda = 365$  nm, 30 minutes), using laser light of 785 nm. Three peaks with \* are at 1399, 1614 and 1711 cm<sup>-1</sup>, respectively.



Fig. S9 Absorption spectra changes of 1 in CH<sub>3</sub>CN solution (c =  $2.4 \times 10^{-5}$  M) upon UV irradiation ( $\lambda$  = 270 nm) for 0 to 12 minutes.



Fig. S10 <sup>1</sup>H NMR spectrum of 1 (500 MHz, CDCl<sub>3</sub>) after UV irradiation ( $\lambda = 270$  nm).



**Fig. S11** <sup>1</sup>H NMR spectra of **1** before and after UV irradiation ( $\lambda = 270$  nm).



Fig. S12 Plots a and b: irradiating ( $\lambda = 365$  nm) 2 for 0, 20 minutes, respectively; plots c and d: using 624 nm light to irradiate the sample corresponding to plot b for 10 and 20 minutes, respectively; plot e: placing the sample corresponding to plot b in the dark for 40 minutes.



Fig. S13 Raman spectra of 2 before and after irradiation ( $\lambda = 365$  nm, 20 minutes), using laser light of 785 nm. Three peaks with \* are at 1457, 1500 and 1573 cm<sup>-1</sup>, respectively.



Fig. S14 <sup>1</sup>H NMR spectrum of the irradiated sample of 1 (500 MHz, CDCl<sub>3</sub>; UV irradiation,  $\lambda = 365$  nm).



Fig. S15 <sup>1</sup>H NMR spectrum of the irradiated sample of 2 (500 MHz, DMSO- $d_6$ ; UV irradiation,  $\lambda = 365$  nm).



**Fig. S16** Low-temperature magnetization data for **2** collected under various applied dc fields. The solid lines represent fits to the data.



**Fig. S17** Low-temperature magnetization data for **2L** collected under various applied dc fields. The solid lines represent fits to the data.



**Fig. S18** Temperature dependence of ac susceptibilities of in-phase  $\chi_M$ ' (left) and outof-phase  $\chi_M$ '' (right) for **2** under zero applied static field.



**Fig. S19** Temperature dependence of ac susceptibilities of in-phase  $\chi_M$ ' (left) and outof-phase  $\chi_M$ '' (right) for **2L** under zero applied static field.



**Fig. S20** Temperature dependence of ac susceptibilities of in-phase  $\chi_M$ ' for **2** under 1000 Oe applied static field.



Fig. S21 Temperature dependence of ac susceptibilities of in-phase  $\chi_M$ ' for 2L under 1000 Oe applied static field.



Fig. S22 Cole-Cole plots of 2 in the temperature range of 1.8-2.6 K under applied static field of 1000 Oe; the solid lines are the best fit by Debye model ( $\alpha < 0.17$ ).



**Fig. S23** Cole-Cole plots of **2L** in the temperature range of 1.8-2.6 K under applied static field of 1000 Oe; the solid lines are the best fit by Debye model ( $\alpha < 0.14$ ).