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## **Supporting Information**

## Hydrogen-Bonded Supercoil Self-Assembly from Achiral Molecular Components with Light-Driven Supramolecular Chirality

Yangyang Wang,<sup>ab</sup> Deyan Zhou,<sup>a</sup> Haining Li,<sup>a</sup> Ruiru Li,<sup>a</sup> Yueyao Zhong,<sup>ab</sup> Xuan

Sun,\*a and Xun Sun<sup>b</sup>

<sup>†</sup>Key Laboratory for Colloid & Interface Chemistry, Shandong University, Education Ministry, Jinan, 250100, P. R. China

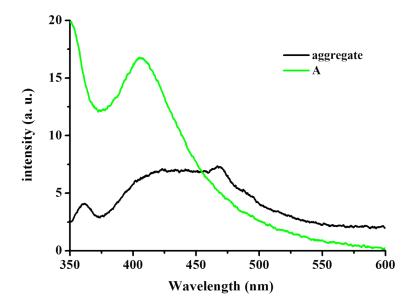
<sup>‡</sup>State Key Lab of Crystal Materials, Shandong University, Jinan, 250100, P. R. China

E-mail: sunxuan@sdu.edu.cn

## Content

- 1. Fluorescence spectra of the aggregate and azobenzene A.
- 2. FT-IR characterization of the intermolecular hydrogen-bonding formed between A and M.
- 3. TGA analysis of the complex  $M \cdot A_3$
- 4. Elemental analysis of the complex  $M \cdot A_3$
- 5. <sup>1</sup>H NMR analysis of the complex  $M \cdot A_3$

- 6. Wide-angle X-ray diffraction (WAXD) patterns of the supercoils
- 7. UV-vis and CD spectra of the supercoils upon 365 nm irradiation
- 8. TEM image of the supercoils after photoirradiation with 365 nm light.
- 9. Crystal structure of M-BA
- 1. Fluorescence spectra of the aggregate and azobenzene A



**Figure S1.** Fluorescence spectra of the self-assembled suspensions in water and the highly dispersed molecule **A** in THF at  $c = 2.5 \times 10^{-5}$  mol L<sup>-1</sup>, respectively.

2. FT-IR characterization of the intermolecular hydrogen-bonding formed between A and M.

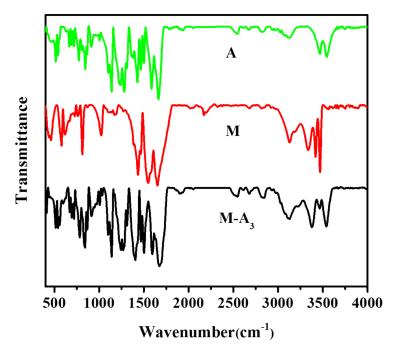


Figure S2. Comparison of the IR spectra of A, M and  $M-A_3$  complex at room temperature.

3. TGA analysis of the complex  $M \cdot A_3$ 

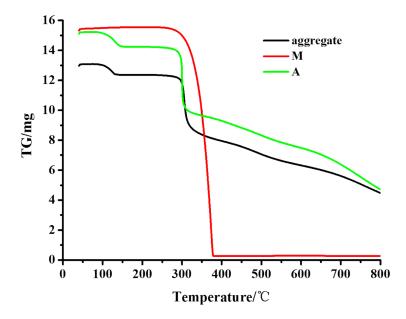
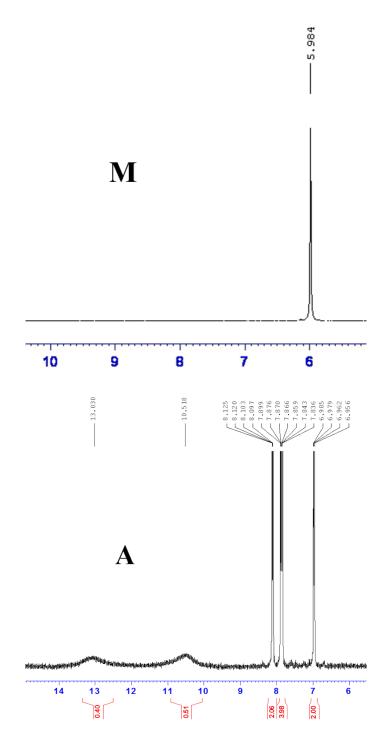


Figure S3. Thermalgravimetric analysis of A, M, and M•A<sub>3</sub> complex in 3:1 ratio.

4. Elemental analysis of the complex  $M \cdot A_3$ 

Table S1. Elemental analysis of the complex $M \cdot A_3$					
Element	Ν	С	Н	0	
Percentage%	17.06	53.85	4.722	24.368	

5. <sup>1</sup>H NMR analysis of the complex  $M \cdot A_3$ 



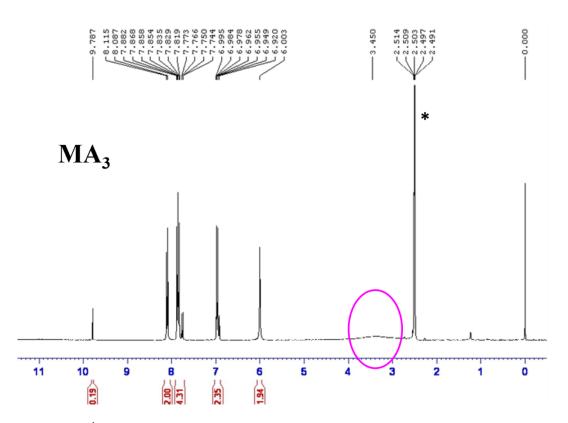


Figure S4. <sup>1</sup>H NMR spectrum of M, A and M•A<sub>3</sub> using the residual solvent resonance of DMSO (denoted as \*) at 2.505 ppm relative to  $SiO_2$  as an internal reference.

6. Wide-angle X-ray diffraction (WAXD) patterns of the supercoils

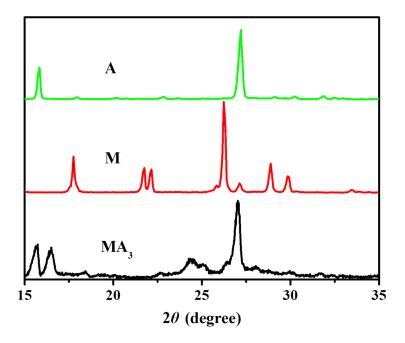
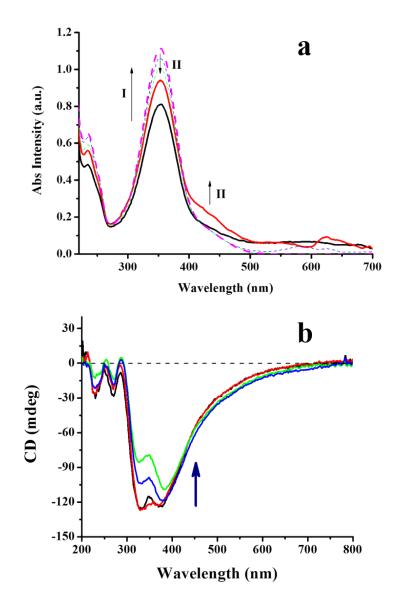


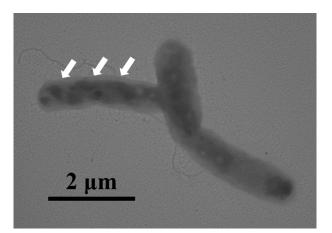
Figure S5. WAXD patterns of the solid A, solid M, and the supercoils formed from  $M \cdot A_3$ 

7. UV-vis and CD spectra of the supercoils upon 365 nm irradiation



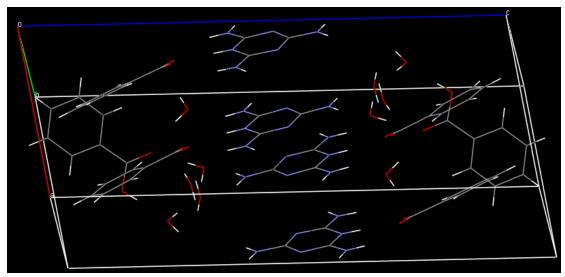
**Figure S6.** (a) UV-vis spectra and (b) CD spectra indicate the structure changes of the supercoils upon 365 nm irradiation. Two stages of changes in the absorption spectra are observed as indicated by the arrows.

8. TEM image of the supercoils after photoirradiation with 365 nm light.



**Figure S7.** TEM image of the supercoils after photoirradiation with 365 nm light. The arrows indicate the remaining helical sense of the supercoils.

9. Crystal structure of M-BA



**Figure S8.** Side view of the hydrogen-bonding network within the crystal of **M-BA**. Protons on the –COOH of **BA** are transferred onto the N atom of the triazine, and the –C–O bonds of the benzoic acid units are identical. Highly ordered water channels inside the nanorods through hydrogen bonding, and the direction of the channels is parallel to the direction of the crystal growth.