Electronic Supplementary Information

Two-Dimensional Networks of Azobenzene Derivative: Bi-Pyridine Mediation and Photo Regulation

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1. Spectra of the filter (365 nm and 435 nm)

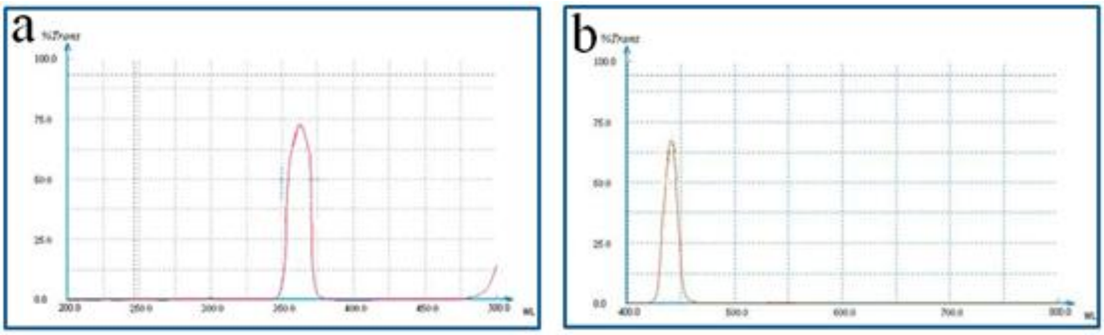

Figure S1. Spectra of Xelon lamp with filter (a) $\lambda_{max} = 365$ nm and (b) $\lambda_{max} = 435$ nm.

2. UV-Vis spectra of NN4A

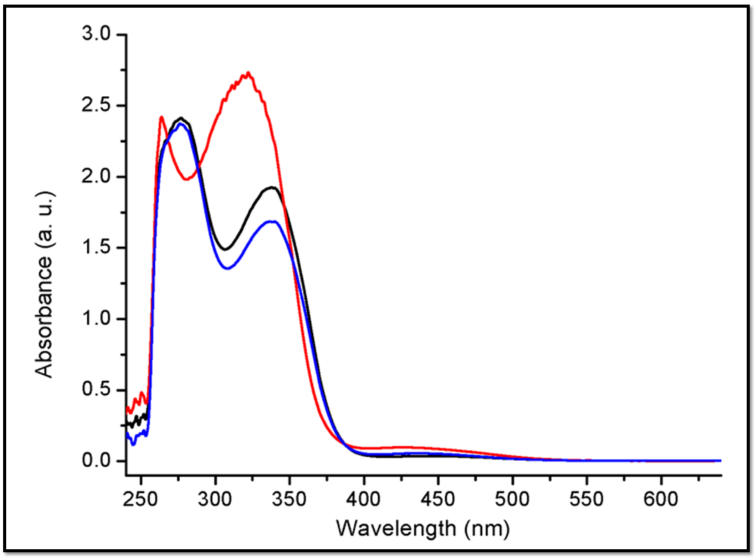

Figure S2. UV-Vis spectra of NN4A (C = 1.0 ×10^{-5} M) in heptanoic acid: Before (*trans*-NN4A, black); after UV light irradiation at 365 nm for 30 mins (*cis*-NN4A, red); and after subsequent visible light irradiation at 435 nm for 15 mins (*trans*-NN4A, blue).

3. UV-Vis spectra of DPE

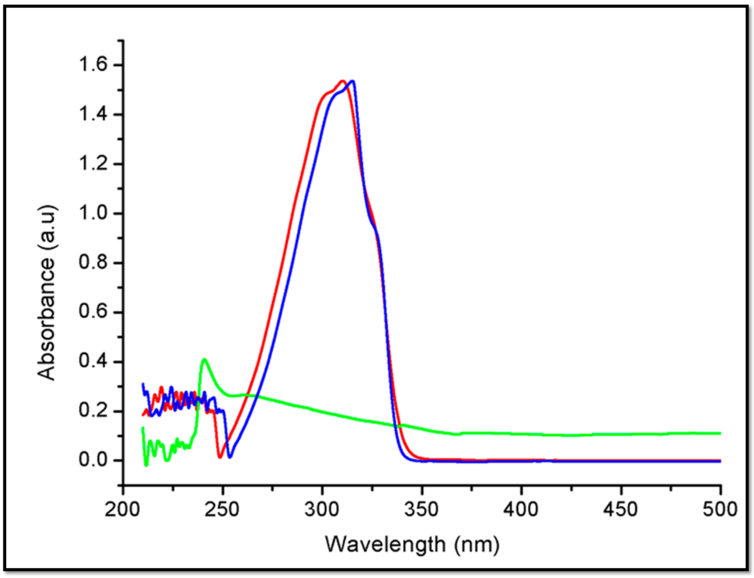

Figure S3. UV-Vis spectra of DPE (C = 1.0 ×10^{-5} M) in heptanoic acid: Before (*trans*-DPE, blue); after UV light irradiation at 365 nm for 30 mins (*cis*-DPE, green); and after subsequent visible light irradiation at 435 nm for 15 mins (*trans*-DPE, red).

4. STM image of DPE

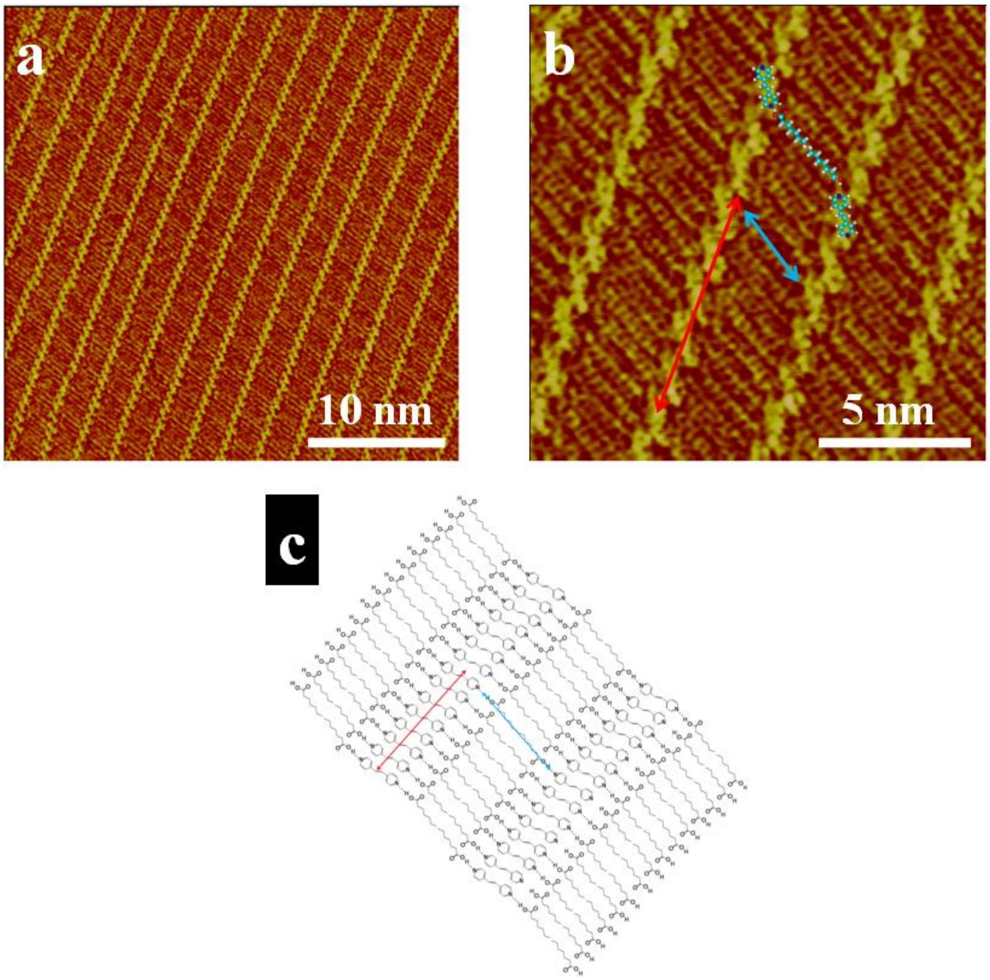

Figure S4. Large scale (a) and high resolution (b) STM image of DPE in heptanoic acid, I_{set} = 320.4 pA, V_{bias} = 548.4 mV. (c) Suggested molecular model for the self-assembled strcture observed in (b). On HOPG surface, DPE molecules form close-packed monolayers through π-π stacking interaction between DPE molecules (along the red arrows) and hydrogen bonds between DPE molecules and heptanoic acid (along the blue arrows). We have tried to perform the isomerization of DPE at the HOPG/hepatonic acid interface, however, we could not obtain any isomerized self-assembled structures. And we think that the π-π stacking interaction between isomerized DPE molecules and hydrogen bonds between isomerized DPE molecules and hepatonic acid become weak because there is strong steric resistance of *cis*-DPE molecules. As a result, the *cis*-DPE molecules could not form the stable structure on graphite surface.

5. STM image of NN4A after successive irradiation by UV light and visible light on HOPG

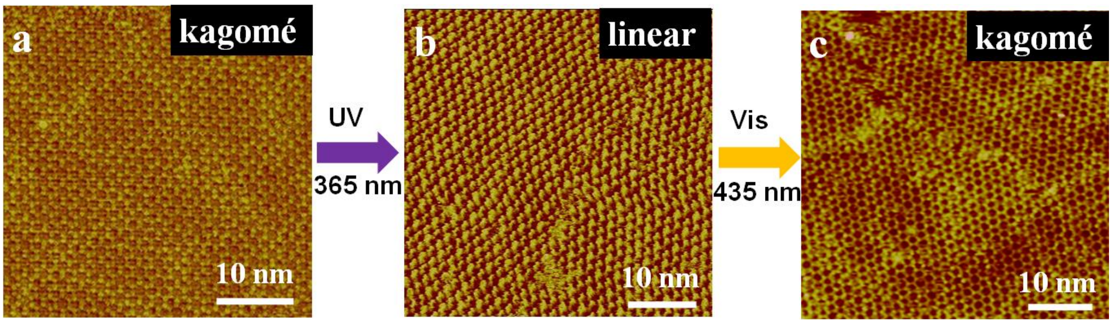

Figure S5. STM images for the photo-cycled self-assembled structures of NN4A in heptanoic acid. (a) *trans*-NN4A (before irradiation), I_{set} = 299.9 pA, V_{bias} = 599.9 mV. (b) *cis*-NN4A (*trans*-NN4A was irradiated by UV light at 365 nm for 30 mins), I_{set} = 323.5 pA, V_{bias} = 658.0 mV. (c) *retrans*-NN4A (*cis*-NN4A was subsequently irradiated by visible light at 435 nm for 15 mins), I_{set} = 311.3 pA, V_{bias} = 616.8 mV.

As shown in Figure S5, the assembled structure of the NN4A could be transformed from the Kagomé networks to the linear characteristics, and then to the Kagomé networks again after successive irradiation by UV light and visible light.

6. Assembled structure of the *cis*-NN4A/*trans*-DPE and its photoisomerization on HOPG

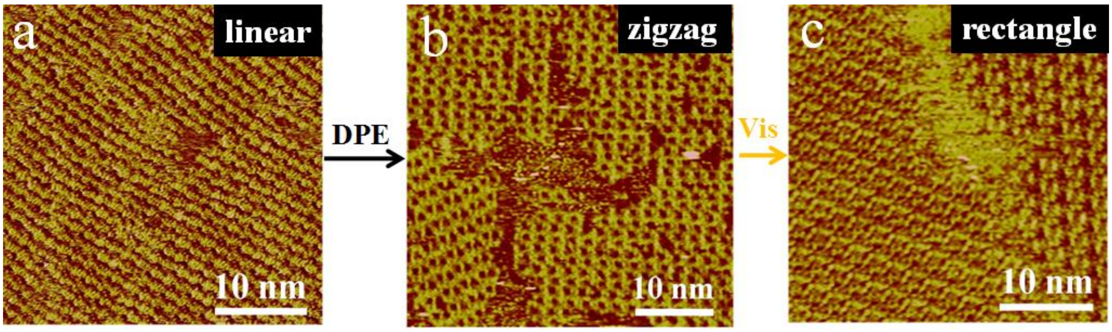

Figure S6. (a) STM image of the *cis*-NN4A assembled structures (NN4A irradiated by UV light for 30 min), I_{set} = 300 pA, V_{bias} = 650 mV. (b) The assembled structure of the *cis*-NN4A/*trans*-DPE (adding some solution containing *trans*-DPE onto the assemblies of the *cis*-NN4A), I_{set} = 300 pA, V_{bias} = 600 mV. (c) STM image of the NN4A/DPE upon irradiation with visible light for 15mins, I_{set} = 310.5 pA, V_{bias} =790 mV.

As shown in Figure S6, the linear structure could be replaced by the zigzag architecture after adding some *trans*-DPE onto the assembled structure of the *cis*-NN4A molecule, which demonstrates that the zigzag structure is formed by the *cis*-NN4A and the *trans*-DPE. Besides, the zigzag characteristics could be transformed to the rectangular networks after irradiation by visible light.