## Electronic Supplementary Information for

## Two-dimensional self-assembly and co-assembly of two

## tetracarboxylic acid derivatives investigated by STM

Xuan Peng,<sup>a, d, \xi</sup> Linlin Gan,<sup>a, c, ξ</sup> Wenchao Zhai,<sup>d</sup> Xiaoling Chen,<sup>d</sup> Ke Deng,<sup>\*a</sup> Wubiao Duan,<sup>\*c</sup> Wei Li,<sup>\*d</sup> Qingdao Zeng<sup>\*a, b</sup>

- a. CAS key Laboratory of Standardization and Measurement for Nanotechnology, CAS Center for Excellence in Nanoscience, National Center for Nanoscience and Technology (NCNST), Beijing 100190, China.
- b. Center of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, China.
- c. Department of Chemistry, School of Science, Beijing Jiaotong University, Beijing 100044, China.
- d. School of Science, Nanchang Institute of Technology, Nanchang 330099, China.

\*Corresponding author E-mail: kdeng@nanoctr.cn; liweidting@nit.edu.cn; wbduan@bjtu.edu.cn; zengqd@nanoctr.cn.

 $\xi$  Xuan Peng and Linlin Gan contributed to this work equally.



**Fig. S1** Large-scale STM image of H<sub>4</sub>BDETP's self-assembled nanostructure at the 1-heptanoic acid/HOPG interface with the tunneling conditions of  $I_{set}$  = 216.7 pA,  $V_{bias}$  = 707.4 mV.



**Fig. S2** (a), (b) Large-scale STM images of H<sub>4</sub>BTB's two self-assembled nanostructure (lamellar nanostructure in domain I and tetragonal nanostructure in domain II) at the 1-heptanoic acid/HOPG interface, both the tunneling conditions were:  $I_{set}$  = 363.2 pA,  $V_{bias}$  = 668.0 mV.



**Fig. S3** Large-scale STM images of two-component system prepared by dropping  $H_4BTB$  to pre-assembled  $H_4BDETP$  with different molar ratios: (a)  $H_4BDETP$ :  $H_4BTB > 2:1$ ,  $I_{set} = 219.7 \text{ pA}$ ,  $V_{bias} = 655.2 \text{ mV}$ ; (b)  $H_4BDETP$ :  $H_4BTB = 2:1$ ,  $I_{set} = 265.5 \text{ pA}$ ,  $V_{bias} = 754.4 \text{ mV}$ ; (c)  $H_4BDETP$ :  $H_4BTB < 2:1$ ,  $I_{set} = 174.0 \text{ pA}$ ,  $V_{bias} = 551.5 \text{ mV}$ . (Domain I:  $H_4BDETP/H_4BTB$  co-assembly structure. Domain II: self-assembly structure of  $H_4BDETP$ . Domain III: self-assembly structure of  $H_4BTB$ )



**Fig. S4** Large-scale STM images of assembled structures prepared by dropping H<sub>4</sub>BDETP to pre-assembled H<sub>4</sub>BTB with different molar ratios: (a) H<sub>4</sub>BDETP: H<sub>4</sub>BTB > 2:1,  $I_{set}$  =296.0 pA,  $V_{bias}$  = 578.9 mV; (b) H<sub>4</sub>BDETP: H<sub>4</sub>BTB = 2:1,  $I_{set}$  = 296.0 pA,  $V_{bias}$  = 578.9 mV; (c) H<sub>4</sub>BDETP: H<sub>4</sub>BTB < 2:1,  $I_{set}$  = 299.8 pA,  $V_{bias}$  = 699.8 mV. (Domain II: self-assembly structure of H<sub>4</sub>BDETP. Domain III: self-assembly structure of H<sub>4</sub>BTB)



**Fig. S5** Large-scale STM images of two-component system prepared by pre-mixing  $H_4BTB$  and  $H_4BDETP$  solution: (a)  $I_{set} = 241.1$  pA,  $V_{bias} = 846.3$  mV, (b)  $I_{set} = 335.7$  pA,  $V_{bias} = 660.4$  mV. The concentration of  $H_4BTB$  increased from (a) to (b).