

## Supporting Information

### Single-Molecule Observation of the K<sup>+</sup>-Induced Switching of Valinomycin within a Template Network

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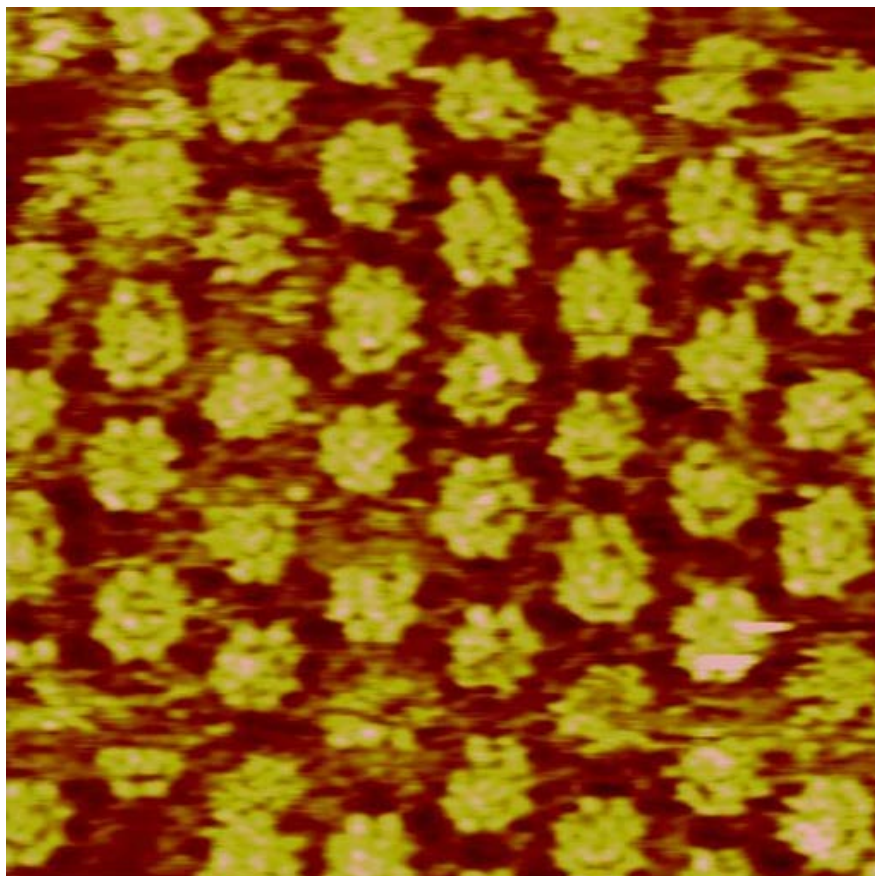
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#### 1. Experimental Section

The synthesis of oligoamide macrocycle has been described previously.<sup>1-2</sup> Valinomycin was purchased from Acros with purity > 99%. The macrocycle was dissolved in chloroform, and then a droplet of the solution was deposited on freshly cleaved graphite substrate (HOPG, grade ZYB) to form the self-assembled monolayers (SAMs). A droplet of valinomycin ethanol solution was dropped on the HOPG surface with the existing oligoamide macrocycle SAMs. After ethanol was evaporated, the STM experiments were performed at the liquid/solid interface after application of 0.5 μL of 1-phenyloctane as accessory solvent to the existing surface assemblies. The valinomycin ethanol solution was mixed with KCl aqueous solution prior to the STM experiments. The sample preparations for the K<sup>+</sup> complex in the STM experiments follow the same procedure as that for valinomycin. Tips were mechanically formed from Pt/Ir wires (80/20). STM images were acquired using the Nanoscope IIIa scanning probe microscopy system (Veeco Metrology). All STM images were recorded in constant current mode. The tunneling conditions are described in the corresponding figure captions.

1. L. H. Yuan, W. Feng, K. Yamato, A. R. Sanford, D. G. Xu, H. Guo, B. Gong, *J. Am. Chem. Soc.*, 2004, **126**, 11120.
2. W. Feng, K. Yamato, L. Q. Yang, J. S. Ferguson, L. J. Zhong, S. L. Zou, L. H. Yuan, X. C. Zeng, B. Gong, *J. Am. Chem. Soc.*, 2009, **131**, 2629.

#### 2. STM image at negative bias condition



**Figure S1.** (a) The STM image of cyclo[16]aramide and valinomycin-K<sup>+</sup> complex (35.4 nm × 35.4 nm,  $I = 452.6$  pA,  $V = -735.2$  mV).