

*Electronic Supplementary Information for*

**Fine-Tuning of Two-Dimensional Metal-Organic Nanostructures *via*  
Alkali-Pyridyl Coordination**

Kun Zhou,<sup>a</sup> Huifang Liang,<sup>b</sup> Miao Wang,<sup>c</sup> Shuaipeng Xing,<sup>a</sup> Honghe Ding,<sup>d</sup> Yang Song,<sup>a</sup>  
Yuxu Wang,<sup>a</sup> Qian Xu,<sup>d</sup> Jing-Hui He,<sup>e</sup> Junfa Zhu,<sup>d</sup> Wei Zhao,<sup>\*,c</sup> Yu-qiang Ma<sup>\*,a,f</sup> and  
Ziliang Shi<sup>\*,a</sup>

**Affiliations:**

<sup>a</sup> Center for Soft Condensed Matter Physics & Interdisciplinary Research, School of Physical Science and Technology, Soochow University, 215006 Suzhou, China

<sup>b</sup> Institute of Functional Nano & Soft Materials (FUNSOM), Soochow University, Suzhou 215123, China

<sup>c</sup> Institute for Advanced Study, Shenzhen University, 518060 Shenzhen, China

<sup>d</sup> National Synchrotron Radiation Laboratory, University of Science and Technology of China, 230029 Hefei, China

<sup>e</sup> College of Chemistry, Chemical Engineering and Materials Science, Collaborative Innovation Center of Suzhou Nano Science and Technology, Soochow University, 215123 Suzhou, China

<sup>f</sup> National Laboratory of Solid State Microstructures and Department of Physics, Nanjing University, 210093 Nanjing, China

**\*Correspondence to:**

weizhao@szu.edu.cn

myqiang@nju.edu.cn

phzshi@suda.edu.cn

## I. Additional STM images

Consecutive deposition of NaCl and TPyP on Ag(111) reproduced a similar Na-TPyP chessboard (CB) structure.

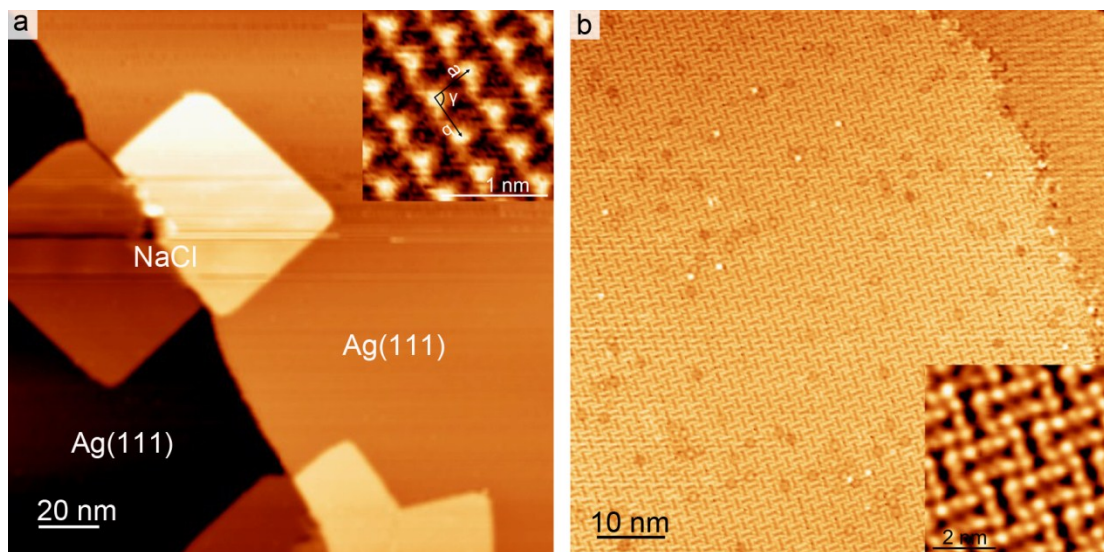


Figure S1. (a) STM overview of the NaCl(100) islands on Ag(111) and high-resolution image (inset) of a NaCl single layer. Unit cell parameters:  $a=0.40\pm 0.02$  nm,  $b=0.40\pm 0.02$  nm,  $\gamma=90^\circ$ . (b) STM overview of Na-TPyP CB structure, coexisting with TPyP close-packing (CP) monolayers (upper-right), resulting from the deposition of TPyP on the Ag(111) surface with NaCl pre-adsorbed. Inset: high-resolution image of the CB structure. Data acquisition conditions: (a) and inset,  $U=-1.2$  V,  $I=50$  pA; (b) and inset,  $U=-0.9$  V,  $I=30$  pA.

## II. XP spectra of Cl 2p.

To measure the evolutions of the XP spectra for Cl 2p, we have prepared a sample where NaCl and TPyP were consecutively evaporated on a clean Ag(111) surface. A post-annealing treatment at 420 K was applied to promote the self-assembly process. The XP spectra of the Cl 2p were measured using a photon energy of 350 eV. Similar with the Na 2p signals for the NaCl (see Fig. 2a in the main text), there are two sets of Cl 2p<sub>3/2</sub> and 2p<sub>1/2</sub> peaks visible for the NaCl submonolayers on Ag(111) due to the double layered structure (dipole) of NaCl (Fig. S2, top). The Cl 2p<sub>3/2</sub> peak at the binding energy of 199.0 eV is attributed to NaCl on top layers, in agreement with the reported value measured for the NaCl submonolayers on other metal surfaces.<sup>1,2</sup> The other Cl species at higher BE is attributed to the 1<sup>st</sup> layer NaCl on Ag(111), and the smaller intensity is due to the damping effect. Upon the deposition of TPyP, a new Cl 2p<sub>3/2</sub> peak appears at BE=197.4 eV (Fig. S2, middle), assigned to the Cl adsorbed on the Ag(111) surface.<sup>3</sup> The emergence of the new Cl species (labeled Cl<sub>-Ag</sub>) reveals that NaCl is dissociated due to the surface reaction between NaCl and TPyP molecules. As shown in the bottom panel of Fig. S2, a further increment of Cl<sub>-Ag</sub> following the thermal annealing of the sample agrees with the evolutions of Na 2p XP spectra and our STM observations.

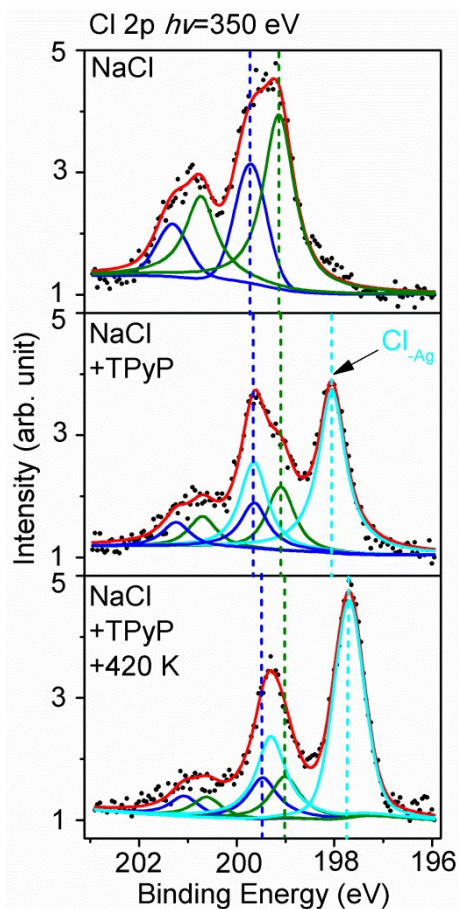


Figure S2. Evolutions of XP spectra of Cl. The spectra are denoted with different colors: Cl in NaCl submonolayers, blue and green; Cl binding with Ag atoms of the substrate, cyan.

## References

- 1 T. K. Shimizu, J. Jung, H. Imada and Y. Kim, *Angew. Chem., Int. Ed.*, 2014, **53**, 13729-13733.
- 2 D. Skomski, S. Abb and S. L. Tait, *J. Am. Chem. Soc.*, 2012, **134**, 14165-14171.
- 3 H. Piao, K. Adib and M. A. Barteau, *Surf. Sci.*, 2004, **557**, 13-20.