Supplementary Materials for

On-surface construction of metal-organic Sierpiński triangle

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Experimental Section

All the STM experiments were carried out in a UHV chamber (base pressure 1×10^{-10} mbar) equipped with a variable-temperature "Aarhus-type" STM,[1,2] a molecular evaporator and standard facilities for sample preparation. The TPDCN molecule (chemical name: [1,1':3',1"-terphenyl]-4,4"-dicarbonitrile) which was prepared according to the literature[3] was deposited by thermal sublimation onto the Au(111) substrate. The chemicals for the synthesis of TPDCN was purchased form Adamas. Ni atom was evaporated by resistively heating a Nickel filament of high purity (99.994%; purchased from Alfa Aesar). The STM measurements were performed in a typical temperature range of 100 K−150 K unless specified, and the typical scanning parameters: $I_t = 0.5 \sim 1.0$ nA, $V_t = \pm 1000 \sim 2000$ mV.

The density functional theory (DFT) calculations were performed by using VASP code.[4,5] The projector augmented wave method was used to describe the interaction between ions and electrons,[6,7] and the PBE exchange−correlation functional was employed,[8] van der Waals corrections to the PBE density functional were also included using the DFT-D2 method of Grimme.[9] The atomic structures were relaxed until the forces on all unconstrained atoms were ≤0.03 eV/Å.
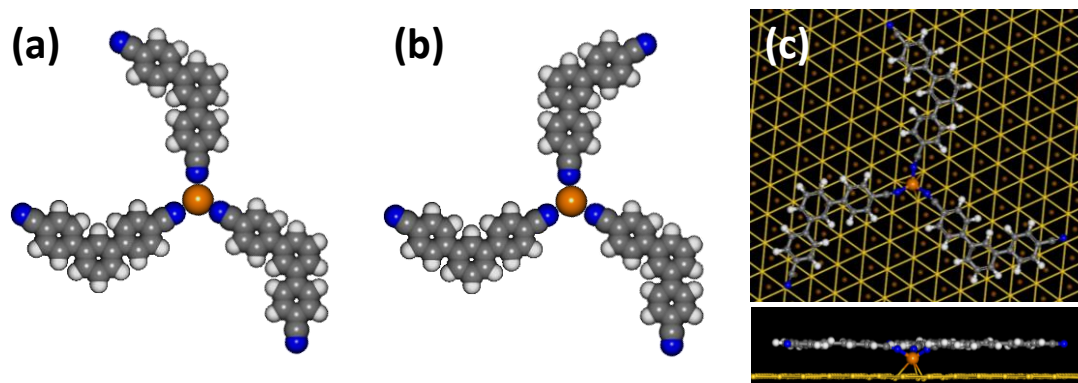

Figure S1. Structural models of (a) heterotactic and (b) homotactic elementary coordination motifs. (c) Top and side views of the DFT optimized model of the homotactic motif on Au(111).
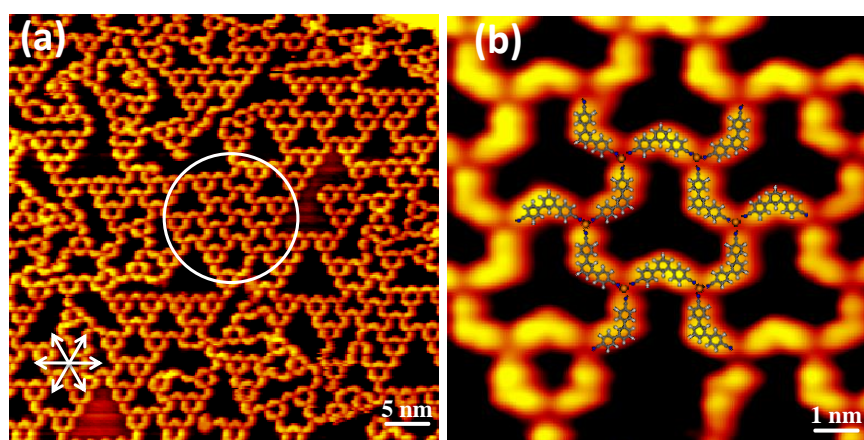

Figure S2. (a) STM image at high molecular coverages, a small region of the structures formed by the homotactic elementary motifs is indicated by white circle. (b) Close-up STM image of the region highlighted in (a) with the proposed model.
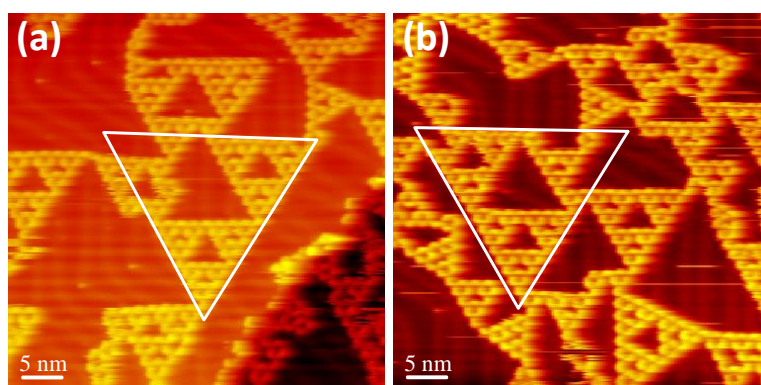
Figure S3. STM images showing the incomplete third generation Sierpiński triangle. Defects increase when growing into molecular Sierpiński triangle with higher generation. The "strict structural requirements" in the main text refer to several points including but not limited to the complete heterotactic arrangements, appropriate TPDCN/Ni ratio to achieve the 3 TPDCN and 1 Ni coordination motifs as well as the stronger local coordination bonding to the substrate.

Reference:

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