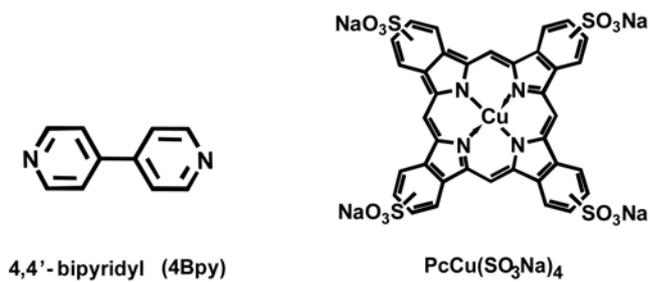


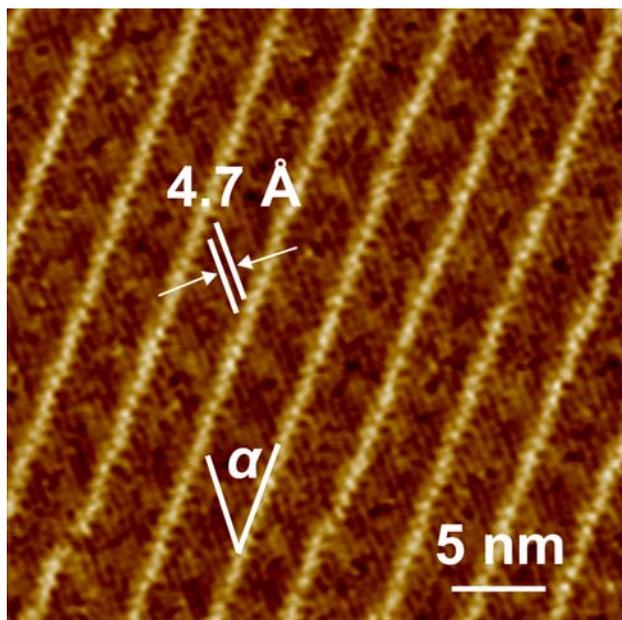
Experimental Details

The lyophilized powder of polyamino acids, solid powder of 4Bpy and $\text{PcCu}(\text{SO}_3\text{Na})_4$ were dissolved respectively in MilliQ water (the concentrations of 4Bpy and $\text{PcCu}(\text{SO}_3\text{Na})_4$ are 6.4 mM and 1.0 mM respectively, and the molar ratio of 4Bpy to polypeptide in solution is approximately 4 : 1) and mixed together. The co-assembly of polypeptide-4Bpy- $\text{PcCu}(\text{SO}_3\text{Na})_4$ was prepared by depositing a drop of the mixed aqueous solution directly onto the surface of freshly cleaved HOPG. After 1 minute deposition, the excess solution was blown away from the HOPG surface and the surface are blown dry by using high purity nitrogen gas. The as-prepared sample was scanned by STM under ambient conditions. STM experiments were performed in constant-current mode (Nanoscope IIIA system, Veeco, USA). The tips were newly mechanically formed Pt/Ir wire (80/20). The STM tunneling conditions are described in the corresponding figure captions. Experiments were repeated independently with different tips for reproducibility.

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Scheme S1 Molecular structures of 4,4'-bipyridyl (4Bpy) and copper phthalocyanine tetrasulfonate sodium ($\text{PcCu}(\text{SO}_3\text{Na})_4$).



15 Fig. S1 The STM image of polyQ₇ co-assembly with 4Bpy. The angle (α) between peptide molecular axes and the stripe directions are measured to be $46 \pm 2^\circ$. The distance between the neighboring polyQ₇ molecules is $4.7 \pm 0.2 \text{ \AA}$ (marked by white bars). Tunneling conditions: $I = 343.2 \text{ pA}$, $V = 598.5 \text{ mV}$.

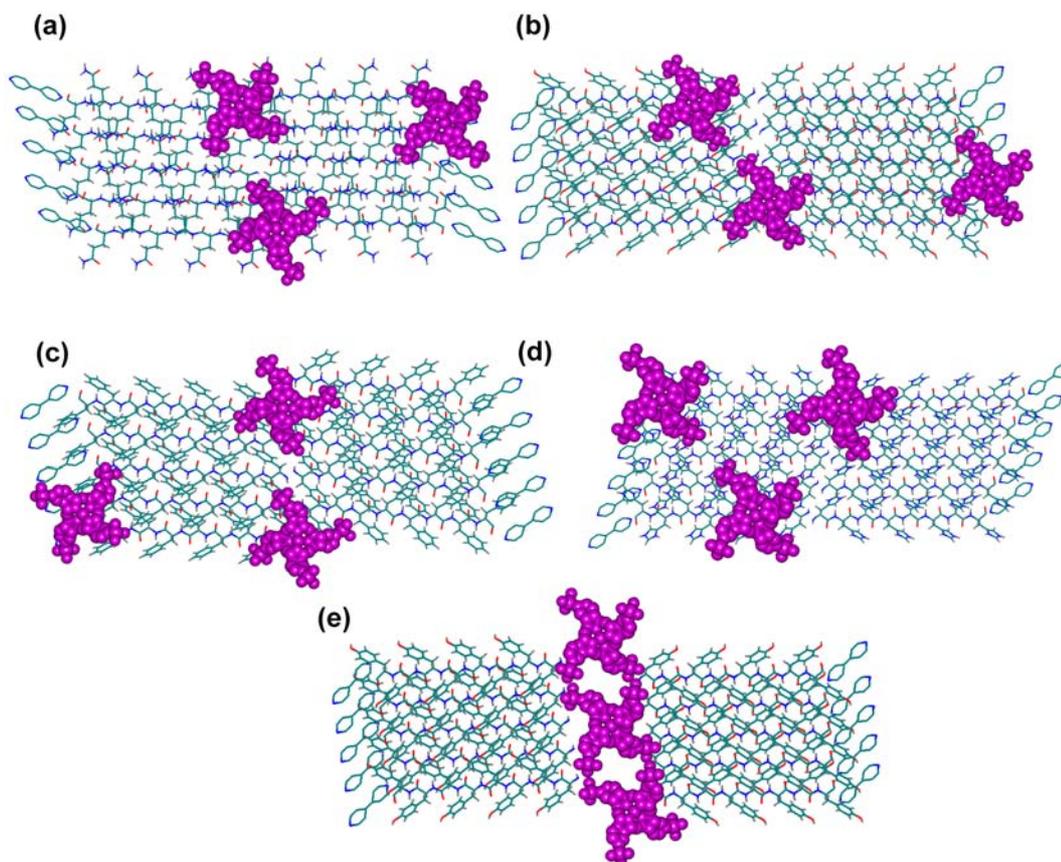


Fig. S2 The proposed structural models of $\text{Cu}(\text{SO}_3\text{Na})_4$ binding on different kinds of peptides-4Bpy co-assemblies derived from STM. The models for 20 $\text{Cu}(\text{SO}_3\text{Na})_4$ binding on polyQ₇-4Bpy (a), polyY₈-4Bpy (b), polyF₈-4Bpy (c), polyH₈-4Bpy (d) co-assemblies, and the model for $\text{PcCu}(\text{SO}_3\text{Na})_4$ array co-adsorbed with polyY₈ stripes denoted as site III (e). $\text{Cu}(\text{SO}_3\text{Na})_4$ molecules are depicted in violet. Color code: cyan for C, white for H, blue for N, and red for O.

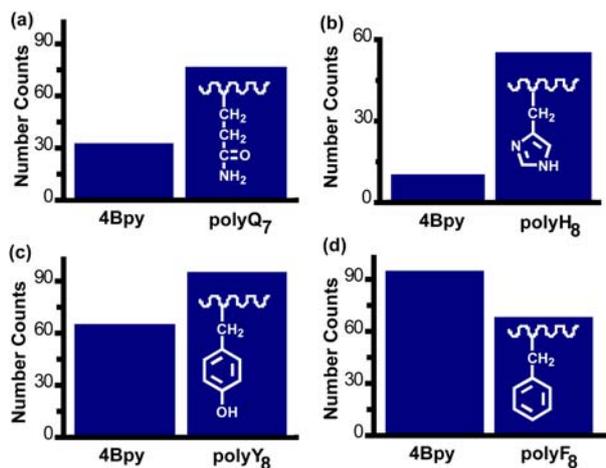


Fig. S3 The statistical result of $\text{PcCu}(\text{SO}_3\text{Na})_4$ binding on different sites derived from STM. (a)-(d) The number of $\text{PcCu}(\text{SO}_3\text{Na})_4$ adsorption on different 25 sites on (a) polyQ₇-4Bpy, polyH₈-4Bpy (b), polyY₈-4Bpy (c), and polyF₈-4Bpy (d) co-assemblies.

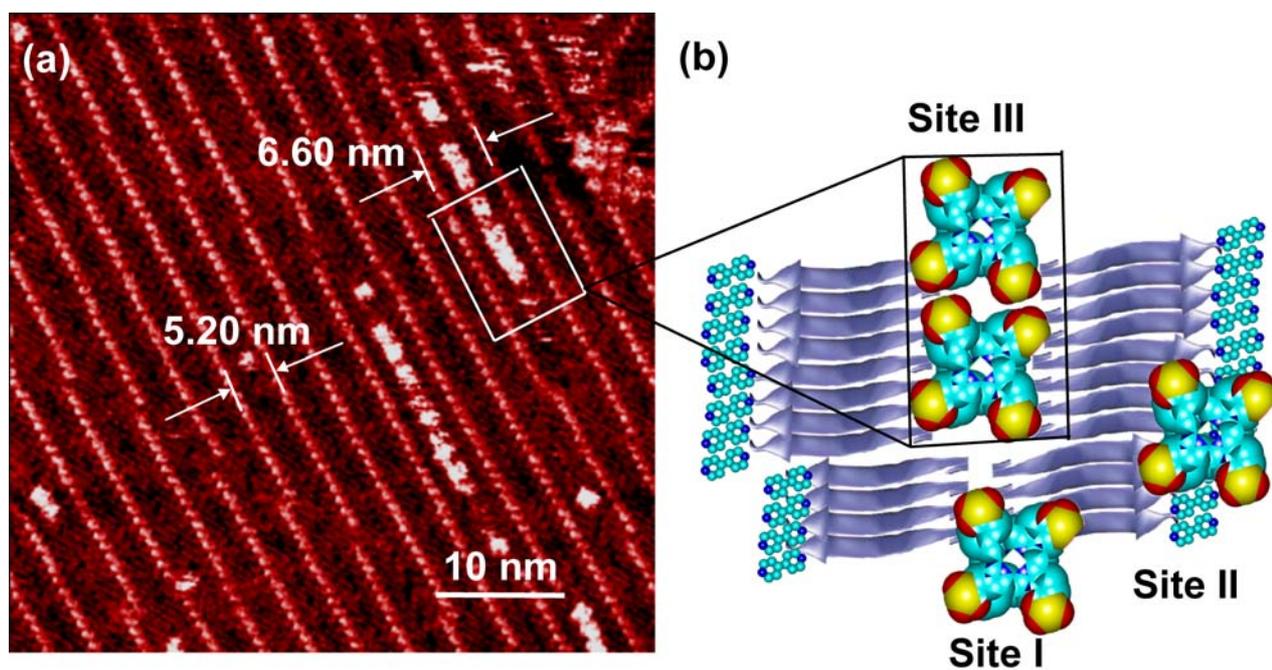


Fig. S4 $\text{PcCu}(\text{SO}_3\text{Na})_4$ adsorption on polyY₈-4Bpy co-assembly identified by STM image. (a) The STM image of $\text{PcCu}(\text{SO}_3\text{Na})_4$ adsorption on polyY₈-4Bpy co-assembly. The domain highlighted by white rectangle is the adsorbed $\text{PcCu}(\text{SO}_3\text{Na})_4$ arrays interacting with N termini of polyY₈ peptides. (b) Proposed model for the $\text{PcCu}(\text{SO}_3\text{Na})_4$ adsorption sites. Sites I and II present adsorption atop peptides and atop 4Bpy respectively. Site III pointed out by the black rectangle presents the binding mode with insertion of $\text{PcCu}(\text{SO}_3\text{Na})_4$ array between polyY₈ rows. Tunneling conditions: (a) $I = 612.2 \text{ pA}$, $V = 311.0 \text{ mV}$.