**Experimental Details**

The lyophilized powder of polyamino acids, solid powder of 4Bpy and PcCu(SO$_3$Na)$_4$ were dissolved respectively in MilliQ water (the concentrations of 4Bpy and PcCu(SO$_3$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-PcCu(SO$_3$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.

![Scheme S1](image1.png)

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

![Fig. S1](image2.png)

**Fig. S1** The STM image of polyQ$_7$ co-assembly with 4Bpy. The angle ($\alpha$) between peptide molecular axes and the stripe directions are measured to be 46 ± 2°. The distance between the neighboring polyQ$_7$ molecules is 4.7 ± 0.2 Å (marked by white bars). Tunneling conditions: $I = 343.2$ pA, $V = 598.5$ mV.
Fig. S2 The proposed structural models of Cu(SO$_3$Na)$_4$ binding on different kinds of peptides-4Bpy co-assemblies derived from STM. The models for 20 Cu(SO$_3$Na)$_4$ binding on polyQ-4Bpy (a), polyY$_8$-4Bpy (b), polyF$_8$-4Bpy (c), polyH$_8$-4Bpy (d) co-assemblies, and the model for PcCu(SO$_3$Na)$_4$ array co-adsorbed with polyY$_8$ stripes denoted as site III (e). Cu(SO$_3$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 PcCu(SO$_3$Na)$_4$ binding on different sites derived from STM. (a)-(d) The number of PcCu(SO$_3$Na)$_4$ adsorption on different sites on (a)polyQ-4Bpy, polyH$_8$-4Bpy (b), polyY$_8$-4Bpy (c), and polyF$_8$-4Bpy (d) co-assemblies.
Fig. S4  PcCu(SO₄Na)₄ adsorption on polyY₈-4Bpy co-assembly identified by STM image. (a) The STM image of PcCu(SO₄Na)₄ adsorption on polyY₈-4Bpy co-assembly. The domain highlighted by white rectangle is the adsorbed PcCu(SO₄Na)₄ arrays interacting with N termini of polyY₈ peptides. (b) Proposed model for the PcCu(SO₄Na)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 PcCu(SO₄Na)₄ array between polyY₈ rows. Tunneling conditions: (a) $I = 612.2 \text{ pA}$, $V = 311.0 \text{ mV}$. 