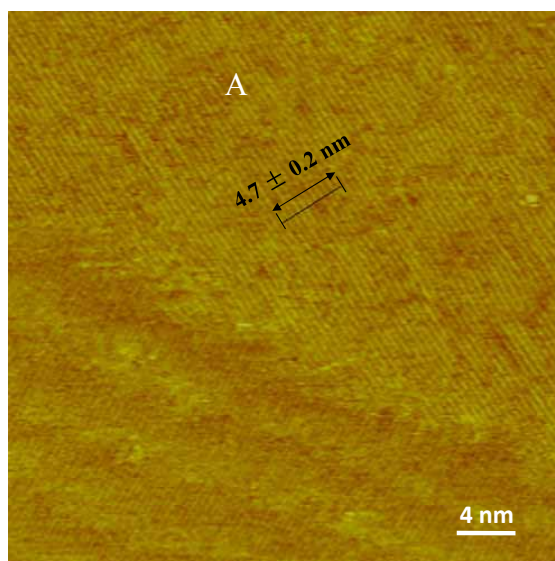
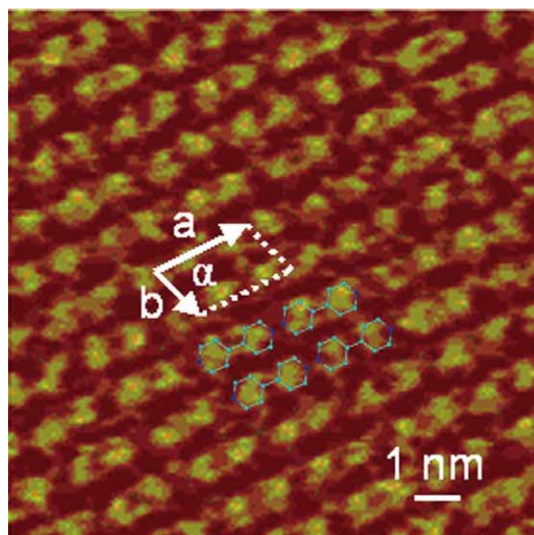


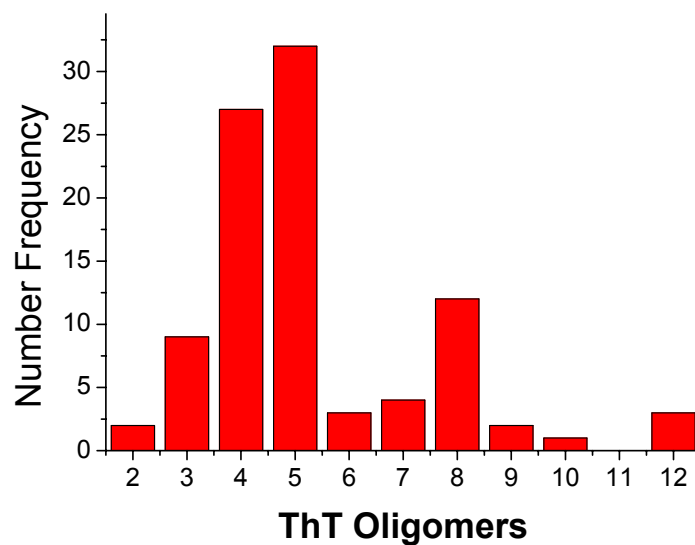
### Supporting Material



**Fig. S1** STM image of pentaalanine assembly. The distance marked covering 10 molecules is  $4.7 \pm 0.2$  nm. Tunneling conditions:  $I = 329.6$  pA,  $V = 880.1$  mV.



**Fig. S2** STM image of 4Bpy assembly with superimposed molecular model and unit cell.  $a = 2.10$  nm,  $b = 1.30$  nm  $\alpha = 81^\circ$ . Tunneling conditions:  $I = 326.5$  pA,  $V = 832.2$  mV.



**Fig. S3** The number distribution of ThT molecules in the oligomers bound to pentaalanine peptides.

## MATERIALS AND METHODS

### *Sample preparation for STM studies*

The STM experiments have been done with different solvents without any apparent difference in the binding modes. Since the similar results, the other solvents (MilliQ water, ethanol) have been deleted in the revised manuscript, and the acetonitrile has been identified as the representative solvent.