Supporting Information

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SI Text

STM Experimental Details. The STM experiments have been reproduced independently after changing the solvents (acetonitrile, Milli-Q water, and ethanol), peptide concentration (the final concentration ca. 1 mg/mL, 0.1 mg/mL), and the solvent evaporation temperature (at room temperature, and heated sample at 60 °C). The specific features of peptide strands in the STM images did have certain variability which is typical for STM tip fluctuations in combination with the molecular conformational fluctuations. The results on length distribution remained nearly the same at the different sample preparation conditions.

Statistical Methods. The lengths of the peptide strands in STM images are measured by using the Nanoscope software (Veeco). A length increment of 0.325 nm for every residue is assumed in the statistical histogram of the length distribution of peptide assemblies. The measured lengths in the histograms represent 300 measured values from the independent experiments. The number frequencies in the statistical results are all based on number of events. The assignments of most probable sites are determined from the peak distribution of peptide length histograms.



Fig. S1. The identification of hIAPP₈₋₃₇ peptide length in STM image. (A and C) The STM images of hIAPP₈₋₃₇ assembly (A) and the coassembly of hIAPP₈₋₃₇ and 4Bpy (C). The white line marks the length measurements of hIAPP₈₋₃₇ lamellae. Tunneling conditions: (A) I = 350.0 pA, V = 490.0 mV; (C) I = 314.3 pA, V = 605.2 mV. (*B* and *D*) Cross-sectional profiles corresponding to the white lines in *A* and *C*. The thick solid lines represent the averaged heights of the peptide strands, and the length measurements for the peptide strand are defined as the full widths at half averaged heights of the peptide strands. (*E* and *F*) The length distribution of hIAPP₈₋₃₇ with longer strands identified by STM. (*E*) The STM image of the coassembly of hIAPP₈₋₃₇ and 4Bpy. The angle α between hIAPP₈₋₃₇ molecular axes and the stripe direction is measured to be $34 \pm 2^{\circ}$. Tunneling conditions: I = 555.7 pA, V = 668.5 mV. (*F*) The length distribution histogram of the core structures of hIAPP₈₋₃₇. The average length is measured to be 7.7 ± 0.6 nm. The step size of the chart is 0.325 nm, which is equivalent to the separation of two neighboring residues in the parallel beta-sheet structure.



Fig. S2. The bimodal distributions of peptide lengths for rIAPP₈₋₃₇ R18H (*A*) and rIAPP₃₇₋₈ R18H (*B*) coassembled with 4Bpy identified by STM. The peptide length measurements are marked by white solid lines. Tunneling conditions: (*A*) I = 612.9 pA, V = 300.4 mV; (*B*) I = 575.9 pA, V = 334.4 mV.

Table S1. Statistical length distribution of the coassembly of $hIAPP_{8\!-\!37}$ and 4Bpy at different ranges

Range of peptide chain length/nm *	Possible numbers of amino acid †	Counts *	Percent (%) §		
4.225 ~ 4.550	14	176	58.7		
4.550 ~ 4.875	15	124	41.3		

*The range is defined by the minimum length and the maximum length measured from STM images using an increment of 0.325 nm (the separation between the two neighboring amino acids in the peptide chain structure).

^tThe possible numbers of residues are calculated by dividing the chain lengths by 0.325 nm.

*Counts indicate the total number of chains in the different measured length ranges.

[§]The percentage of peptide chains in the ranges are shown in the fourth column.

Table S2. Statistical length distribution of the coassembly of $hIAPP_{8-37}$ and 4Bpy with longer peptide length

Range of peptide chain length/nm *	Possible numbers of amino acid †	Counts *	Percent (%) §	
6.825 ~ 7.150	22	19	6.3	
7.150 ~ 7.475	23	70	23.3	
7.475 ~ 7.800	24	91	30.3	
7.800 ~ 8.125	25	73	24.3	
8.125 ~ 8.450	26	43	14.3	
8.450 ~ 8.775	27	4	1.3	

*The range is defined by the minimum length and the maximum length measured from STM images using an increment of 0.325 nm (the separation between the two neighboring amino acids in the peptide chain structure).

^tThe possible numbers of residues are calculated by dividing the chain lengths by 0.325 nm.

*Counts indicate the total number of chains in the different measured length ranges.

[§]The percentage of peptide chains in the ranges are shown in the fourth column.

Table S3. Statistical length distribution of the coassembly of hIAPP₃₇₋₈ and 4Bpy

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Range of peptide chain length/nm*	Possible numbers of amino acid †	Counts *	Percent (%) §
3.575 ~ 3.900	12	8	2.7
3.900 ~ 4.225	13	217	72.3
4.225 ~ 4.550	14	27	9.0
4.550 ~ 4.875	15	11	3.7
4.875 ~ 5.200	16	3	10.0
5.200 ~ 5.525	17	5	1.7
5.525 ~ 5.850	18	9	3.0
5.850 ~ 6.175	19	17	5.7
6.175 ~ 6.500	20	3	1.0

*The range is defined by the minimum length and the maximum length measured from STM images using an increment of 0.325 nm (the separation between the two neighboring amino acids in the peptide chain structure).

'The possible numbers of residues are calculated by dividing the chain lengths by 0.325 nm.

*Counts indicate the total number of chains in the different measured length ranges.

[§]The percentage of peptide chains in the ranges are shown in the fourth column.

Table S4. Statistical length distribution of the coassembly of rIAPP₈₋₃₇ and 4Bpy

Range of peptide chain length/nm*	Possible numbers of amino acid †	Counts *	Percent (%) §	
4.225 ~ 4.550	14	31	10.3	
4.550 ~ 4.875	15	203	67.7	
4.875 ~ 5.200	16	57	19.0	
5.200 ~ 5.525	17	9	3.0	

*The range is defined by the minimum length and the maximum length measured from STM images using an increment of 0.325 nm (the separation between the two neighboring amino acids in the peptide chain structure).

^tThe possible numbers of residues are calculated by dividing the chain lengths by 0.325 nm.

[†]Counts indicate the total number of chains in the different measured length ranges.

[§]The percentage of peptide chains in the ranges are shown in the fourth column.

Table S5. Statistical	length distributior	ı of the c	coassembly o	f rIAPP37_8	and 4Bpy

Range of peptide chain length/nm *	Possible numbers of amino acid †	Counts *	Percent (%) §	
2.925 ~ 3.250	10	8	2.7	
3.250 ~ 3.575	11	89	29.7	
3.575 ~ 3.900	12	108	36.0	
3.900 ~ 4.225	13	61	20.3	
4.225 ~ 4.550	14	23	7.7	
4.550 ~ 4.875	15	11	3.7	

*The range is defined by the minimum length and the maximum length measured from STM images using an increment of 0.325 nm (the separation between the two neighboring amino acids in the peptide chain structure).

⁺The possible numbers of residues are calculated by dividing the chain lengths by 0.325 nm.

⁺Counts indicate the total number of chains in the different measured length ranges.

[§]The percentage of peptide chains in the ranges are shown in the fourth column.

Tab	le S6	. Statistical	length	distri	bution	of t	he	coassem	bl	y of	[:] rIAPP _{8–37}	R18H a	and	4Bp	Ŋ
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Range of peptide chain length/nm *	Possible numbers of amino acid ⁺	Counts *	Percent (%) ^s	
3.900 ~ 4.225	13	58	19.3	
4.225 ~ 4.550	14	88	29.3	
4.550 ~ 4.875	15	29	9.7	
4.875 ~ 5.200	16	19	6.3	
5.200 ~ 5.525	17	50	16.7	
5.525 ~ 5.850	18	39	13.0	
5.850 ~ 6.125	19	17	5.7	

*The range is defined by the minimum length and the maximum length measured from STM images using an increment of 0.325 nm (the separation between the two neighboring amino acids in the peptide chain structure).

^tThe possible numbers of residues are calculated by dividing the chain lengths by 0.325 nm.

^{*}Counts indicate the total number of chains in the different measured length ranges.

[§]The percentage of peptide chains in the ranges are shown in the fourth column.

Table S7. Statistical length distribution of th	he coassembly of	rIAPP _{37–8} R18H an	d 4Bpy at different
ranges			

Range of peptide chain length / nm *	Possible numbers of amino acid $^{\scriptscriptstyle +}$	Counts *	Percent (%) §	
3.250 ~ 3.575	11	74	24.7	
3.575 ~ 3.900	12	48	16.0	
3.900 ~ 4.225	13	18	6.0	
4.225 ~ 4.550	14	12	4.0	
4.550 ~ 4.875	15	7	2.3	
4.875 ~ 5.200	16	20	6.7	
5.200 ~ 5.525	17	41	13.7	
5.525 ~ 5.850	18	36	12.0	
5.850 ~ 6.175	19	16	5.3	
6.175 ~ 6.500	20	5	1.7	
6.500 ~ 6.825	21	7	2.3	
6.825 ~ 7.150	22	6	2.0	
7.150 ~ 7.475	23	10	3.3	

*The range is defined by the minimum length and the maximum length measured from STM images using an increment of 0.325 nm (the separation between the two neighboring amino acids in the peptide chain structure).

^tThe possible numbers of residues are calculated by dividing the chain lengths by 0.325 nm.

^{*}Counts indicate the total number of chains in the different measured length ranges.

[§]The percentage of peptide chains in the ranges are shown in the fourth column.