Electronic Supplementary Material (ESI) for Soft Matter.
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Supporting Information

Atomic Force Microscopy (AFM)

AFM experiments were performed in air at room temperature (21 ± 1°, humidity 22%) using a Multimode SPM system with a Nanoscope VIII controller (Veeco Instruments Inc., Santa Barbara, CA). All the recorded AFM images consist of 512 x 512 pixels, and several images were obtained at separate locations across the mica surfaces to ensure a high degree of reproducibility of the recorded molecular nanostructures. All the AFM images were analyzed by means of the commercial Scanning Probe Image Processor (SPIP™) software. Samples are prepared by pouring 10 µl of the peptide solution on a freshly cleaved mica surface, rinsed with distilled water and dried with pressurized air.

Transmission Electron Microscopy (TEM)

TEM images were recorded with a Philips C20 transmission electron microscope. To prepare TEM samples, small aliquots of fiber solutions were deposited onto freshly glow-discharged copper grids covered by a thin carbon layer. A droplet of 3 µl of the fiber solution was deposited on the carbon film for absorption for 1 minute – the excess of fluid was blotted away. Grids were rinsed by applying some droplets of distilled water, followed by blotting. All samples for TEM measurements were negative stained with a solution 1% Phosphotungstic Acid (PTA) tuned at pH 7.4.

Synchrotron Radiation Circular Dichroism (SR-CD)

Synchrotron Radiation Circular Dichroism (SR-CD) spectra were collected on beamline CD1 at the ASTRID storage ring (ISA, Aarhus University, Denmark). The beam from CD1 (Miles2007, Miles2008) was polarized with a MgF_2 Rochon polarizer (B-Halle GmbH, Berlin) and a photo elastic modulator (Hinds, USA) produced alternating left and right handed circular polarized light. Samples were measured at concentrations of 1 mg/mL. The light passed though the sample and was detected by a photo multiplier tube (Type 9406B, ETL, UK). Spectra of the water were recorded for baseline subtraction. Samples were measured in 1 mm path-length Suprasil cells (Hellma GmbH). All sample and baseline spectra were collected 3 times with 1 nm step size and 2 seconds dwell time.

Peptides incubation

Monomers were purchased by CASLO Laboratory ApS (Lyngby, Denmark). Monomers were first let to dissolve one day at room temperature in a hexafluoro-2-propanol (HFIP) concentrated solution (2 mg of peptide in 200 µl of HFIP solution). Then, the peptide was incubated in an aqueous solution (in which the pH was adjusted by using HCl and NaOH) at room temperature and continually shaken.

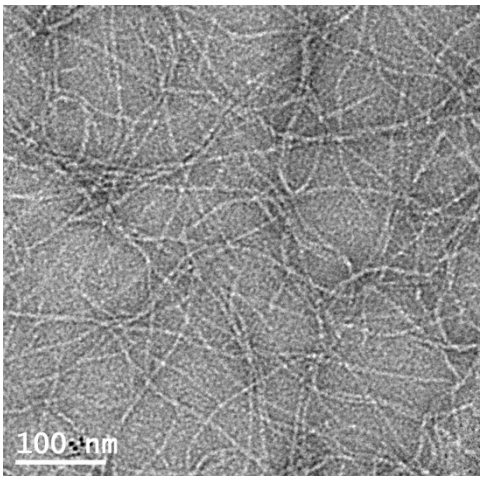

Fig. S1 TEM image of **P2** fibrils obtained at pH2 (corresponding to the AFM image of Fig.3a).

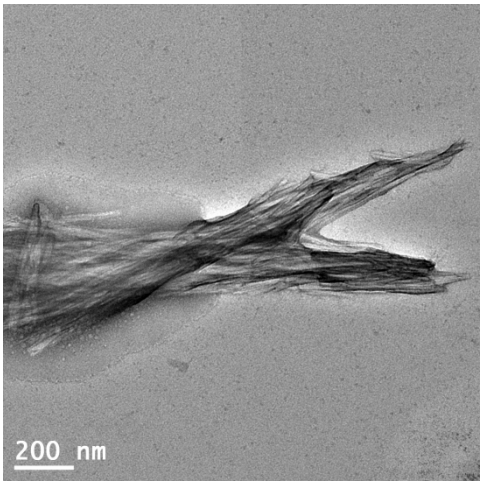

Fig. S2 TEM image of **P2** amorphous aggregates obtained at pH12 (corresponding to the AFM image of Fig.3c).

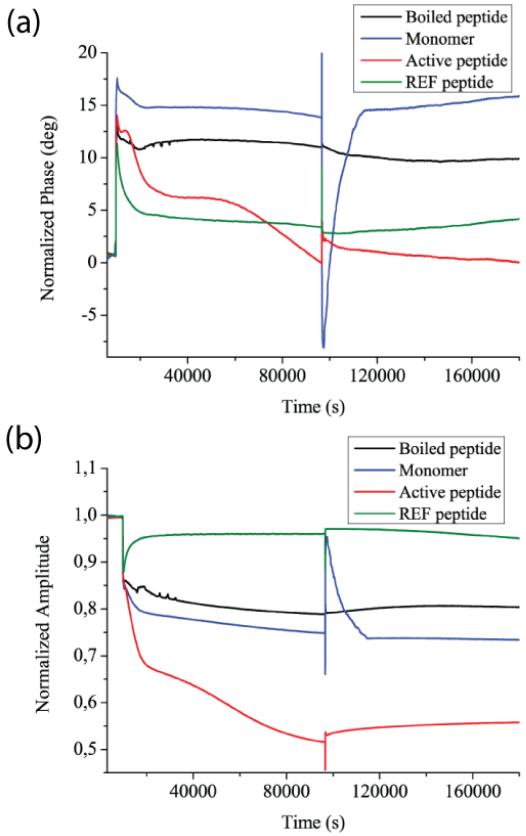

Fig. S3 SAW complete measurement of **P3**. Time is in abscissa and the variation of the phase is in ordinate. Color coded lines show the phase variation on chip surfaces with 4 different compounds. **P3** was injected into running ddH_2O directly followed by a 24 hour incubation period at 0 flow with the remaining **P3** solution until 90,000 s. Then, ddH_2O was run over the surface at 12.5 µl/min to compare dissociation or restructuring.

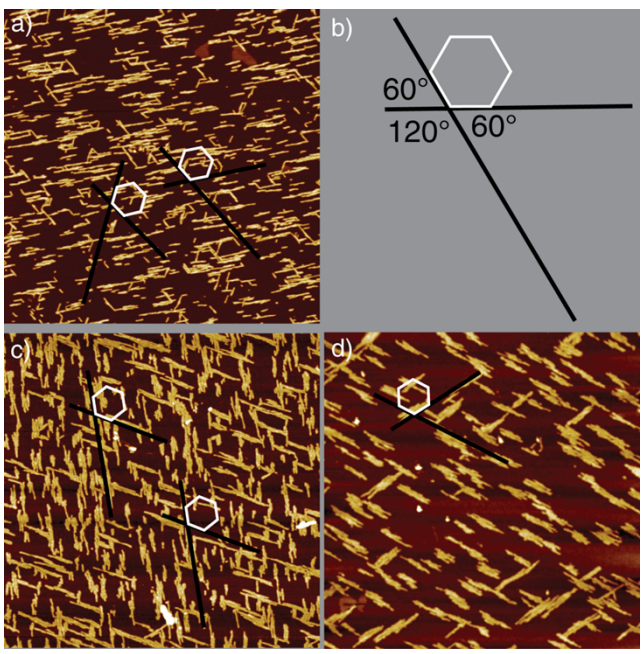

Fig. S4 AFM images exhibiting the hexagonal symmetry arrangement. A white hexagon and two black lines are placed on top of the AFM images to help indicating the underlying symmetry. a) **P2** sheet-like structures formed at pH 7; b) Schematic model of hexagonal symmetry; c) **P1** sheet-like structures formed at pH 2; d) Other example, but with different surface orientation of **P1** sheet-like structures.

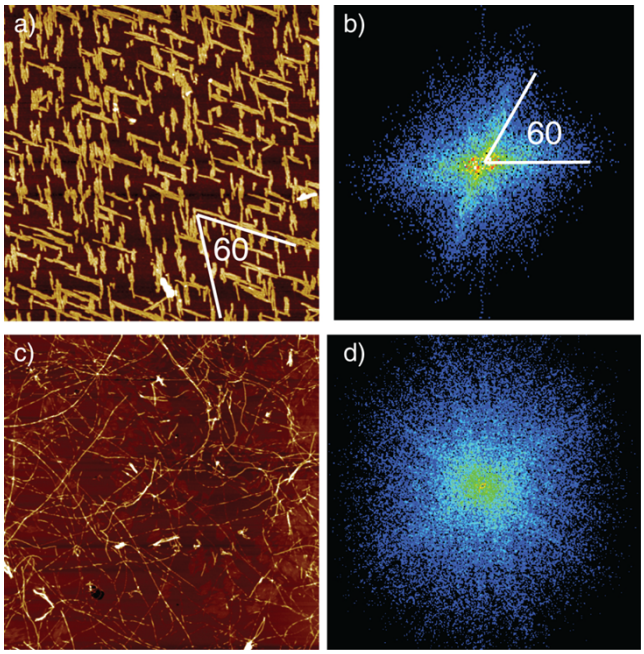

Fig. S5 AFM and Fourier transform images of **P1**: sheet-like structures (a) and Fourier transform (b), fibrils (c) and Fourier transform (d). Two white lines on (a) and (b) highlight the symmetry in real and Fourier space respectively, to support the hexagonal symmetry proposed in Fig. S4.

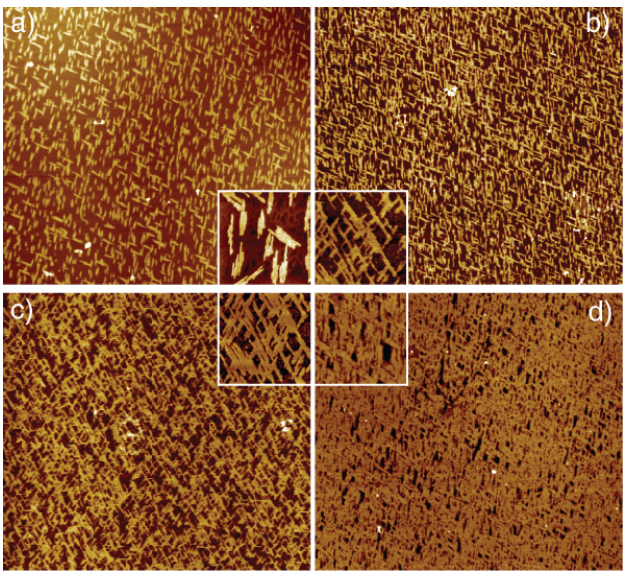

Fig. S6 AFM images of **P1** sheet-like structures showing the coverage dependence on the deposition time. a) 1 min; b) 3 min; c) 4 min; d) 5 min. All images are 10 x 10 µm^2; all insets are 0.5 x 0.5 µm^2.

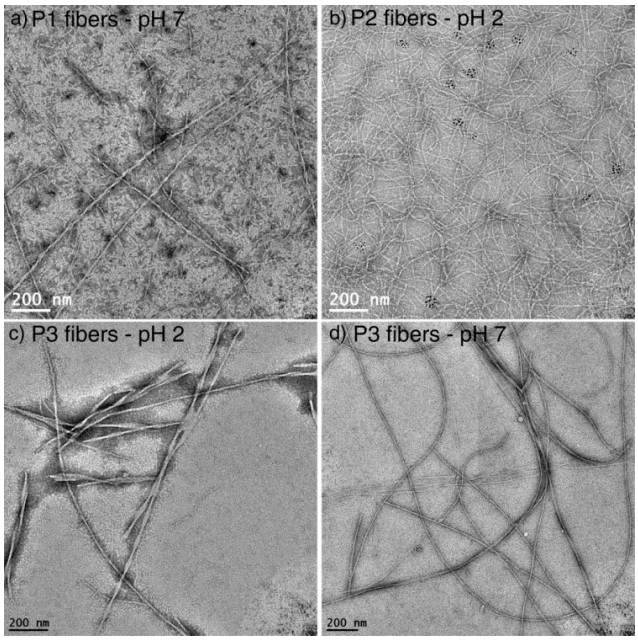

Fig. S7 TEM images of negative stained peptide fibrils; a) **P1** fibrils obtained at pH 7. On the background are visible other structures than fibrils – these have not been accounted in the coverage computation (see Fig. 2e in the manuscript); b) **P2** fibrils obtained at pH 2; c) **P3** fibrils obtained at pH 2; d) **P3** fibrils obtained at pH 7.

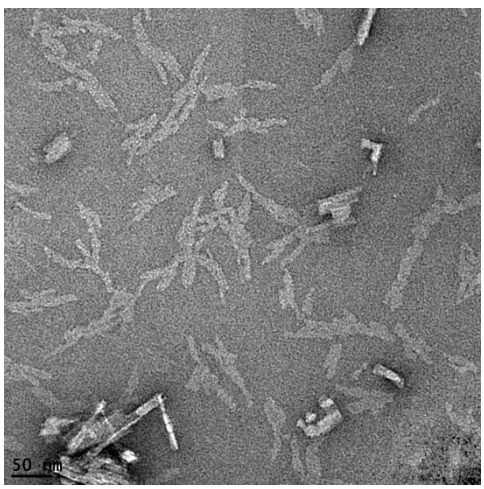

Fig. S8 TEM image of negative stained **P1** sheet-like structures formed at pH 2 (corresponding to Fig. 2a).

	Sequence	pH 2	pH 7
P1	SSFAFASSC	-	fibrils w = 20 ± 4 nm
P2	FASSSSFAC	fibrils w = 8 ± 2 nm	-
P3	SSSSFAFAC	fibrils w = 24 ± 4 nm	fibrils w = 16 ± 4 nm

Table S1 Fibrils width (w) values extracted by TEM images of Fig. S7.

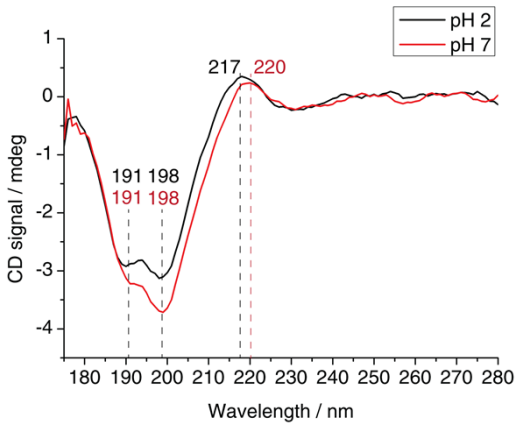

Fig. S9 Synchrotron Radiation Circular Dichroism (SR-CD) spectra of **P2** structures formed at pH 2 (fibrils of Fig. 3a) and pH 7 (sheet-like structures Fig. 3b). Spectra suggest the presence of random coil secondary structures both at pH 2 and pH 7.