## SUPPORTING INFORMATION: Surface Defect Sites Facilitate Fibrillation: Insight from Adsorption of Gold-binding peptide on Au(111)

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## **Experimental Section:**

Materials: Gold substrates Au(111) were purchased from Phasis Sarl. Stock solutions of A3 (1%) in water were stored at 4°C. Samples were diluted to desired concentrations in PBS buffer, pH of 7.5 and were kept frozen at -20° C. Gold substrate was immersed in peptide solution at room temperature for intended amount of adsorption time at desired concentration. Substrates were washed thoroughly with nanopure water for several minutes to remove any salt crystallines and dried with a stream of nitrogen.

AFM imaging: AFM imaging was performed on Dimension 3000 and Innova (Bruker) using silicon cantilevers with a spring constant of 40N/m within a week from sample preparation time. AFM imaging was performed in light tapping regime by maintaining the set point ratio greater than 0.95. At least 5 different areas of the sample were imaged to confirm the uniformity of the observed morphology of the peptides and acquire representative images.

SPR measurements: SPR was performed on a SensiQ dual channel SPR biosensing system using an unmodified bare Au SPR sensor. 100 µL of A3 peptide at 100 ng/mL, 1 ng/mL, 0.1 ng/mL, and 10 pg/mL was co-injected with water as a reference simultaneously through each channel and subtracted in real time. 10 pg/mL of A3 peptide was not detected. SPR measurements at each concentration was performed in triplicate.

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Figure S1. a) Topography image and b) Phase image of A3 at 20 minutes of adsorption on Au(111) from 100ng/ml A3 in PBS. c) Topography image and d) Phase image of A3 at 20 minutes of adsorption on Au(111) from 10  $\mu$ g/ml A3 in PBS. Z-scale: 2 nm in a,c and 12 degree in b,d. At high concentration regime, A3 fibers or coils have not been observed neither at early stages of adsorption nor later.



Figure S2. a) Topography of A3 fiber aggregates at a,b,c) 40 minutes d,e) 60 minutes and f) 80 minutes of adsorption on Au(111) from 0.1 nM A3 in PBS.



Figure S3. a) Topography image and b) Phase image of A3 at 100 minutes of adsorption on silicon from 0.1 nM A3 in PBS. Z-scale: 2 nm In a and 100 mV in b. The formation of A3 fibers and coils did not happen on Silicon substrate where A3 adsorption appeared to rely on non-specific interactions.



Figure S4. a) AFM images of 1 ng/ml gold binding peptide MHGKTQATSGTIQS on Au(111) which form fibrils and coils near grain boundary sites or step edges after 60 min of adsorption. Z-scale: 10 nm in a, 5 nm in b, 6 nm in c,d.