Self-Assembly of Ultralong Aligned Dipeptide Single Crystals

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Supporting Information

Results and Discussion

The influence of initial concentration of NH_4OH and temperature on the structures of FF fibers. The concentration of NH_4OH, solvent evaporation rate show great influence on the formation of FF fibers. We have made crystallization experiments over a broad range of concentrations of NH_4OH, from 0.006 to 14.80 mol/L. The obtained horizontally aligned fibers with the diameter of about 1-2 μm are obtained on the silicon wafer from 0.006 and 2.89 mol/L NH_4OH, however masses formed from 14.80 mol/L NH_4OH (**Figure S3b-d**). The different morphology of FF fibers may attribute to the different FF concentration when the fibers strat to grow. It takes different time for nucleation with different NH_4OH concentration of 10 min (0.006 mol/L), 30 min (2.89 mol/L) and 60 min (14.80 mol/L). In 0.006 and 2.89 mol/L NH_4OH, the solubility of FF reduces in the whole process with evaporation of solvent (**Figure S3a**). Nevertheless, in 14.80 mol/L NH_4OH, the solubility increases firstly and decrease, leading to long time for supersaturation. The FF concentration is pretty high and more FF fibers formed on the silicon wafer, which make the fibers amorphous (**Figure S3d**). Temperature can influence the evaporation speed. In 0.006 mol/L NH_4OH solution, when temperature is decreased to be 38 ^oC, the nucleation time of FF fibers on the scratch becomes longer (40 min) than that of at 75 ^oC (10 min) and less aligned FF fibers with the larger diameter of 2-3 μm form on the silicon wafer (**Figure S3a**). The FTIR and XRD analyses confirm the β-sheet secondary structure and highly crystalline character of all the FF fibers (**Figure S3e, f**). This

results indicate that our process makes the growth of aligned crystals with different structure controllable, which is great importance for their functional devices.

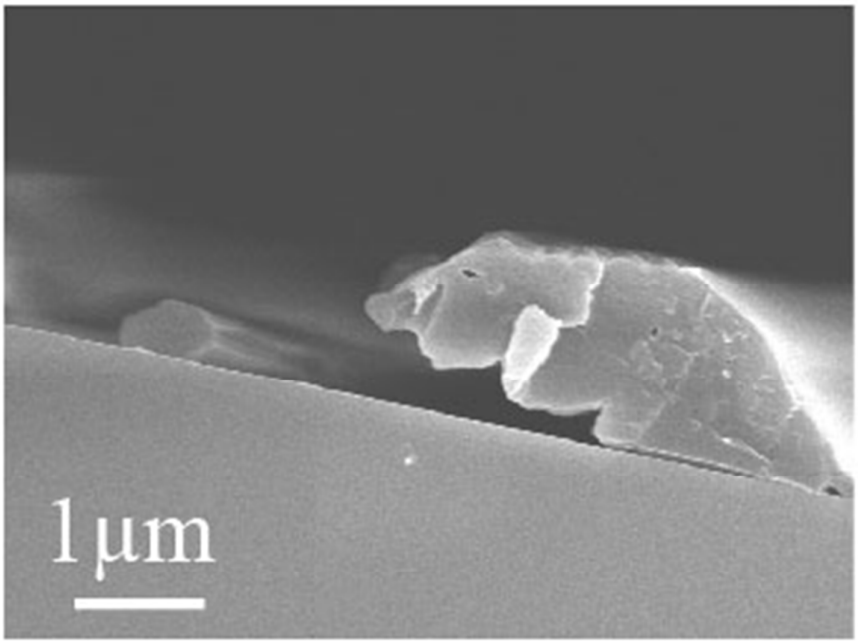

Figure S1. SEM image of a broken end of FF fibers.

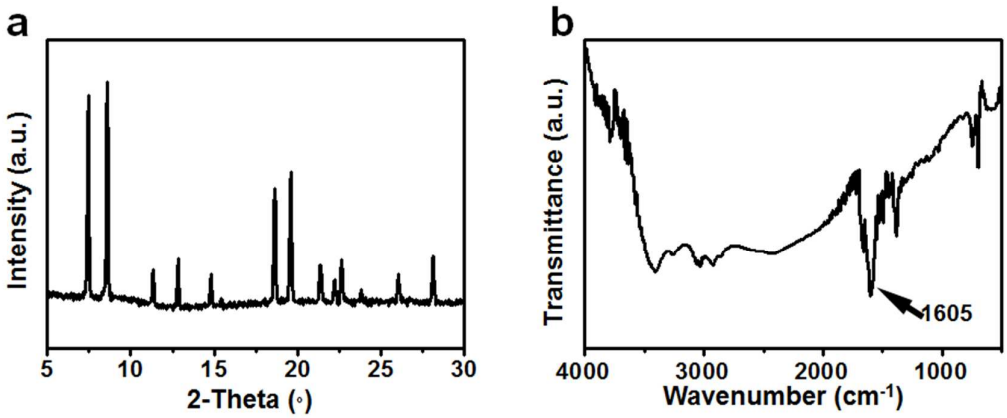

Figure S2. a) XRD and b) FTIR of powder of FF fibers.

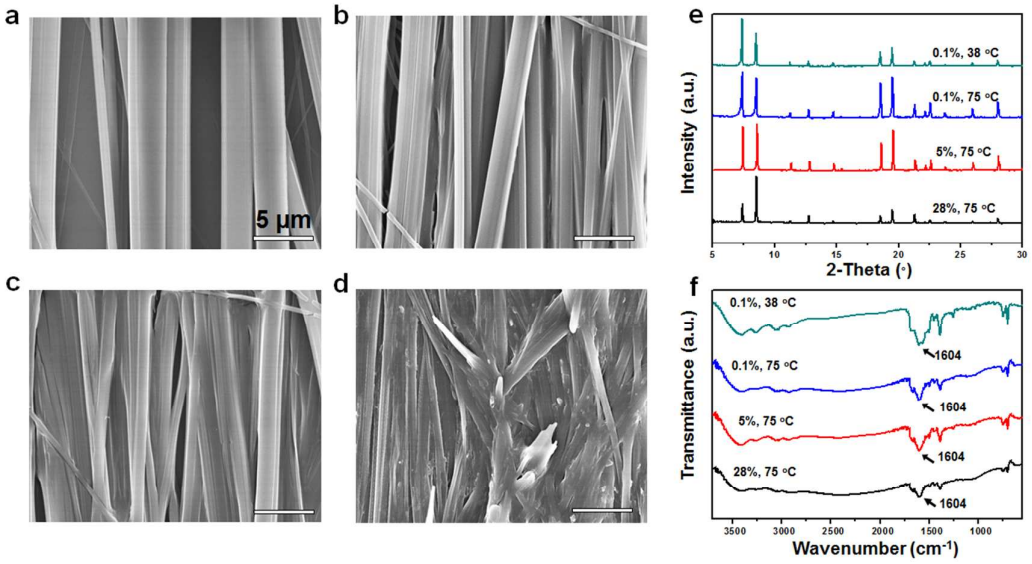

Figure S3. Aligned FF fibers formed under different NH_4OH concentration and temperature. a) SEM image of fibers in 0.006 mol/L NH_4OH at 38 ^oC. b) SEM image of fibers in 0.006 mol/L NH_4OH at 75 ^oC. c) SEM image of fibers in 2.89 mol/L NH_4OH at 75 ^oC. d) SEM image of fibers in 14.80 mol/L NH_4OH at 75 ^oC. Scale bar, 5 µm. e) FTIR and f) XRD analyses of the FF fibers. C_0=0.026 g/L, withdrawal speed, 2 µm/s.

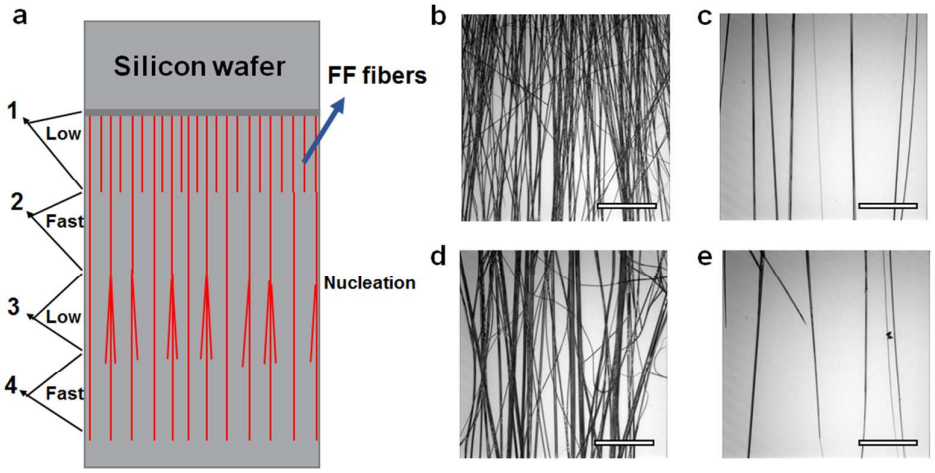

Figure S4. The "gradient single crystals" at a silicon wafer (positions 1,2,3,4) obtained by four steps with different withdrawal speeds. a) The schematic of the formation. The withdrawal speed at step 1,2,3,4 is b) 2.0 μm/s, c) 4.6 μm/s, d) 2.0 μm/s, e) 4.6 μm/s, respectively. The scale bar is 100 μm.

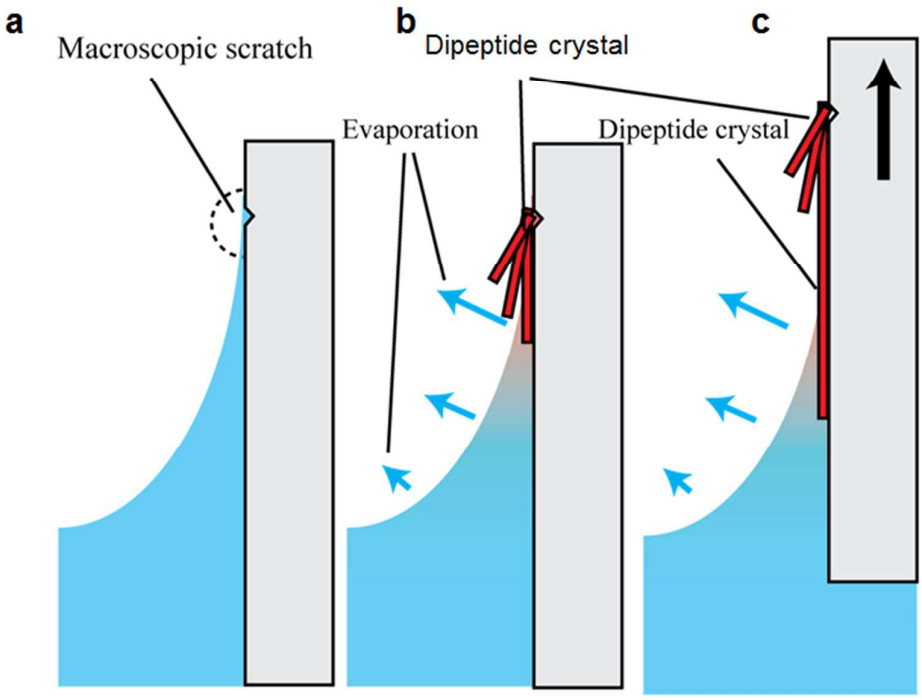

Figure S5. Schematic of proposed assembly mechanism of the aligned FF single crystal growth process. a) Macroscopic scratch provides a nucleation side. b) FF fibers (coffee-stain) act as "crystal seed". c**)** Dip-coating starts to withdraw FF fibers.

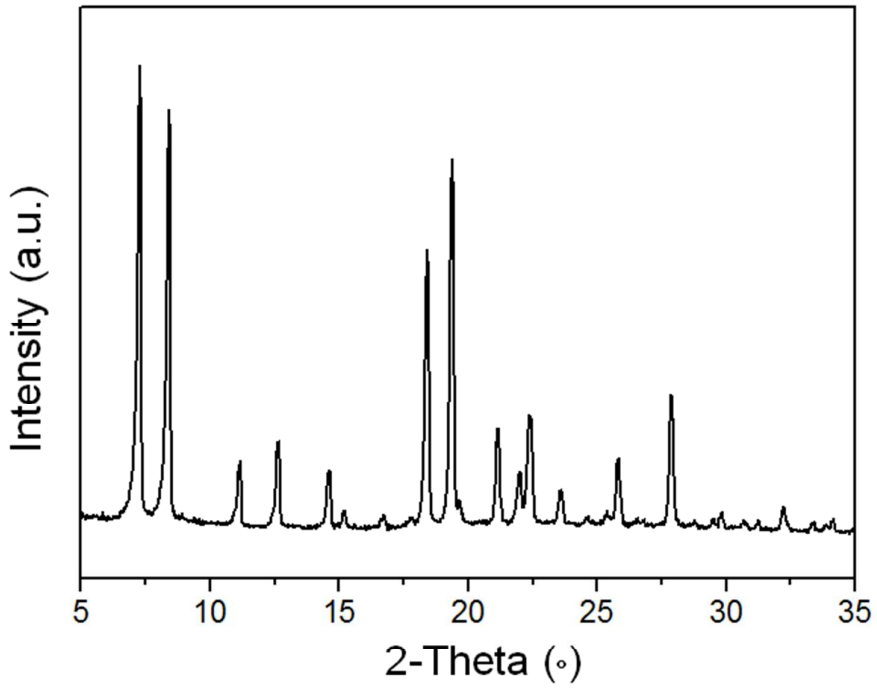

Figure S6. XRD pattern of the FF-RhB fibers. 0.006 mol/L NH_4OH at 38 ^{o}C, withdrawal speed, 2 μm/s.

Movie S1. Growth of aligned FF single crystals on silicon wafer with the withdrawal of substrate.