[Supporting Information]

Amyloid Fibrils are "Alive": Spontaneous Refolding from One Polymorph to Another

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Experimental:

Preparation of apo-α-lactalbumin fibrils

Bovine apo-α-lactalbumin (Sigma, St.Louis, MO) solution (5 mg/ml, 150mM NaCl, pH 2.0 adjusted by adding HCl) was incubated at 37 °C with constant stirring for 72 h. The incubation was terminated by separating the gelatinous fraction by centrifugation at 14000**g** for 30 min at 37 °C. The separated fibrils were redispersed in pH 2.0 HCl solutions with and without added NaCl.

Atomic force microscopy (AFM)

Redispersed fibrils were diluted with aqueous pH 2.0 HCl solution in a 1:400 ratio and deposited onto a freshly-cleaved mica. AFM imaging was performed by using a MFP-3D™ Bio Asylum Research microscope (Asylum Research, CA, USA) in non-contact mode with Olympus AC160TS tips.

Deep UV resonance Raman spectroscopy

197-nm excited DUVRR spectra were measured using a home-built Raman spectrometer as described elsewhere.[29] A spinning NMR tube with a magnetic stirrer inside was used for sampling. All reported Raman spectra are an average of at least three independent measurements. GRAMS/AI 7.0 software (Thermo Galactic, Salem, NH) was used for spectral data processing.

H/D exchange

The method of hydrogen-deuterium exchange combined with DUVRR spectroscopy is described in detail elsewhere[15]. Briefly, samples (1mL) of fibrils dispersed in water were centrifuged at 14 000**g** for 30 min. The precipitate was divided into two parts, each of which were washed two times, one with D_2O and the other with H_2O, by subsequent spinning-redispersion. The washed dispersions (in D_2O and H_2O) were used for Raman spectroscopic measurements.

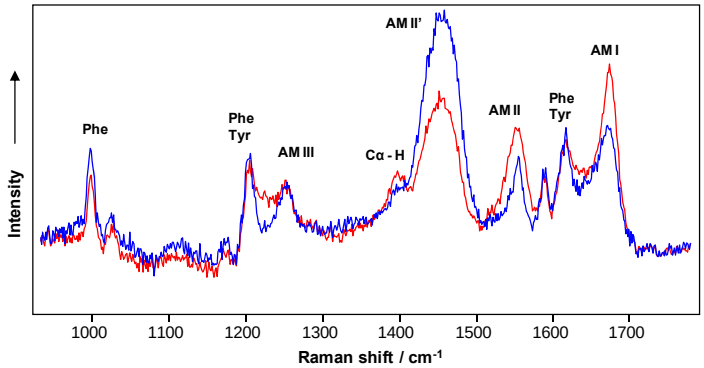

Figure S1. DUVRR spectra of fibril Polymorphs I (red) and II (blue) in D_2O. H-D exchange of Polymorph II was performed after fibril incubation in H_2O for 7 hours at 25 °C.