Fichman, G., Guterman, T., Damron, J., Adler-Abramovich, L., Schmidt, J., Kesselman, E., Shimon, L. J.W., Ramamoorthy, A., Talmon, Y., Gazit, E. Spontaneous structural transition and crystal formation in minimal supramolecular polymer model. ***Sci. Adv.***; 2016: 2, e1500827.

**Abstract:** The association of building blocks into supramolecular polymers allows the fabrication of diverse functional architectures at the nanoscale. The use of minimal assembly units to explore polymer dynamics and phase transitions significantly contributes to the application of polymer physicochemical paradigms in the field of supramolecular polymers. We present a minimal model that displays spontaneous coordinated structural transitions between micro- and nanostructures, hydrogels with nanoscale order, and single crystals. The simple amphiphilic 9-fluorenylmethoxycarbonyl-3,4-dihydroxyphenylalanine (Fmoc-DOPA) modified amino acid undergoes a noninduced transition from spherical assemblies into nanofibrils followed by sol-gel transition, nanotube formation via intermediate assembly, and crystallization within the gel. Notably, the transition kinetics is slow enough to allow both multistage and multiscale characterization of the supramolecular arrangement using electron microscopy, vibrational and circular dichroism spectroscopies, nuclear magnetic resonance, and x-ray crystallography. This minimalistic system is the first comprehensive model for a complete spontaneous structural transition between diverse states governed by distinct molecular interactions.