Title:	Liquid–Liquid Phase Separation Is Driven by Large-Scale Conformational Unwinding and Fluctuations of Intrinsically Disordered Protein Molecules
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Keywords:	Noncovalent Intermolecular Interactions
Issue Date:	2019
Publisher:	American Chemical Society
Citation:	Journal of Physical Chemistry Letters, 10(14), pp.3929-3936.
Abstract:	Liquid–liquid phase separation occurs via a multitude of transient, noncovalent, and intermolecular interactions resulting in phase transition of intrinsically disordered proteins/regions (IDPs/IDRs) and other biopolymers into mesoscopic, dynamic, nonstoichiometric, and supramolecular condensates. Here we present a unique case to demonstrate that unusual conformational expansion events coupled with solvation and fluctuations drive phase separation of tau, an IDP associated with Alzheimer's disease. Using intramolecular excimer emission as a powerful proximity readout, we show the unraveling of polypeptide chains within the protein-rich interior environment that can promote critical interchain contacts. Using highly sensitive picosecond time-resolved fluorescence depolarization measurements, we directly capture rapid large-amplitude torsional fluctuations in the extended chains that can control the relay of making-and-breaking of noncovalent intermolecular contacts maintaining the internal fluidity. The interplay of these key molecular parameters can be of prime importance in modulating the mesoscale material property of liquid-like condensates and their maturation into pathological gel-like and solid-like aggregates.
URI:	https://pubs.acs.org/doi/abs/10.1021/acs.jpclett.9b01731 (https://pubs.acs.org/doi/abs/10.1021/acs.jpclett.9b01731) http://hdl.handle.net/123456789/1991 (http://hdl.handle.net/123456789/1991)
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