NANOSCIENCE COLLOQUIUM

Following and controlling nanoscale formation and function of bottom-up assembled materials

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ABSTRACT:

Short-range-interacting particles can in principle crystallize via so-called non-classical pathways invoking a metastable liquid intermediate, yet non-equilibrium gelation often occurs before a metastable liquid can form. Using in situ small angle X-ray scattering, we nevertheless watch electrostatically stabilized colloidal semiconducting nanocrystals self-assemble into long-range-ordered superlattices via this non-classical pathway and show how the pathway increases the rate of crystallization over that of direct crystallization from the colloidal phase. Furthermore, by mapping the phase behavior and kinetics as a function of nanocrystal density and electrostatically tuned driving force for assembly, we demonstrate a highly unusual degree of control of a nanoscale system. This control is exemplified by varying the self-assembly rate by over three orders of magnitude, along with predictive control of superlattice yield, size, and crystallinity. Most strikingly, we reveal that this non-classical pathway increases crystallinity of the superlattice simultaneously with the crystallization rate. To further elucidate the elusive nature of the short-range interactions at the nanoscale, we also study the microscopic fluctuations of colloidal suspensions and liquid droplets of the nanocrystals via MHz X-ray photon correlation spectroscopy (XPCS). We discover suppressed nanocrystal self-diffusion in the liquid state, which we attribute to the explicit attractive interactions that are not captured by typical charged particle hydrodynamic models. The combined results suggest design rules for the shape of interaction potentials not only to leverage liquid intermediates in crystallization processes but also to avoid gelation for better control of phase behaviors.

To subtly modify the nanocrystal interactions we take advantage of their light absorption. Current results from ultrafast optical transient absorption

spectroscopy suggest that exciton dissociation into charges trapped at the nanocrystal surface alters the local configuration of molecular ions modify longer time scale self-assembly kinetics and resulting superlattice lattice constants. Time-resolved wide angle X-ray scattering and associated pair distribution function analysis preliminarily show the corresponding molecular reorganization in the nanocrystal solvation shell. Ultimately, the multiscale characterization of and manipulation of electrostatically stabilized nanocrystals paves the way to more clearly explain the design rules for nanoscale interaction potentials so that nanomaterial assemblies can achieve more effective functionalities via deterministic and predictive control.



For a bio, please page to the bottom of the following URL (photo is at the top): http://www.cchem.berkeley.edu/nsggrp/ginsberg.html



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