

Abstract Submitted
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Dynamic, Directed Self-Assembly of Nanoparticles via Toggled Interactions ZACHARY SHERMAN, JAMES SWAN, Massachusetts Institute of Technology — Crystals self-assembled from nanoparticles have useful properties such as optical activity. During fabrication, gelation and glassification often leave these materials arrested in defective metastable states, a key difficulty preventing adoption of self-assembled nanomaterials at scale. Dynamic, directed self-assembly processes in which interactions are actuated temporally, offer a promising method to suppress kinetic arrest while accelerating growth of nanostructures. We show with simulation and theory how time-dependent, periodically toggled interparticle interactions can avoid kinetic barriers and yield large crystalline domains for a dispersion of nanoparticles. The growth mechanism and terminal structure are controlled by parameters of the toggling protocol, allowing for selection of processes that yield rapidly assembled, low defect crystals. Though toggled self-assembly forms dissipative materials inherently out-of-equilibrium, its outcome is predicted by a nonequilibrium thermodynamic theory, requiring equality of time-averaged pressure and chemical potential in coexisting phases of the dispersion. The predicted phase behavior agrees with that from simulations. We also present kinetic models to predict the rate of crystallization for several observed growth mechanisms.

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