Field-Directed Self-Assembly of Colloidal Particles for Engineering Phononic Materials

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The self-assembly of nanoparticles serves as a way to build periodic crystalline structures. Externally-applied stimuli enable one to manipulate interparticle interaction potentials, and thus, avoid kinetically-arrested states to reach equilibrium phases. By using suitable pairs of particle chemistry and external stimulus, colloidal suspensions can be programmed to fabricate a broad range of functional materials.

We study the effect of time varying fields on the self-assembly of colloidal suspensions. The microstructural growth of paramagnetic colloids is observed in toggled magnetic fields. The toggled magnetic field repeats, switching on and off periodically with a square waveform. The suspension begins in a solid, gel-like state and anneals to droplet-like domains governed by time-averaged bulk and surface energies. The terminal structure of this process depends on the frequency and duty cycle of the toggled field. Four structures are observed: fluid, columnar, percolated, ellipsoidal states. In addition to these four states, we identify a new perpendicular state. In this state, the suspension forms zigzag pattern of millimeter scale structures that grow perpendicular to the applied field direction. The kinetics of the structural evolution are classified into two distinctive pathways. The ellipsoidal and perpendicular states involve the coalescence and breakup of aggregates that leads the suspension to an energetically favorable state. These phenomena are demonstrated for both spherical particles and shape-anisotropic dumbbell particles.

The periodic nanostructures of colloidal crystals lead to unique mechanical and acoustic properties. The phononic structure of colloidal crystals is an important property accessible by colloidal self-assembly. We fabricate anisotropic nanostructures with nonspherical dumbbell particles. Brillouin light scattering (BLS) is used to measure the phonon dispersion relation and verifies that a tunable bandgap arises that depends on the propagating direction of acoustic waves relative to the lattice structure of the colloidal crystal. Normalizing the phonon dispersion relation scales the bandgap frequency, providing a new structure-property relation. This relation may play a key role in engineering phononic materials with a desired phononic property.

Finally, the thermomechanical properties of polymer nanoparticles are characterized using temperature-dependent BLS measurements. The temperature-dependent vibrational modes of nanoparticles reveal the existence of a polymer mobile layer at the surface. We further extend this observation to a core/shell particle structure. A shell atop of the seed particle leads to a strong suppression of a polymer mobile layer on the nanoparticle surface. The modification of polymer dynamics at interface can improve the thermal stability of polymer nanomaterials.

These results suggest that colloidal self-assembly is a promising way to develop functional nanomaterials. One can design specific structures to achieve desired phononic bandgaps from versatile building blocks.