Dissipative Self-assembly in Paramagnetic Colloidal Suspensions

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Dissipative colloidal self-assembly offers a way to create materials whose structure is determined not by dynamic arrest (e.g. colloidal gels) or thermal equilibrium (e.g. colloidal crystals), but by the persistent input and viscous dissipation of energy. It is a design strategy to produce non-equilibrium colloidal assemblies. Paramagnetic colloidal particles are excellent starting materials for these processes because the strength of their interactions can be easily and precisely varied in time. Yet a quantitative framework linking the applied field protocols to morphology and dynamics to the magnetostatic energies of self-assembled particle aggregates is still lacking. This dissertation addresses that gap by analyzing microgravity experiments on the International Space Station using quantitative image and video analysis which supplements theoretical and computational investigations that relate unsteady field protocols to the steady-state suspension structure; ultimately we seek to identify when observed steady states resemble equilibrium versus nonequilibrium structures.

First, we develop a metrology to quantify the shape, spatial organization, and dynamics of complex colloidal aggregates from microscopy data. These structural and dynamical metrics are combined into compact morphological descriptors that define a coherent phase diagram. Within this framework, we identify dissipative steady states, including a ribbon phase in which aggregates continually split, merge, and exchange small clusters, maintaining sustained internal motion despite a nearly stationary average structure. These surface instabilities correspond to drastic increases in viscous dissipation.

Next, we construct a hierarchy of magnetostatic models that connect aggregate shape and crystal packing to suspension energy and effective susceptibility, ranging from continuum descriptions to descriptions incorporating local structure and interfacial energies. These models predict how local fields and surface energies depend on packing and orientation, explain the emergence of needle-like and sheet-like aggregates, predict large enhancements in effective susceptibility upon aggregation, and clarify where aggregates are forming low-energy structures.

Together, these results affirm paramagnetic colloidal suspensions in toggled magnetic fields as a model platform for dissipative self-assembly and yield important inroads into the creation of quantitative design principles for producing *non-equilibrium*, field-responsive colloidal materials.