TOPICAL: FIELD-DRIVEN EVOLUTION OF 4D COLLOIDAL MATTER IN MICROGRAVITY

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by

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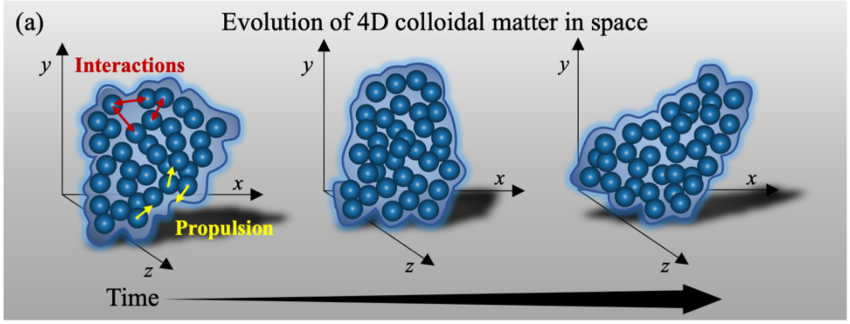
1. Overview

Understanding, controlling, and directing the assembly of colloids and nanoparticles requires a delicate interplay of non-equilibrium, long-range, competing interactions[1]. An ability to "dial-in" the range, strength, and cooperativity of these interactions in a pre-programmable manner is the key to tailor the soft microstructures and thus develop active and reconfigurable materials. The presence of a gravitational field can hinder, even prevent such self-assembly as a competing force acting to disrupt the intended structure[2]. In this white paper, we focus on the critical need and merit of investigating nonequilibrium, long-range competing interactions using colloidal particles in microgravity and studying their external field-driven behaviors in four dimensions (4D), i.e., structural evolution in space and time (Figure 1a). 4D matter is ubiquitous in living systems such as the unicellular organism Amoeba, which dynamically reconfigures and alters its shape and spatial configuration (Figure 1b). Establishing the principles of encoding such non-equilibrium response in synthetic materials using external electric and magnetic fields is a grand scientific challenge and the focus of this white paper. A fundamental obstacle in addressing the challenge is our inability to decouple surface and field-induced interactions from gravitational effects on earth.



(b) Amoeba; Source: YouTube (Dr. Ralf Wagner)

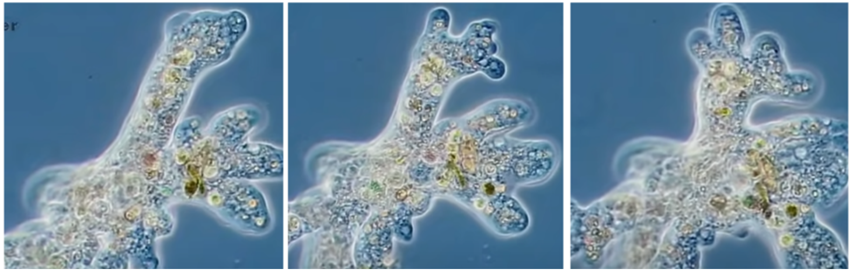


Figure 1. *Schematics representing the change in structure of an ensemble of colloidal particles over time in external field. The evolution of structure is driven by a combination of interparticle interactions and active forces operational on individual particles in external electric and magnetic fields. (b) The change in structural characteristics of a unicellular organism, here Amoeba. The road to understand the origin of the structural evolution in the living matter goes through investigating the collective behavior and emergent properties of colloidal matter.*

These challenges become more significant when weak competing interactions, such as the ones induced by external fields, or emerging during self-propulsion are at play and driving the structure and dynamics of an ensemble of colloidal building blocks. These limitations can be overcome by performing systematic studies in microgravity which would allow decoupling the surface and field-induced interactions from gravitation forces, thus enabling new insights into the non-equilibrium colloidal matter. The fundamental understanding of the non-equilibrium phenomena from nano- to millimeter-scale systems is critical for designing the next generation of materials capable of structural evolution - moving, morphing, and transforming energy from one form to another.

2. Introduction

Competing and cooperative interactions are critical to the existence of matter, from the attraction-repulsion balance of electrons and protons leading to the formation of atoms to the joint operation of proteins and bacteria in colonies. However, there is a lack of understanding of the factors governing the weak balance of competing and cooperating forces leading to unusual structural and dynamical features of the self-assembled matter. One fundamental class of poorly understood interactions is long-range interactions induced by external electromagnetic fields. Here we demonstrate a critical need to investigate such interactions using colloidal particles. Because of our ability for *in situ* visualization, colloids are ideal models for studying the structure, dynamics, and kinetics of assembly, especially under non-equilibrium conditions controlled and guided by external fields.

Microgravity provides a unique environment to study colloidal assembly and dynamics in ways that cannot be accomplished on earth[3]. The lack of buoyancy-driven convection and sedimentation in microgravity have furthered our understanding of the structure and dynamics of colloidal suspensions. The first series of experiments performed at the International Space Station (ISS) demonstrated how micron-sized hard-sphere systems undergo disorder-order phase transitions, characterized by optical microscopy and light scattering measurements[4]. Colloidal suspension that could not crystallize on earth were able to form millimeter-sized crystallites in microgravity. Experiments with colloids and polymers demonstrated the role of polydispersity on colloidal phase separation and gelation[5]. Magneto-rheological colloidal fluids can be driven to form kinetically trapped and percolated structures by toggling the field strength at varied frequencies[6]. The benefits of the microgravity environment include understanding of the fundamental thermodynamic and kinetic processes of phase separation of colloidal systems without being hindered by sedimentation. These initial experiments in space were seminal in providing new insights into colloidal assembly based on intrinsic interactions. The next phase of colloidal science will be built on this fundamental research to understand the external-field-driven assembly of colloids and engineer new colloidal materials to improve space exploration.

Directed assembly with magnetic, electric, and optical fields offers precise control and structural organization of colloidal materials[7-9]. These assemblies are typically non-equilibrium, whereby a constant input of energy provides continuous reorganization of colloidal structures which can grow and respond to their environment, analogically to living systems[10-11]. Structures formed under nonequilibrium conditions can be much more diverse than those governed by classical thermodynamics[12-13]. Therefore, fundamental research is needed to isolate and study non-gravitational phenomena, prepare defect-free materials, and enable container-free processing of materials. The advent of a new commercial space age has made it possible to manufacture colloidal materials in space that can be brought back to earth. This opens unprecedented challenges and immense opportunities for fabrication using evolving soft matter. For example, in the future the legendary, but elusive Star Trek “replicator” may be realized using nonequilibrium processes such as electromagnetic fields to 3D print colloidal fluids into free-standing objects.

Furthering this field, active colloids are an emerging class of synthetic matter where an external input of energy induces non-Brownian dynamics in colloids resembling that of living matter. The unusual dynamics and non-ergodic nature of the active particles derive collective and emergent behaviors which are far from understood[14-15]. There is a critical need to understand the push-pull balance of equilibrium and non-equilibrium forces on both structure and dynamics of colloids. Understanding the cooperative and collective flocking of particles will enable better control the structures and dynamics at multiple length scales ranging from angstrom to millimeters. Using active particles as non-living counterparts to model interactions in "living" matter could develop underlying principles in making materials with life-like features, including self-replication, self-propulsion, morphogenesis, and an ability to self-heal. In summary, the key to the design of out-of-equilibrium colloidal materials is understanding and directing the equilibrium and non-equilibrium forces at multiple length scales with a high degree of control over the orientational and positional order of the colloids.

3. Future of research on field-driven colloids: Scientific challenges

Microgravity offers vast opportunities to solve the following key scientific challenges:

1. Understand the behavior of colloidal systems near and far from equilibrium, especially the time evolution of configurations during the assembly process, phase transitions, non-equilibrium steady states, transient dynamics, and arrested dynamics. Based on such understanding, we should seek to gain in-depth knowledge on the correlation between the dynamical evolution of the system configurations and the properties of the resultant materials (e.g., those from superlattice structures), as guided by an external field[16-19].
2. Investigate the rich and subtle behavior of colloidal systems (e.g., meta-stable states) that occur when the interactions between colloids, interfaces, external fields, suspending fluids, etc. are of comparable magnitude and cooperativity (including time-varying inputs self-propulsion, unsteady fields, etc.)[20-24]. The microgravity environment will allow us to explore the electromagnetic-field-mediated interactions over orders of magnitude, from weaker than the gravitational potential energy to much stronger.
3. Identify the inherent coupling between molecular intricacies and forces/torques of colloids, leading to emergent properties at the macroscopic scale[25-27]. This involves careful consideration of friction landscapes (e.g., configuration-dependent hydrodynamics and contact friction) as well as free energy landscapes (e.g., potential energy and entropy landscapes) similar to atoms[28-30].

3.1. Experiments on earth and in microgravity

Earth's gravity limits the choice of fluids, colloid material and size, and the means to observe colloidal dynamics. Furthermore, density matching often is incompatible with the optical transparency needed for observation and may modify surface chemistry and thus interparticle interactions. Finally, density matching is also nearly impossible in mixtures of colloids of different types or colloids with dissimilar composition such as particles with surface patches. Therefore, microgravity is necessary and would greatly expand the parameter space, enabling exploration in four major directions:

1. *Three-dimensional (3D) suspensions*: Active colloids have been shown to undergo activity-induced phase separation to form dense phases (liquid or crystalline) as a result of a kinetic trapping mechanism whereby the swim speed decreases with increasing particle density[31]. Experimental investigation of activity-induced phase separation in 3D is hindered by gravity leading to the following observations: (1) the dense particles sediment onto a planar substrate, (2) the anisotropic particles experience gravitational torques that dominate over those due to Brownian motion and interparticle interactions, (3) the consumption of chemical fuel (e.g., hydrogen peroxide) and/or the absorption of light leads to concentration and thermal gradients that drive buoyant flows. Novel structures and behavior, e.g., superfluidity, are expected in 3D. Activity can be used to deform droplets and create on-demand 3D shapes. On earth, gravitaxis causes particles to accumulate at interfaces. As a result, monolayers of active colloids have

been primarily studied. Even if 3D structures are formed on earth, infinitesimal density mismatch can lead to artifacts and structure degradation over time.

2. *Assembly and dynamics of active colloids on curved interfaces*: The interplay of curvature and field-driven colloid activity could create dynamic, reconfigurable assemblies of more complex patterns[32-33]. While in vicinity of flat surfaces, colloids typically form hexagonal lattices. The absence of gravity allows surface tension to shape liquids in large droplets and various curved interfaces.
3. *Isotropic and anisotropic interactions*: Knowledge of the interaction potentials would inform simulations to predict colloidal dynamics on earth, where particle interactions are affected by suspending fluids and it is challenging to disentangle intrinsic interactions and gravity effects. Microgravity will help isolate the interparticle interactions and untangle the influence of a suspending fluid (e.g., particles need not be suspended in fluids in microgravity), introducing various combinations of isotropic/anisotropic interactions that allows unprecedented analogous systems. For convenient observation, 2D systems without gravity, e.g., colloids confined by acoustic field, can be used to extrapolate behavior to 3D systems.
4. *Anisotropic medium*: Interactions of colloids and thus their assembly in isotropic media such as water is controlled primarily by short-range forces, such as the screened Coulomb interactions. Isotropic environment, especially the microgravity environment offers limited means to control directional assembly, dynamics, and modes of collective motion of active and passive colloids. An additional sense of direction could be imparted by an orientationally ordered medium, namely, a liquid crystal[34-36]. Orientational elasticity and anisotropic surface properties of liquid crystals produce long-range forces among microparticles, both inanimate and living, thus allowing one to commands self-assembly and collective behavior at the microscale.

3.2 Role of theory and computation

The application of theory/simulations/artificial intelligence to predict colloidal assembly in the microgravity environment will play a central role in addressing the key science challenges as follows:

1. It will help disentangle the individual contributions made by the various forces/torques in the overall assembly process, greatly facilitating the design, control, and optimization of new experiments in microgravity. Specifically, in complex systems of interest, theory/simulations can be used to understand how the absence of gravity (that impacts the experiments on earth) will lead to different structures formed in space. By bridging the gap between experiments on earth and in microgravity, theory/simulations will be used to predict the possible outcomes of experiments in the microgravity arena[37-40]. It can provide hypotheses to be tested and will form an integral part of a loop in which experiments inform models, which then inform the design of new experiments to revise models.
2. It will provide reliable methods to interpret experiments by matching observables between experiments and simulations (i.e., particle coordinates, radial distribution functions, etc.). More importantly, it will provide an opportunity to advance science by implementing state-of-the-art modeling techniques suitable to non-equilibrium systems. In comparison to experiments (e.g., microscopy and scattering) where spatial and temporal resolutions are often limited along with practical difficulty in numbers of trials, simulations are more amenable to generate necessary statistics without experimental artifacts (noise, resolution, etc.). Such noise-free statistically significant data sets are essential to machine learning methods such as inverse

analyses, dimensionality reduction (principal component analysis, diffusion mapping). Moreover, reinforcement learning can be readily and reliably applied.

3. Modeling and computation, combined with experiments, will provide a direct pathway to correlate molecular details to forces/torques at colloidal length scale. For example, colloids in aqueous solutions organize into unique structures at the solid-liquid interface that are dependent on the surface chemistry of both the substrate and particles compared to the bulk structures[41-43].

4. Soft matter and colloid science at NASA: A vision

NASA has an unmatched track record of impactful colloidal science experiments conducted under microgravity, evolving into the recent Advanced Colloids Experiments (ACE) series of investigations. In this work, it has been demonstrated that insights into the fundamental phenomena governing colloidal self-assembly that are vital to the design of advanced materials, can only be obtained in microgravity conditions. Ultimately, the ability to control colloidal assembly and behavior within three-dimensional environments opens a broad spectrum of new structures with novel functionalities. Here, the long-term goal is to expand NASA-sponsored research with new capabilities at the international forefront of dissipative soft matter and long-range self-organizing materials and structures. With a focus on directing structures and dynamics through the application of external fields, this research will both establish the principles of guided four-dimensional soft matter design and also enable the development of new microscale engineering and biomedical devices. Our research paradigm will enable future space-based manufacturing of intelligent, self-reconfiguring, self-repairing, and self-replicating soft materials for building and expanding extraterrestrial bases critical to improving life on earth and space exploration. The development of advanced materials, sensors, and products will impact a variety of NASA Space Technology Roadmap objectives, including those in Nanotechnology (TA10), Materials, Structures, Mechanical Systems, and Manufacturing (TA12), Space Power and Energy Storage (TA3), and Thermal Management Systems (TA14).

The collective organization of weakly interactive particles has, to date, been a topic of intensive research and the focus of a large and well-recognized community of physics, condensed soft matter, and colloid science researchers. However, long-range colloid ‘signaling’ and organization in external electric and magnetic fields poses a significant research challenge on earth due to the interference of gravitational forces in the evolution of structure and dynamics of colloidal matter. The unique microgravity environment overcomes these fundamental barriers and allows precise control of particle dynamics and organization, leading to long-range structuring. In addition to providing precise control of particle interactions conducive to direct assembly of active particles, soft matter swarms, and new types of ultrasoft and ultraflexible microstructures, external fields provide the energy required to power responsive micromachines able to perform complex tasks. The emerging topic of collective dynamics of such micromachines would be a natural and logical evolution of active colloids in the years ahead. Thus, the proposed research will be of high fundamental importance and intellectual impact and will be at the forefront of new active materials and structures as the future basis of developing micromachines.

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