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Complex collective dynamics of active torque-driven colloids at interfaces

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ABSTRACT

Modern self-assembly techniques aiming to produce complex structural order or functional diversity often rely on non-equilibrium conditions in the system. Light, electric, or magnetic fields are predominantly used to modify interaction profiles of colloidal particles during self-assembly or induce complex out-of-equilibrium dynamic ordering. The energy injection rate, properties of the environment are important control parameters that influence the outcome of active (dynamic) self-assembly. The current review is focused on a case of collective dynamics and self-assembly of particles with externally driven torques coupled to a liquid or solid interface. The complexity of interactions in such systems is further enriched by strong hydrodynamic coupling between particles. Unconventionally ordered dynamic self-assembled patterns, spontaneous symmetry breaking phenomena, self-propulsion, and collective transport have been reported in torque-driven colloids. Some of the features of the complex collective behavior and dynamic pattern formation in those active systems have been successfully captured in simulations.

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1. Introduction

Self-assembled colloidal structures and materials capable of supporting structural complexity and functional diversity must consume energy from the environment and as a result remain out of equilibrium [1,2]. Such dissipative self-assembly is often called dynamic or active self-assembly [1] in contrast to more conventional static assembly happening at or near thermodynamic equilibrium [3–7].

A significant work has been dedicated to various aspects of equilibrium colloidal structures obtained as a result of static self-assembly [8–23]. Static magnetic and electric fields have been successfully used to direct and control the self-assembly processes through in situ modification of particle interactions [24–35]. It allowed to slightly expend the amount of available self-assembled structures. Once formed, these assemblies do not require an external energy to sustain the structure. In contrast, dynamically assembled structures rely on external energy input and cease to exist once the energy source is removed. Due to the fact that these externally driven particles are not in thermodynamic equilibrium, these dynamically assembling systems are called "active." Alternating electric/magnetic fields demonstrated the potential to introduce dynamics into the self-assembly process, and a number of nontrivially ordered dynamic structures have been reported [36–50]. Remarkable toroidal vortices and pulsating rings have been reported in electric field-driven colloidal ensemble comprised of conductive spherical particles in low-electrolyte liquids [50]. Dynamic two-dimensional hexagonal sheets assembled in rotating magnetic [38,51] or electric [47] fields were observed in suspensions of paramagnetic and PMMA particles, respectively. Particle foams, honeycomb-structured composites, and complex dynamic vortex patterns have been reported in triaxial time-varying magnetic fields [38,52]. Dynamically assembled colloidal structures "living" outside of equilibrium have made accessible properties that are usually attributed to biological systems, such as self-healing [42]. In all scenarios of the dynamic self-assembly in colloidal suspensions, the observed dynamic patterns are often a result of a fine-tuned dynamic balance between magnetic/electric dipole interactions, steric repulsions, and induced hydrodynamic flows.

The present paper reviews a less conventional case of collective dynamics and self-assembly of particles with externally driven torques coupled to a liquid or solid interface. Hydrodynamic interactions are believed to play a crucial role in the onset of collective dynamics and pattern formation in torque-driven suspensions.

2. Magnetic field-assisted torque-driven particles: collective dynamics and self-assembly

Magnetic field is an effective tool to exert torques on any material with magnetic moment (permanent or induced). One of the first realizations of torque-driven dynamic self-assembly powered by a magnetic field was accomplished in a system of magnetized millimeter-sized discs suspended at a liquid–air interface and subjected to a rotating field produced by a rotating permanent magnet [53,54]. The disks were spinning around their axis with the frequency of the permanent

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magnet, see Fig. 1. The fluid motion associated with the disk spinning induced repulsive hydrodynamic interactions between spinning disks. The competition between axisymmetric magnetic attraction and hydrodynamic repulsion of the disks let to the formation of a number of dynamic patterns exhibiting various types of ordering [53,54]. Experiments clearly demonstrated that inertial effects are significant in such system (the Reynolds number, $\mathrm{Re} = \frac{\rho\omega a^2}{\mu} < O(1)$, is small but finite). Each particle experiences a lift force transverse to the streamlines. Hydrodynamic repulsion facilitated by the lift forces balances the time-averaged magnetic attraction and leads to a formation of steady dynamic patterns. Each particle dissipates energy supplied by the external rotating magnetic field while maintaining dynamic order. The frequency of the rotating magnetic field can be tuned to adjust the force balance in the system, resulting in dynamically assembled patterns [53,54].

Magnetic particles with pinned (or fixed) magnetic moments automatically have a potential to be torque-actuated in alternating magnetic fields in contrast to paramagnetic particles that acquire magnetic moment along the field direction only when subject to the external magnetic field. Ferromagnetic suspensions proved to be scientifically rich systems to study collective dynamics and self-assembly in active torque-driven ensembles [43,2].

Ferromagnetically ordered particles subjected to a uniform constant magnetic field experience a torque, forcing their magnetic moment to be aligned with the applied field direction. There are two scenarios on how a ferromagnetic particle can change magnetic moment orientation: (a) it can rotate magnetic moment inside the particle by an adjustment of the internal magnetic domain structure, or (b) it can mechanically adjust the orientation of the particle to align the magnetic moment with the external field direction. In a typical ferromagnetic microparticle, magnetic domain walls are pinned by the internal defects, and the reorientation of its movement is often associated with high energy losses. As a consequence, in most cases, it is more energetically favorable for a ferromagnetic microparticle at a liquid interface (where the friction is low) to proceed with the mechanism (b) and mechanically adjust the orientation of the particle("magnetic shaking") [45]. During this process, we transfer the particle's torque to the local excitations of the liquid interface and induce vortical hydrodynamic flows around each particle.

Maintained in a state away-from-equilibrium by applying alternating (*ac*) magnetic fields, a magnetic colloidal suspension at liquid interfaces exhibits a strong tendency toward dynamic self-organization [43,37]. Two distinctive geometries of the torque-driven actuation of particles are possible for ferromagnetic suspension at liquid interfaces: (a) alternating magnetic field is perpendicular to the interface [55], and (b) alternating field is along the interface [56].

2.1. Dynamic assembly of particles driven by a magnetic field transversal to the liquid interface

In a transverse orientation (the alternating field axis is perpendicular to the liquid interface), astounding dynamically assembled structures (see Fig. 2) emerge in a certain range of excitation parameters (magnetic field amplitude and frequency) [45].

These structures are dynamic by nature and exist only while we supply energy by means of an external driving field. Once formed, the patterns are stable, provided the driving field is unchanged. Each structure is composed out of segments; each segment consists of ferromagnetically aligned chains of microparticles whose magnetic moments are aligned along the chain direction. The segments, however, are anti-ferromagnetically aligned: the total magnetic moment per segment reverses its direction from section to section as demonstrated in the Fig. 2(c). Amazingly, the long-range order has a completely unconventional origin. It is facilitated by a structure-induced surface wave [55,45,57].

In the process of dynamic self-assembly, ferromagnetic suspensions at liquid–air interfaces often develop strong large-scale hydrodynamic surface flows in the vicinity of assembled structures [58]. The strongest flows are concentrated at opposite ends of the dynamic pattern where the centers of the vortices are located (dark spots in Fig. 3(a)). The flow velocity can be as fast as a few centimeters per second and is controlled by the frequency of the driving magnetic field. It was demonstrated that under certain conditions [39] the reported dynamic structures spontaneously break the symmetry of self-generated surface flows and turn into self-propelled entities, Fig. 3(b). This type of magnetic surface swimmer is rather unique due to the unusual mechanism of self-propulsion exploiting symmetry breaking of self-generated surface flows and the intrinsic antiferromagnetic nature of the swimmer's structure, in contrast to previously reported colloidal swimmers [59,60]. In a multiple-swimmer state, dynamic self-assembled swimmers create a highly disordered and non-periodic surface velocity field [61] with Kolmogorov energy spectra that are characteristic of two-dimensional systems; this result indicates that self-generated flows are highly localized near the interface.

One of the key ingredients facilitating active self-assembly of ferromagnetic suspensions at liquid–air interfaces and leading to the formation of magnetic snakes has been hydrodynamic long-range interactions produced by particles. Consequently, the modification of this component of particle interactions provides an efficient tool to modify and control the results of dynamic assembly. Changes in the interface liquid–air viscosity [62] or introduction of a top liquid layer (liquid–liquid interface) [37] produces another remarkable self-assembled dynamic structures – localized asters – illustrated in Fig. 4.

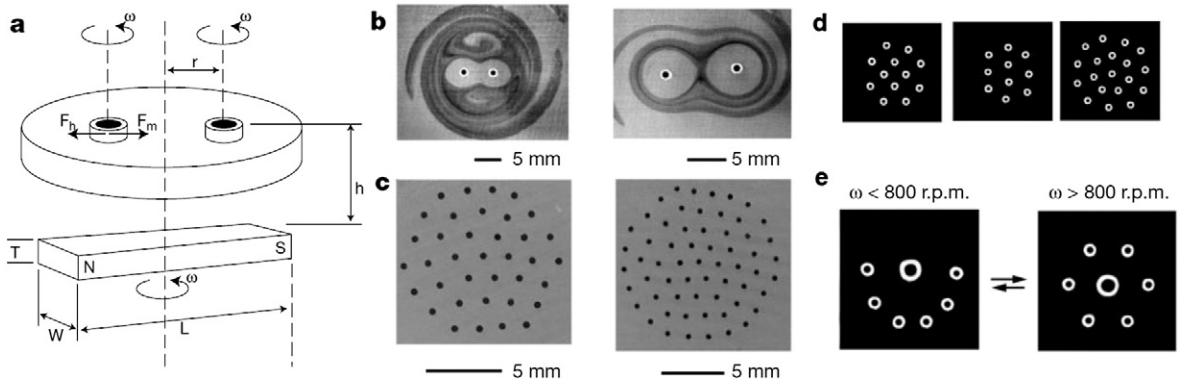

Fig. 1. Dynamic assembly of rotating disks at a liquid–air interface. (a) A scheme of the experiment. A rotating with angular velocity ω bar magnet is used to drive magnetic disks placed on the liquid–air interface. The streamlines are visualized by placing drops of rodamine/water solution. (b) Two 1.27 mm disks spinning at the ethylene glycol–water interface. Left panel: disks are at 700 rpm; right panel:1100 rpm. (c) Hexagonally ordered aggregates of spinning 570 μm disks at 1100 rpm. (d) Various dynamic patterns formed by rotating disks suspended at ethylene glycol–water interface. All disks are spinning around their centers at $\omega = 700$ rpm. (e) Reversible dynamic assembly depending on the rotational speed of the ω. Reproduced from Ref. [53].

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