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, and 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.

Keywords: collective dynamics, dynamic self-assembly, rotors

Preprint submitted to Current Opinion in Colloid & Interface Science November 25, 2015

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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, 4, 5, 6, 7].

A significant work has been dedicated to various aspects of equilibrium colloidal structures obtained as a result of static self-assembly [8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 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, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 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 number of nontrivially ordered dynamic structures have been reported [36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49, 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 torquedriven 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 magnet, see Figure 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, $Re = \rho \omega a^2/\mu < O(1)$, is small but finite). Each particle experience 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. Frequency of the rotating magnetic field can be tuned to adjust the force balance in the system and resulting 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 a 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)mechanically adjust 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 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 application of an alternating (ac) magnetic fields, a magnetic colloidal suspension at liquid interfaces exhibits a strong tendency towards 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 Figure 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 liquidair 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 Figure 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, Figure 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 multiple-swimmers 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, 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 - localised asters- illustrated in Fig. 4.

Dynamic self-assembly in this system is caused by the interactions between ferromagnetic particles responding to an external periodic magnetic field (magnetic torque transfer) and both self-induced interface deformations and hydrodynamic flows in the bulk of the liquids [37]. A short-range magnetic order, governed by dipole-dipole magnetic interactions between the particles, promotes formation of chains. Self-induced hydrodynamic flows with non-negligible fluid inertia(typical Reynolds number for asters is of the order of 10) and waves provide a necessary feedback mechanism that leads to the formation of asters.

The arrangement of chains within an aster is governed by the self-induced hydrodynamic flows and dipole-dipole repulsion of chains. In contrast to magnetic snakes observed at liquid-air interfaces [45, 55, 39], a presence of a top liquid drastically changes the overall force balance and, correspondingly, the outcome of a dynamic self-assembly. An excited circular wave leads to a formation of radial ordering of the magnetic chains, and the chains decorate slopes of the self-induced circular standing wave. Asters are composed of ferromagnetically ordered chains of microparticles decorating circular interfacial wave (the asters net magnetic moment is zero). This arrangement implies two permissible magnetic configurations (flavors): magnetic moments pointing inward, towards the center of the aster, and outward (anti-aster), Fig.4b. Both flavors are usually present in the system.

Asters generate large-scale three-dimensional toroidal flows in both liquids (or in the bottom layer in case of liquid-air interface of high viscosity [62]) as

shown in Fig. 5. This result is in a stark contrast to the quasi-two dimensional flows created by magnetic snakes at a liquid-air interface [58]. Fluid jets, pointing perpendicular to the interface, emanate in both top and bottom liquids. Figure 5a,b shows the amplitude of the flow velocities created by an aster in the bottom liquid (flow in the upper layer has similar structure). The jet's velocity can reach a magnitude up to 2 cm/s and depends on the frequency of the applied magnetic field (higher frequency yields faster jet flows).

Dynamic order and hydrodynamic flows are tightly bound to each other due to a fine balance between particle interactions and self-induced streaming flows. The aster's shape change in response to external stimuli (static in-plane field) results in a controlled collective locomotion of asters facilitated by hydrodynamic jets rather than a magnetic force [37].

2.2. Collective dynamics and assembly triggered by an in-plane field actuation

Yet another fascinating realization of a torque-driven active self-assembly comes from a seemingly trivial system (it was also initially overlooked and filtered out by the author due to 'apparent' triviality)- suspension of ferromagnetic particles at liquid interface energized by alternating field in-plane with the interface (fig. 6a). Kept at out-of-equilibrium condition by alternating uniaxial magnetic field the system demonstrates remarkable complexity [56].

Surface tension confines the ferromagnetic particles with intrinsically pinned magnetic moments to the surface of the water-air interface. An alternating magnetic field applied parallel to the interface exerts torque on them. The torque is dissipated locally in the liquid and generates hydrodynamic flows around each particle. Consequently, the particles interact by two temporally related, but physically distinct types of forces: magnetic (dipole-dipole interactions) and hydrodynamic. Dynamic self-assembly reflects the interplay between magnetic interactions and hydrodynamic flows. The relative contributions of these two primary interactions are modulated by the parameters of the energizing alternating magnetic field [56, 63]. As a result, the system exhibits a remarkable diversity of quasi-stable active states that are dynamic, and exist only while

energy is supplied by the ac magnetic field.

Loose clusters (Fig. 6b) extended along the ac magnetic field are formed at low frequencies. The clusters exhibit periodic changes in shape (pulsations): over time the cluster extends and contracts with a fraction of the driving magnetic field frequency. At elevated frequencies of the applied field, the clusters transform into a cloud of continuously rearranging short chains (Fig. 6c). In striking contrast with loose clusters, the cloud switches the axis of elongation and extends perpendicular to the ac magnetic field. Further increase in the frequency yields a new remarkable dynamic phase: spinners (see Fig. 6d). The spinners emerge via spontaneous breaking of the uniaxial symmetry of the energizing magnetic field. In this phase the particles self-assemble into short chains, and rotate in the plain of the air-water interface at the frequency of the applied ac magnetic field. This rotation creates strong in-plane hydrodynamic flows, see Fig. 6f.g. The spinners exhibit complex dynamic behavior: they move across the surface, collide, disintegrate, and re-assemble [56, 63]. The multi-spinner state (gas) has no apparent preferred direction and covers the entire area of the container uniformly. At higher frequencies of the applied field the spinners give way to dynamic wires (see Fig. 6e). All phases form and disassemble reversibly.

Dynamic spinners are composed of short self-assembled chains of microparticles ferromagnetically ordered due to the prevailing magnetic dipole-dipole inter-particle interactions. Spinners rotate clockwise or counterclockwise with the frequency of the applied field. Since for the case of uniaxial magnetic field the clock/counterclockwise senses of rotation are equally probable, the initial direction is selected by a variety of factors, like interactions with neighboring particles and flows. The rotating chains exert viscous torques on the liquid which trigger strong long-range [64] hydrodynamic vortical flows at the interface. Spinners move (seemingly randomly) due to magnetic interactions and the flows generated by other spinners. Their collective motion creates an overall gas-like appearance of the phase. These rotors collide, disintegrate and reassemble, and thus create complex time-dependent hydrodynamic patterns at the interface.

2.3. Driven magnetic Janus colloids

Torque-assisted active self-assembly has been reported also in ensembles of magnetic Janus colloids [41, 65]. By applying a precessing field to spherical silica particles with a thin nickel film directionally deposited onto one of their hemispheres the magnetic torques were generated that drove the particles to spin around the precession axis and oscillate perpendicular to the rotating plane, Fig. 7a. Subjected to precessing fields magnetic Janus particles demonstrated tendency towards synchronization [41]. At low precession angles long virtually defect-free microtubes spontaneously formed in suspension of these Janus spheres, see Fig. 7b. Structurally these microtubes were achiral and consisted of a staggered set of regular polygons. Once formed these microtubes rolled on the substrate with spheres remained in continual motion. Additional dynamics induced transitions with the frequency of the precessing field have been observed [41].

One of the useful features of the magnetic Janus particles is a shifted dipole moment with respect to a geometrical center of the particle. Recent computer simulation results of the cluster structures in systems of particles with shifted magnetic dipole moments suggested an importance of this additionally introduced anisotropy for the topology of assembled clusters [66, 67]. Subjected to time-varying fields this tunable property could be used to control complex dynamics of the suspension and an outcome of the dynamic assembly. The shift of the magnetic dipole from the geometrical center of a particle depends on the directional coating thickness. The thinner the film the larger the dipole offset from the center [65]. In rotating magnetic fields of moderate strength (2-5mT) magnetic Janus spheres assembled in extended hexagonal planar crystals (Fig. 8) that rotate as a whole with an angular velocity that is fraction of the rotational field [65]. At elevated strengths of the rotational field (about 30mT) unexpected dynamic transition has been observed: single particles formed dumbbells with their magnetic hemispheres pointing inward, see Fig. 8c. Once formed, dicolloids start to rotate around their center of mass. In rotating magnetic fields these dicolloids retain hexagonal symmetry with lattice constant twice as large as that for single particles [65], Fig. 8. Denser aggregates of the dicolloids exhibit more complex dynamic packing, see Fig. 8d.

Janus spherical particles have demonstrated a rich variety of dynamic colloidal superstructures when driven by magnetic torques. Control of the magnetic coating thickness allowed precise tuning of magnetic properties of Janus colloids which govern the symmetry of the resulting dynamic assembles.

3. Quincke rollers: electric field assisted torque driven colloids

Electric field can also be used to exert mechanical torque on colloids taking advantage of electrohydrodynamic phenomenon known as Quincke rotation [68, 69]. Above a certain critical electric field applied to an insulating sphere immersed in a conducting liquid spontaneous symmetry breaking of the charge distribution at the sphere's surface results in a net electrostatic torque that causes the sphere to rotate in a random direction perpendicular to the applied field and also generating hydrodynamic torques in the media. This instability can be effectively exploited to generate large ensembles of rotating particles to tune rheological properties of suspensions [70, 71] and create large ensembles of active self-propelled colloids at solid interfaces [72, 73].

At low densities the rollers formed an isotropic gas-like phase. As area fraction of rollers increases above some critical density a macroscopic fraction of rollers self-organizes in a band and cruise in the same direction, see Fig. 9b. No stationary state involving more than one band has been observed. Remarkably, the velocity of the band (or colloidal flock) is found to be close to the single particle velocity at the front of the band. Comparison of experiments with theoretical prediction suggests that hydrodynamic interactions promote and protect the formation of macroscopic bands at low densities and homogeneous polar phases at high densities [72].

Being subjected to an additional boundary constraints (convex polygonal regions) Quincke rollers may develop a non-equilibrium phase transition into heterogeneous vortex state [73], see Fig. 10. As the area fraction of rollers in

the confinement increases above some threshold value, collective motion emerges spontaneously in the system. Globally ordered state with equally probable left-and right-handed vortices sets in the colloidal ensemble. It was also shown that this vortex state of the system is not very sensitive to a specific geometry of the boundary and rather is a genuine state of the polar active matter [73].

4. Theoretical aspects

Dynamics of driven suspensions of ferromagnetically ordered particles has been investigated by means of Brownian dynamics (BD) computer simulations [74, 75]. There for the case of a bulk system without a hydrodynamic coupling a full non-equilibrium phase diagram as a function of the driving frequency and field strength has been constructed. Appearance of the layered states (similar to previously reported in the system of induced dipoles [76]) has been captured in simulations. Implementation of the hydrodynamic interactions in a quasi-two-dimensional setup in a presence of a rotational driving magnetic field allowed to access and study in detail the interplay between permanent dipolar and hydrodynamic interactions [75]. Results indicated a profound effect of the hydrodynamic interactions on the dynamics of structure formation and behavior of the assembled clusters.

A number of studies have become available that theoretically investigate collective effects in suspensions of particles driven by a Quincke rotation [77, 78]. Within a mean field approximation considering the dynamics of particle orientations in the flow induced by rotating particles a non-equilibrium transition to the polar order at sufficiently high concentration of particles has been established [78].

Some of the aspects of the torque driven dynamics of particles at liquid interfaces has been captured by simulations within Stokes approximation (negligible inertia) [79, 80]. In contrast, the investigation of active rotors in the absence of the hydrodynamic interactions resulted in phase separation of spinners (clockwise-counterclockwise direction) via spinodal decomposition as well as for-

mation of rotating clusters [81]. Most experiments with active rotors strongly suggest the importance of the fluid inertia in the formation of the dynamic order [43, 53]. While the Reynolds number is usually considered low and often of the order of unity in those experiments, the effects of the flows associated with the fluid inertia (such as the Magnus force) are crucial for the resulting collective dynamics and order formation in the active torque-driven ensembles. Thus, in order to capture the relevant physical behavior of the systems with active rotors Navier-Stokes equation has to be solved with finite Reynolds numbers. A few computational algorithms have been successfully applied to tackle the complex out-of-equilibrium dynamic of active rotors in 2d. Fluid particle dynamics (FPD) method [82], force coupling method(FCM) [83, 84], multiparticle collision dynamics (MPC) scheme [85] and Fourier spectral method [86] have been used to treat hydrodynamics in such systems. Hybrid simulations combining fluid dynamics with Molecular Dynamics (MD) for particles [87, 86], fluid particle dynamics method [88] and force coupling method [89] were able to capture in detail the essential dynamics and order formation of the active rotors at 2d interfaces, see Fig. 11.

5. Summary and outlook

Torque driven particles demonstrate remarkably diverse dynamics and tunable active self-assembly. Up to recent years they have been rarely the subject of the detailed studies in a context of out-of-equilibrium self-assembly and self-propulsion. Ensembles of torque driven particles exhibit complex collective behavior often reminiscent of a more 'conventional' active particle systems such as synthetic self-propelled particles [90, 91, 92, 93] or bacteria [94, 95]. Understanding the fundamental principles guiding out-of-equilibrium dynamics in torque-driven system will add an essential tool in a toolbox for assembly of novel materials which might exhibit the multi-level functionality and hierarchical organization that up to now are only found in biological world. Smart materials that can adjust their shape and functionality in response to external stimuli

offer an enormous potential for applications.

Significant progress in the last years has been made in the understanding of a role the hydrodynamics plays in out-of-equilibrium systems. Effects of a fluid inertia (small but finite Reynolds numbers) proved to be important and in most of the cases crucial for understanding complex collective dynamics of particle ensembles at the microscale. To date mostly spherical and disk-like particles have been employed in the studies of active torque-driven systems. The use of shape-anisotropic particles may significantly change the dynamic behavior and outcome of the active self-assembly in the system since hydrodynamic feedback of the particles strongly depends on the shape. This will likely be one of a major research thrusts in the future. Also realization of a dynamic self-assembly powered by torque transfer in three dimensions is a promising research area.

A few efficient computational schemes have been developed to capture hydrodynamic interactions in 2d geometries. A variety of dynamic phases was reproduced and predicted in simulations. A great challenge is to develop more refined models and algorithms incorporating realistic 3d hydrodynamics that to date is still considered to be computationally prohibitive. Access to realistic predictive modeling will enable to speed up and direct the search for future smart materials with bio-mimetic functionalities.

6. Acknowledgment

This work was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division.

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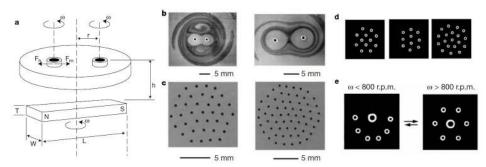


Figure 1: Dynamic assembly of rotating disks at a liquid-air interface. a) A scheme of the experiment. A rotating with angular velocity ω bar magnet is used to drive magnetic disks placed on the liquid-air interface. The streamlines are visualized by placing drops of rodamine/water solution. b) Two 1.27mm 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 μ m 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 ω =700rpm. e)Reversible dynamic assembly depending on the rotational speed of the ω . Reproduced from [53].

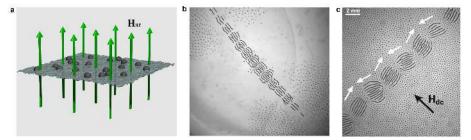


Figure 2: Dynamic self-assembly of torque-driven magnetic particles at liquid interface. a) Schematics of the external magnetic field forcing. Alternating magnetic field is applied perpendicular to the liquid interface. Torques acquired by particles are transferred to the interface resulting in strong vortical flows around each particle and interface oscillations. b) Dynamic multi-segmented self-assembled pattern (magnetic snake) promoted by an alternating magnetic field (110 Oe, 50 Hz.). c) Unusual magnetic ordering of the magnetic snake. White arrows designate directions of the magnetization vector at corresponding segments. Segments composed of ferromagnetically ordered chains of particles are antiferromagnetically ordered to each other. The structure was generated by a 110 Oe, 50 Hz magnetic field. Reproduced from Ref. [45, 55, 43]

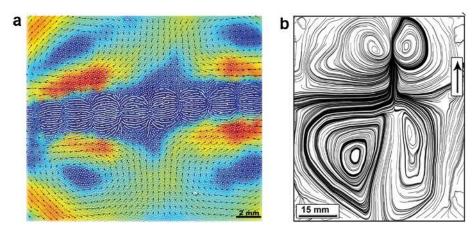


Figure 3: Induced interface hydrodynamic flows by active magnetic particles. a) Flow patterns generated by dynamically assembled magnetic snakes. b) stream lines of the flows produced by self-propelled magnetic snake. The arrow depicts the direction of swimming. Swimmers spontaneously brake the symmetry of the induced quadrupole vortex structure [39]. Reproduced from Ref. [58, 39].

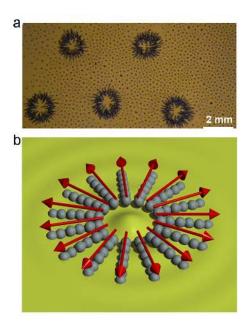


Figure 4: Dynamically assembled asters at liquid-liquid interface. a) array of asters. b) Magnetic order of an aster. Two flavors of magnetic order are permissible: magnetic moments pointing inward and outward (pictured). Reproduced from Ref. [37].

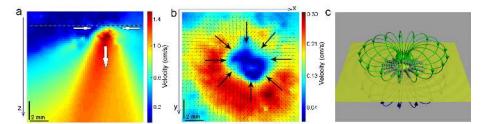


Figure 5: Induced hydrodynamic flows by self-assembled asters. a) Flow velocity field generated by an aster in the bottom liquid layer (vertical slice) obtained by particle image velocimetry. The dashed line depicts the position of the interface, frequency of the alternating magnetic field is 15Hz. Flow direction is shown schematically by arrows. b) Horizontal slice of the flow velocity generated by the aster. Arrows show flow direction. The slice was taken 0.5mm below the interface. c) schematics of hydrodynamic streaming flows induced by an aster structure. Toroidal flows accompany each aster with the liquid jets pointing perpendicular to the interface. Reproduced from Ref. [37].

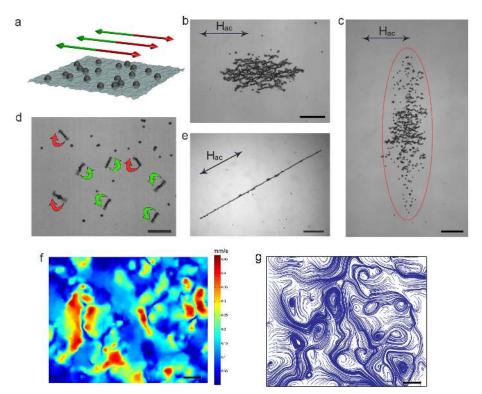


Figure 6: Active dynamics and self-assembly of ferromagnetic suspension at the air-liquid interface with planar magnetic field actuation. a) schematics of the experiment. Uniaxial magnetic field is applied parallel to the air-liquid interface. b) Loose pulsating cluster observed at 20 Hz, 27 Oe magnetic field. Scale bar is 2 mm. (e) Dynamic particle-thick wire formed at 180 Hz, 40 Oe magnetic field. Scale bar is 2 mm. (c) A snapshot of a perpendicular cloud consisting of continuously rearranging short chains. The cloud is extended perpendicular to the axis of the applied magnetic field (40 Hz, 40 Oe) in striking contrast to the cluster phase or dynamic wires. Scale bar is 2 mm. (d) Spinner phase formed at 50 Hz, 29 Oe magnetic field. Short chains of particles rotate in either direction with the frequency of the applied magnetic field. Clock- and anti-clock-wise spinners are illustrated by arrows. Scale bar is 1 mm. f) A typical snapshot of a magnitude of the hydrodynamic flow velocity field generated by spinners at the air-liquid interface. The in-plane applied alternating magnetic field is 29 Oe, 70 Hz. Scale bar is 2 mm. (c) A typical streamline pattern of hydrodynamic surface flows. Spinners produce a complex time dependent vorticity distribution at the liquid interface. Scale bar is 2 mm. Reproduced from Ref. [37].

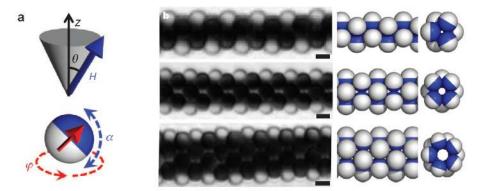


Figure 7: Active synchronisation-induced self-assembly in suspension of magnetic Janus particles. a) Janus particle with the director (red) rotating around precession axis in a precessing magnetic field. b) Experimental images and corresponding model of self-assembled microtubes. Reproduced from Ref. [41].

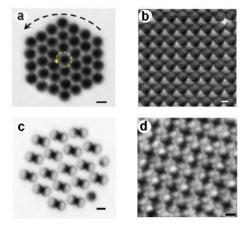


Figure 8: Assembly of magnetic Janus particles in rotating magnetic fields. a) 2d hexagonal crystal in rotating magnetic field (20Hz, 2mT). Black arrow indicates the rotation of the entire crystal. b) Planar crystal with a square symmetry formed by 3 μ m Janus colloids in 20Hz, 5mT rotating field. Scale bar is 2 mm. c) self-assembled crystal of dicolloids. d) snapshot of a square lattice dicolloid assembly. Scale bar is 2 μ m. Reproduced from Ref. [65].

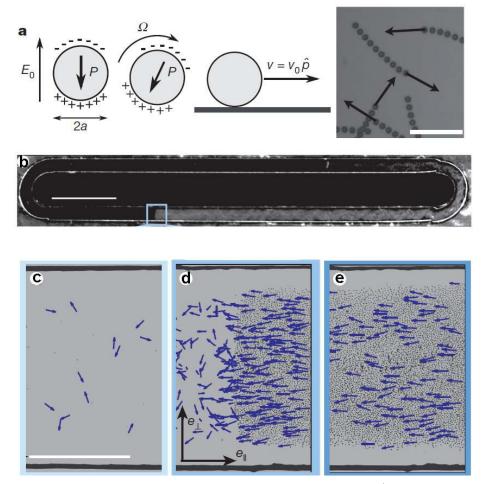


Figure 9: Colloidal rollers dynamics facilitated by Quincke rotations. a) Sketch of the mechanism of the Quincke rotation and resulting propulsion. b) Dark-field image of population of rollers formed a density band propagating along the predesigned enclosed racetrack. c)close-up view of the density band. The arrows correspond to the rollers velocities. Reproduced from Ref. [72].

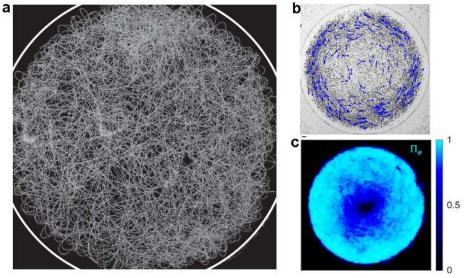


Figure 10: Colloidal rollers inside a circular cavity. a) superimposed images of a dilute ensemble of rollers Colloids follow a persistent random walks. b) snapshot of a dynamic vortex state of rollers. Arrows represent the instantaneous speeds of rollers. c) time-averaged polarization field. Reproduced from Ref. [73].

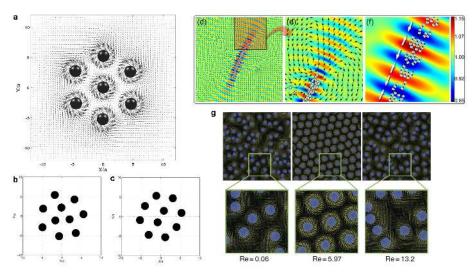


Figure 11: Simulation of active rotators. a) fluid velocity field of the stable agregate of rotating spheres (N=7, Re=2). b,c)stable dynamic patterns for N=10 at Re=2. d) flow generated by magnetic snake in the entire domain. Background color reflects the amplitude of self-induced surface wave [2]. e)flow pattern in the vicinity of the snake's tail. f)antiferromagnetic order of the particles within the snake structure. Reproduced from Ref. [88, 89, 86].