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Collective behavior in out-of-equilibrium colloidal suspensions

Comportement collectif de suspensions colloïdales hors équilibre

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ABSTRACT

Colloidal suspensions, heterogeneous fluids containing solid microscopic particles, play an important role in our everyday life, from food and pharmaceutical industries to medicine and nanotechnology. Colloidal suspensions can be divided in two major classes: equilibrium, and active, i.e. maintained out of thermodynamic equilibrium by external electric or magnetic fields, light, chemical reactions, or hydrodynamic shear flow. While the properties of equilibrium colloidal suspensions are fairly well understood, out-of-equilibrium colloids pose a formidable challenge and the research is in its early exploratory stage. The possibility of dynamic self-assembly, a natural tendency of simple building blocks to organize into complex functional architectures, is one of the most remarkable properties of out-of-equilibrium colloids. Examples range from tunable, self-healing colloidal crystals and membranes to self-assembled microswimmers and robots. In contrast to their equilibrium counterparts, out-of-equilibrium colloidal suspensions may exhibit novel material properties, e.g. reduced viscosity, enhanced self-diffusivity, etc. This work reviews recent developments in the field of self-assembly and collective behavior of out-of-equilibrium colloids, with the focus on the fundamental physical mechanisms.

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RÉSUMÉ

Une suspension colloïdale est un liquide hétérogène contenant des particules solides microscopiques. Les colloïdes jouent un rôle important dans notre vie quotidienne, des industries alimentaires et pharmaceutiques à la médecine et aux nanotechnologies. Il est pratique de distinguer deux classes majeures de suspensions colloïdales : à l'équilibre, et active, c'est-à-dire maintenue en dehors de l'équilibre thermodynamique par des champs électriques ou magnétiques externes, de la lumière, des réactions chimiques, ou un flux de cisaillement hydrodynamique. Alors que les propriétés des suspensions colloïdales à l'équilibre sont assez bien comprises, les colloïdes actifs constituent un formidable défi, et la recherche en est encore à l'etape exploratoire. Une des propriétés les plus remarquables des colloïdes actifs est la possibilité d'auto-assemblage dynamique, une tendance naturelle des composants simples à s'organiser dans des architectures fonctionnelles complexes. Les exemples s'étendent des cristaux et membranes colloïdaux modifiables et auto-réparables aux micro-nageurs et robots auto-assemblés. Les suspensions colloïdales actives peuvent montrer des propriétés matérielles qui ne sont pas présentes dans leurs homologues à l'équilibre, comme, par exemple, une viscosité réduite, une auto-diffusivité augmentée, etc. Ce travail examine les développements les plus récents dans

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le domaine de la physique des colloïdes actifs, dans le but d'élucider les mécanismes de physique fondamentale régissant l'auto-assemblage et le comportement collectif.
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1. Introduction

Colloids are suspensions of macroscopic particles, typically from tens of nanometers to tens of microns, distributed in continuum phase, such as liquid or gas [1]. Colloidal suspensions are routinely encountered in our everyday life (e.g. milk, pigmented ink, blood) and critical to many industries, from food, pharmaceutical, medicine to nanotechnology [2] and electronics.[1] The cross-disciplinary field of colloidal suspensions is an active area of research. Physics of colloidal systems is an important part of the "soft matter", a rapidly expanding field of contemporary science dealing with the physical states easily deformable by external forces, stresses, or thermal fluctuations. Broad scope of soft matter ranges from inanimate colloids, polymers, foams, gels, granular materials, to some biological systems, like suspensions of motile microorganisms, see for recent reviews [4–9].

Forces of various nature govern interactions between colloidal particles, from steric repulsion, electric, magnetic forces (for magnetic colloids), van der Waals forces due to electric dipole moments of colloidal particles, gravity, entropic forces, to hydrodynamic and forces due to gradients of surfactant, etc. [9]. Depending on the specific experimental conditions, interacting colloidal particles may form a variety of steady states, from disordered colloidal glasses [10] and gels [11] to highly ordered colloidal crystals [12]. Ordered colloidal crystals find application as materials with photonic band gap [13,14].

A significant body of work is dedicated to various aspects of mostly equilibrium colloidal structures obtained as a result of static self-assembly [15–27]. Here we survey recent progress in active (or driven) colloidal systems, where formation of dynamic structures occurs under *out-of-equilibrium conditions*. Dynamically self-assembled colloidal structures offer functionality not available under equilibrium conditions: an ability of self-sustained motion (self-propulsion) [28–30], self-repair [31,32], manipulation of cargo particles [33], and many other functions usually associated with living systems [34]. However, there are only a few reviews focused on colloidal systems out of equilibrium [35–38], especially from the point of view of physics.

Collective motion in out-of-equilibrium (active) colloidal systems can be classified according to energy injection mechanisms. One large class is represented by the systems driven by external fields: electric, magnetic fields, or by hydrodynamic flows: the external field exerts *forces or torques* on the colloidal particles, either in the bulk or at liquid/solid or liquid/liquid interface. In the second case the particles are driven internally by chemical reactions, ultraviolet light, etc., see, e.g. [39]. In this situation no net external force is applied to the particle, and the particles are propelled due to generation of local *force dipoles* [40].[2] A large class of self-propelled particles, such as bimetallic chemical microswimmers, asymmetric Janus particles (named after two-faced Roman god), and even a majority of swimming microorganisms belong to this class [41]. It was demonstrated on the example of motile bacteria that a suspension of self-propelled colloidal particles (active swimmers) exhibits material properties different from their equilibrium counterpart: seven-fold reduction of viscosity [42],[3] dramatic increase of diffusivity [46,47], etc.

2. Colloidal systems energized by external fields

Here we consider collective motion in colloidal systems energized by external fields: electric or magnetic. Fig. 1 illustrates some self-assembled states observed in this broad class of systems, from non-equilibrium steady-state patterns, like separated bands and self-healing membranes [32,48] to truly dynamic structures: rotating binary vortices, pulsating rings [49] and self-assembled surface swimmers [30].

Systematic classification of collective motion and self-assembled states emerging in externally driven colloidal systems is not simple: the outcome of self-assembly is sensitive to the amplitude, frequency of the driving field, properties of the suspending fluids, and size and materials properties of the colloidal particles. For example, the system of oppositely-charged colloidal particles in non-aqueous solution subject to a low frequency alternating electric field segregates into bands perpendicular to the applied field direction, [48], see Fig. 1(a)–(d). The bands disintegrate due to the particles' Brownian diffusion once the field is switched off. The authors argue that band formation is caused by collisions between particles moving in opposite directions. The same system behaves differently when a constant electric field of higher magnitude is applied: instead of the static bands positioned perpendicular to the field a phenomenon of lanes formation was observed. Lanes are stripes formed by particles of opposite polarity and moving in opposite directions. In contrast to bands, the lanes are aligned parallel to the applied field [16]. According to [50], the physical mechanism, responsible for lanes' formation, is

[1] Colloidal pigment particles are used in low power consumption electrophoretic displays for e-readers like Kindle and Nook [3].

[2] We will identify active colloids with systems where particles are propelled internally, whereas colloids energized by external fields of shear flow fall into a more general class of out-of-equilibrium systems.

[3] In contrast, suspension of other type of microswimmers (ciliated unicellular algae) exhibits increase of the effective viscosity with increase of the self-propelling velocity [43–45]. The apparent viscosity increase is due to opposite sign of the force dipole generated by the microorganism.

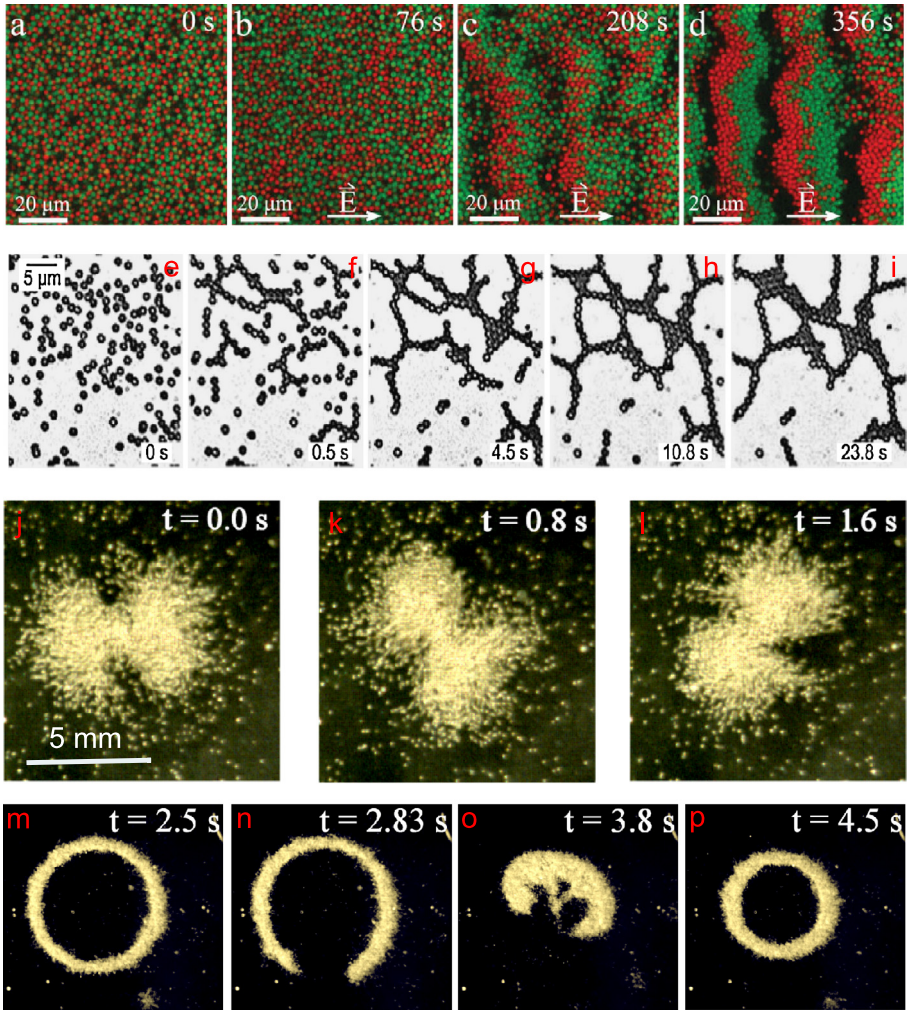

Fig. 1. (a)–(d) Formation of bands of oppositely-charged colloids with a diameter of 1 μm in an alternating electric field with the magnitude $|E| = 17.5$ V/mm and frequency $f = 0.02$ Hz. (a) The mixture is isotropic if no field is applied. Bands start to form after 76 s when the field is switched on, from [48]. (i)–(m) Illustration of self-healing membrane formation in a dilute suspension of superparamagnetic particles (0.15 particles per μm^3). After a rotating magnetic field has been switched on, short chains (0.5 s) appeared, followed by their growth, branching, and formation of a loose network (4.5 s). Remaining unconnected clusters are rapidly absorbed and the network is subsequently coarsened such that the membrane patches grow (10.8 s and 23.8 s), from [32]. (j)–(p) Self-assembled dynamic structures formed by conducting microparticles (100 μm bronze spheres) in a weakly-conducting non-polar liquid in constant (dc) electric field. Images (j)–(l) illustrate rotating binary star-like vortices. The rotating "binary stars" transform into pulsating rings (m)–(p) after the direction of the applied electric field is reversed, from [49].

an enhanced lateral mobility of particles induced by collisions with particles moving in the opposite direction. Once the lanes are formed, a particle's mobility is sharply decreased. The authors argue that the particles in a lane can be regarded as being in a dynamically "locked-in" state. A survey of various instabilities in oppositely-charged colloidal systems can be found in [50].

A simple system of conducting microspheres (100 μm bronze particles) suspended in a thin layer of weakly-conducting liquid (toluene/alcohol mixture) and energized by static (dc) electric field (up to 2–3 kV/mm) exhibit a variety of dynamic self-assembled states [49], see Fig. 1(j)–(p). Depending on the amplitude of dc electric field, its direction with respect to gravity, and the fraction of alcohol in toluene, various states are observed. For low applied field and low alcohol concentration, the patterns are static: clusters, colloidal crystals or honeycomb lattices. For higher applied field values and higher alcohol concentrations, the static patterns transform into dynamics states: self-assembled binary star-like vortices, Fig. 1(j)–(l). Upon polarity reversal of the applied electric field, the rotating vortices transform into chaotically pulsating rings, Fig. 1(m)–(p).

The physical mechanisms responsible for the interaction between particles are *electrophoresis*, i.e. transport of charged particles by electric field in the liquid, and *electroosmosis*, transport of liquid by the electric field. While toluene is a non-polar non-conducting fluid, the presence of small amount of alcohol makes the mixture a weak ionic conductor due to dissociation of the alcohol molecules. For typical experimental conditions the corresponding Debye length is above

1–100 μm, i.e. many orders of magnitude higher than that in water. Consequently, the electric field in the mixture becomes practically unscreened, making it very different from the colloids in aqueous solutions [16,50] where the Debye length is very short, of the order of 10 nm.

Unscreened electric field results in migration of ions in the bulk of the liquid. Conducting particles affect the flow of the ions, resulting in creation of toroidal electroosmotic flows in the proximity of the particles [49,51]. The combination of these vortex flows, electrostatic interactions between the particles, and gravity, results in creation of various dynamic states. The majority of these dynamic states were captured in the framework of phenomenological theory developed in [52]. Experiments conducted with much smaller particles (2–3 μm gold spheres) revealed, in addition to vortices, new dynamic states: self-assembled wires formed along the field direction and assembling/reassembling tree-like structures [53].

A colloidal assembly can be tuned or directed by external magnetic field if the particles possess a magnetic moment, either permanent (ferromagnetic) or induced (superparamagnetic). Static magnetic field generates either chain-like clusters or colloidal crystals [54]. A variety of complex self-assembled structures forms when the suspension of magnetic particles is energized by an alternating magnetic field: networks and quasi-static sheets and membranes [32,55] and dynamic self-assembled swimmers [30]. Magnetic colloidal suspensions can be energized by uni-axial [30], two or tri-axial fields [32,56].

An alternating field, either electric or magnetic, may assemble the dipolar colloidal particles into planar sheets [32, 56–58]. A rotating planar field generates $1/r^3$ dipolar pair interaction between superparamagnetic particles with in-plane attraction (r is the distance between particles) and repulsion along the axis of rotation. This anisotropic interaction facilitates assembly of the particles into flat sheets parallel to the field plane [57]. A tri-axial field case is more subtle: the interaction under a certain condition can become isotropic [56,58]. The effect of precessing tri-axial magnetic field was studied in [32]: the field has a cone opening angle θ_m, controlled by the ratio of the static component of (z-component) to rotating in-plane component (x–y components). In this case the field-induced interaction between superparamagnetic particles decays as α/r^3. The constant α depends on the opening angle θ_m. For small θ_m the colloids behave as dipolar liquid. For $\theta_m \approx 90^\circ$ the colloidal particles experience effective in-plane attraction [57,59]. For the "magic angle" $\theta_m = \arctan(1/\sqrt{3}) \approx 54.7^\circ$ the time-averages interaction term vanishes irrespectively of the relative position of the particles, and the interaction becomes isotropic, attractive, and decays as $1/r^6$.

Ref. [32] reports on a gradual formation of ordered planar membranes consisting of closed-packed particles when a tri-axial magnetic field with the opening angle close to the magic angle, is applied to a suspension of micron-size superparamagnetic particles. A formation of these membranes from a semi-dilute suspension of particles is illustrated in Fig. 1(e)–(i). The particles initially assemble into dimers, the dimers form chains, and Y-junctions. The Y-junctions interconnect, coarsen, and produce membrane patches of all orientations. The membranes show self-healing capability: the membrane reconstitutes its structure after artificial perforation is created. In addition to membranes, a variety of complex patterns were observed in rotating (vortex) magnetic fields, from periodic lattices to honeycomb-like "particles foams" [58], sheets of spinning chains [60], various dynamically-assembled clusters, etc. [61]. Similar phenomenon of planar membrane formation is observed in a precessing tri-axial electric field with the opening angle close to the magic one [19]. Novel staggered chain structures and reversible assembly/disassembly of magnetic Janus particles were observed when both electric and magnetic fields are applied [62]. Remarkable self-assembled microtubes were discovered in the suspension of magnetic micron-size particles with different surface chemistry on opposing hemispheres [63]. The suspension, energized by the precessing magnetic field, forms microtubes in which the particles rotate and oscillate continuously.

Self-assembled structures can be confined at interfaces or near solid surfaces. A variety of dynamic self-assembled states are observed in a dispersion of ferromagnetic microparticles (100 μm nickel spheres), suspended at the water/air interface, and energized by an alternating (ac) magnetic field applied perpendicular to the interface, Fig. 2(a)–(d). Depending on the frequency and amplitude of applied magnetic field, the particles self-assemble into linear snake-like objects (magnetic snakes) [64]. The snakes are formed by several segments arranged along a straight line. The segments, in turn, consist of parallel ferromagnetically-aligned chains of particles. The segments, however, have anti-ferromagnetic alignment. The distance between the neighboring segments is controlled by the frequency of the applied ac field, and the width of the snake is determined by the field's amplitude. The primary physical mechanism of snakes' formation is the interplay between magnetic dipolar interaction of individual particles, promoting chains formation, and hydrodynamic surface flows excited by oscillating chains at the air/water interface.

A symmetry-breaking instability occurs with the increase of magnetic field's frequency: the snakes, immobile for low frequencies, become self-propelled objects for higher frequencies [30]. Due to the fluid's inertia,[4] snake's tails excite large-scale rectified vortex flows [65], each tail serving as a powerful self-assembled pump, see Fig. 2(e), compare to Rayleigh or acoustic streaming phenomenon [66]. For relatively low frequencies of the applied field (below 60 Hz) the snake's flow is symmetric. However, the symmetry becomes spontaneously broken for the higher frequencies, resulting in self-sustained swimming of the snake, see Fig. 2(a)–(b). This mechanism of swimming does not have a direct analog in nature. The symmetry of the snake's large-scales flows can be also broken artificially, by placing a large non-magnetic bead in the proximity of one of the snake's tails. In this case the snake attaches to the bead and is propelled by the remaining free tail, Fig. 2(c)–(d). Many aspects of dynamic self-assembly in this system are captured in the framework of a first-principle quasi-two-dimensional model based on the particles dynamics coupled to surface deformations in thin liquid layer [67].

[4] Corresponding Reynolds number *Re* of the snake's tail flow is about 10.

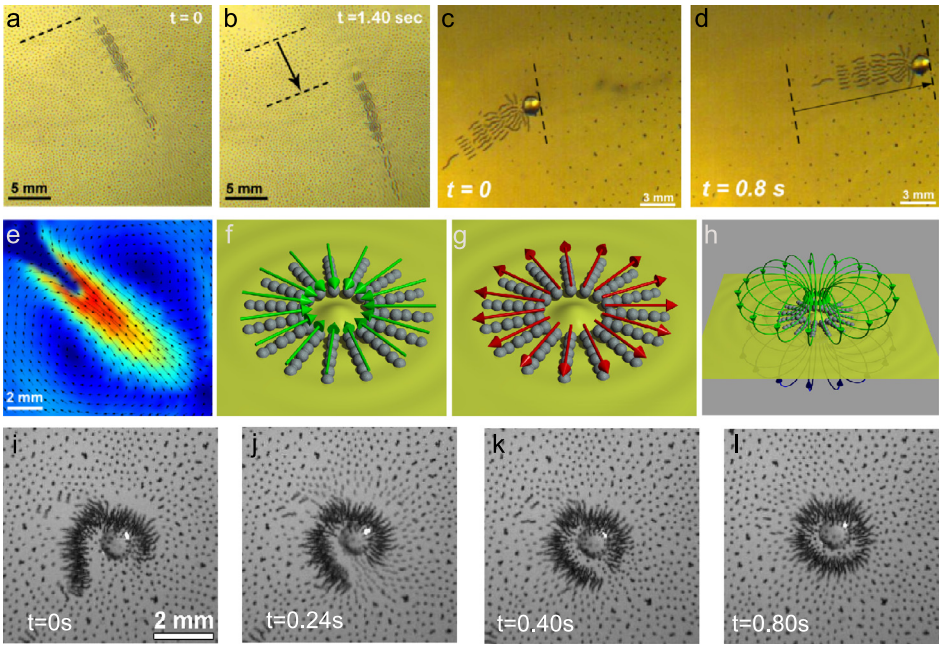

Fig. 2. (a)–(d) Microscopic surface swimmers (magnetic snakes) self-assembled from a dispersion of ferromagnetic particles (80–100 μm nickel spheres) suspended at water–air interface. Homogeneous alternating magnetic field (60–80 Hz) is applied perpendicular to the interface. Images (a), (b) illustrate the magnetic snake formed spontaneously from a random dispersion of ferromagnetic particles, and images (c), (d) show a structure formed by the snake attached to 1 mm non-magnetic glass bead, from [30]. (e) A large-scale vortex flow generated by the tail of a magnetic snake, arrows show streamlines, and colors indicate the magnitude of the surface velocity, from [30]. Illustrations of aster and anti-aster (f), (g) and the toroidal vortex flows generated by the aster in top and bottom liquids (h), from [33]. (i)–(l) Action of self-assembled aster-robot. Ferromagnetic microparticles, confined at the interface between water and oil, self-assemble into a radially-symmetric aster. The asters, remotely controlled by external in-plane dc magnetic field, perform simple robotic functions: capture, transport, and release of large non-magnetic beads [33].

In addition to magnetic snakes, a multitude of novel states is observed when ferromagnetic microparticles are placed at the interface between two non-miscible liquids (silicone oil and water) [33]. For rather large viscosity of the top liquid (viscosity of silicone oil is about 20 times higher than the viscosity of water), a new type of dynamically self-assembled patterns is observed: axisymmetric asters and clusters of asters. The asters are composed of chains with the magnetic moments pointing towards the center (asters) or out of the center (anti-asters), Fig. 2(f), (g). While the asters are immobile, they generate rectified toroidal vortex flow pointed down in the bottom liquid and up in the top liquid layer, Fig. 2(h). Application of a small in-plane dc magnetic field results in controlled self-propulsion of the asters: asters open up and swim in the direction of the field, while asters and anti-asters swim in an opposite direction. It is important to emphasis that the homogeneous constant magnetic field exerts no net force on an aster! The propulsion is an outcome of a reorganization of the aster's magnetic structure in a response on the in-plane field and consequent deflection of the rectified flow from vertical direction. The in-plane component of the aster's rectified flow propels the aster along the field direction. Consequently, asters' shape and swimming direction can be remotely controlled by a small in-plane magnetic field. This functionality enables asters and clusters of asters to perform simple robotic functions: capture, transport, and positioning of target particles [33], see Fig. 2(i)–(l).

3. Internally driven colloids: suspensions of self-propelled particles

In this section we consider internally driven colloidal suspensions of self-propelled particles. The interest to such systems is supported by a growing demand for nanotechnological applications based on autonomously moving devices performing useful function on a microscale, such as delivery of drug-laden nanoparticles to specific cellular targets (targeted drug delivery) or massive parallel assembly of microscopic machines by autonomously moving agents. A number of original design concepts were realized in the recent years, from chemical swimmers energized by catalytic chemical reaction, to swimmers harvesting energy of electric or magnetic field or ultraviolet light radiation, see for review [39].

A few practical realizations of synthetic microscopic swimmers are shown in Fig. 3. A gold–platinum rod, designed by A. Sen and T. Mallouk groups [68,72], swimming in solution of hydrogen peroxide is shown in Fig. 3(a). A typical swimming speed for the 2–3 μm rod is of the order of 6–10 μm/s. Current consensus is that the rod is propelled by self-induced electrophoretic flow powered by the catalytic decomposition of H_2O_2 at the gold/platinum contact. A related design concept, a polystyrene bead half coated by platinum (Janus particle) and propelled by the gradient of osmotic pressure, is shown in Fig. 3(b). The platinum catalyzes the reduction of hydrogen peroxide (fuel) to oxygen and water, producing more molecules

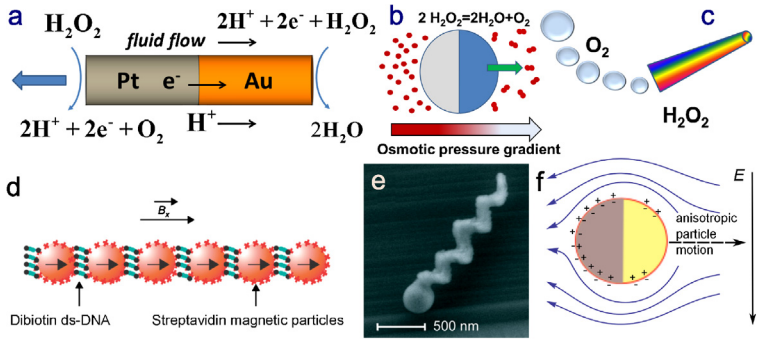

Fig. 3. Practical realizations of microscopic swimmers. (a) Gold–platinum rod propelled by self-electrophoresis in aqueous solution of H_2O_2, from [68]. The potential difference created at the gold/platinum contact in the course of catalytic decomposition of H_2O_2 generates electrophoretic flows propelling the rod. Blue arrow indicates swimming direction. (b) Two-sided (Janus) particle propelled by self-diffusiophoresis in H_2O_2 solution, from [69]. Particles are 1.62 μm polystyrene spheres with one side coated by platinum. (c) High-speed bilayer polyaniline/platinum microtubes grown in the conically shaped pores of a polycarbonate template membrane and propelled by O_2 microbubbles in H_2O_2 solution, from [70]. (d) Artificial magnetic microswimmer. The streptavidin-coated superparamagnetic particles (red cross symbols) form chains along the field (B_x) direction. Double-stranded DNA with biotin at each end binds the particles together. The chain is attached to a red blood cell and energized by ac magnetic field perpendicular to B_x, from [28]. (e) Scanning electron microscope image of an individual glass screw (nanopropeller). A 30 nm layer of cobalt is deposited on one half of the helix. The propeller then is spun by a rotating tri-axial magnetic field, from [71]. (f) A metal-dielectric particle (4 μm polystyrene bead partly covered by gold) is propelled by ac electric field. The electric double layer on the gold side (dark grey hemisphere) is more strongly polarized and thus creates a stronger induced-charge electroosmosis slip than the polystyrene side, resulting in induced-charge electrophoresis motion in the direction of the dielectric side, from [41]. For interpretation of colors, see the online version of this article.

of reaction product than consumed fuel (self-diffusiophoresis). Characteristic swimming speed is about 2–3 μm/s, from [69]. A high-speed microtube conical rocket developed by J. Wang group, UCSD, is shown in Fig. 3(c). The rocket is propelled by O_2 bubbles in the hydrogen peroxide solution and swims up to 350 body length per second, typical rocket size is 5–10 μm [70]. Similar design concept of bubble-propelled particles (nanosubmarines) was developed in O. Schmidt group, Dresden, [73]. Theoretical analysis of the locomotion mechanisms for these types of swimmers can be found in [74,75].

Self-propelled colloidal particles can be energized by alternating electric or magnetic fields. Many of these design concepts were inspired by motile microorganisms, like bacteria or algae. Fig. 3(d) shows artificial microswimmers ("magnetic sperm") [28]. The swimmer is assembled from a dilute suspension of superparamagnetic nanoparticles linked by double-stranded DNA molecules with complimentary biotin/streptavidin protein bonds and attached to a red blood cell. The swimming direction is prescribed by an applied dc magnetic field B_x which orients the magnetic chain (flagellum). The swimmer is energized by the ac magnetic field applied perpendicular to B_x. Typical size of the swimmer is about 5 μm, and speed is about 20 μm/s. Another design of a microswimmer, inspired by a bacterium propelled by rotation of helical flagellum, is presented in Fig. 3(e). The half of a nanoscale glass propeller is covered by ferromagnetic material (cobalt) and brought into motion by a rotating magnetic field [71]. These cork-screw particles can swim up to 40 μm/s. A magnetic swimmer based on a rotation of a magnetic doublet coupled to a boundary interaction was studied in Ref. [29]. Note that the self-assembled snakes [30] are different from all of these designs since they utilize spontaneous symmetry breaking for self-propulsion.

Metal-dielectric (Janus) microparticles can be propelled by an ac electric field [41,76]. In this particular case, the particles move perpendicular to the applied field direction by the induced-charge electrophoresis, arising due to "induced-charge electroosmosis", i.e. the action of an applied electric field on its own induced diffuse charge near a polarizable surface. For typical experimental conditions the speed of particles perpendicular to applied field was the order of 30 μm/s, see Fig. 3(f).

Typical trajectories of individual self-propelled gold/platinum rods are shown in Fig. 4(a). The trajectories are not straight lines, with the orientation of particles changing abruptly, likely due to thermal diffusion and/or interaction with gas microbubbles created by other particles. Additional factors contributing to the apparent diffusive behavior are imperfections of the rods, like roughness of the surface, curvature, etc. Recent studies [77] suggested that these abrupt changes in the swimming direction are mostly caused by the fluctuations-driven flipping of slightly curved rods. This diffusive-like behavior is overall similar to the so-called run-and-tumble motion of some motile bacteria [78,79].[5] One of the fundamental issues in the context of artificial microswimmers is a possibility of collective motion for higher concentrations, similar to that exhibited by swimming bacteria. The bacteria often develop large-scale patterns of collective locomotion, arising purely from collisions and hydrodynamic interactions [81,82], see Fig. 4(b). Collective motion of microswimmers has many advantages compared to individual swimming, for example, from the perspective of collective cargo delivery or harvesting mechanical energy of chaotic motion [83]. To date, no collective behavior similar to that of the bacterial systems was observed in the concentrated suspensions of artificial swimmers. Main reason is possibly a too high rotational diffusion of individual

[5] Some distinctions between angular diffusion of active Brownian particle and run-and-tumble movement of bacteria are discussed in Ref. [80].

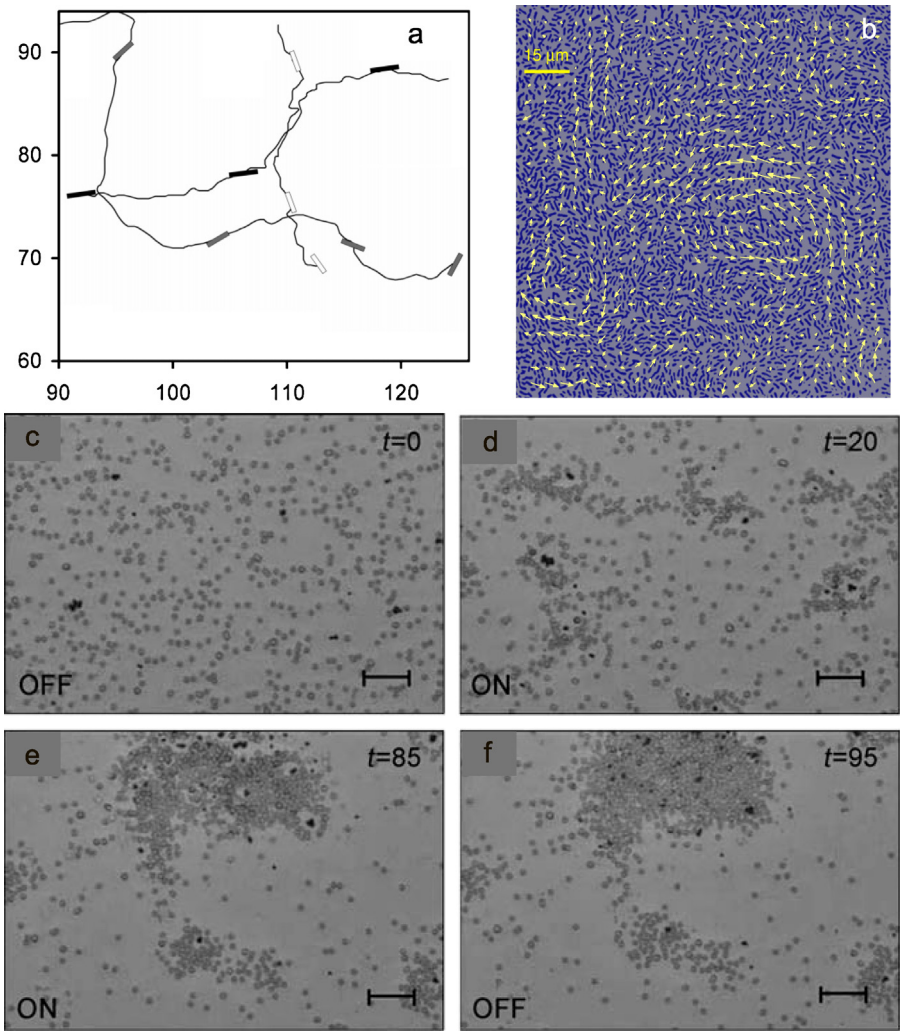

Fig. 4. (a) Trajectories of several individual Au/Pt rods in H_2O_2 solutions, from [72]. Intervals of ballistic motion are interrupted by sharp turns, similar to run-and-tumble motion of motile bacteria. (b) Large-scale collective motion in concentrated suspension of bacteria *Bacillus subtilis*, arrows show the direction of bacterial flow, individual bacteria are seen as short dark rods. Average size of a bacterium is about 4–5 μm, swimming speed of the order of 20 μm/s, image is courtesy of A. Sokolov (color available in the online version of this article). (c)–(d) "Predator–prey" or "schooling" behavior shown by 1-μm AgCl particles (darker objects) and 2.34-μm silica spheres mixed in deionized water. When irradiated with UV light (c)–(e) the silica spheres actively seek out the AgCl particles and surround them. A 2–3-μm exclusion zone is seen around the AgCl particles while the UV light is on; the exclusion zone disappears when the UV light is turned off (f). Scale bars are 20 μm, from [89].

particles destroying the collective swimming state. In addition, gold/platinum particles show a tendency to form permanent aggregates after a collision.[6]

Dynamic clustering of the self-propelled spherical gold–platinum colloidal particles in H_2O_2 solution was observed in [84]. Various dense cluster phases, characterized by merging/separation of particles were observed, with the average cluster size growing as a linear function of the self-propelling velocity. According to Ref. [84], aggregation is primarily caused by the diffusiophoretic chemical interaction between colloidal swimmers, similar to chemotactic aggregation of bacteria. Various aspects of "active" sedimentation of self-propelled particles were studied in [85,86]. In this situation the self-propulsion results in significant increase of suspension's effective temperature. Self-propulsion may also lead to formation of rotating clusters of several active particles [87].

Collective chemical responses of artificial swimmers under ultraviolet (UV) or visual light irradiation were reported in Refs. [88–91]. A behavior, reminiscent of a phototactic response (a tendency of microorganisms to navigate towards or away from light) shown, e.g., by some green algae, was observed when 1 μm silver on silica Janus particles migrated away from UV irradiated regions [88]. An interesting "schooling" behavior was also exhibited by micron-size silver chloride (AgCl) particles

[6] In very concentrated suspensions, the microswimmers also compete for the fuel, which can affect their swimming ability.

upon irradiation by ultraviolet light [89]. These particles can move autonomously in deionized water by self-diffusiophoresis due to asymmetric AgCl decomposition under the UV light. Since moving AgCl particle releases ions, other particles respond by drifting or "schooling" into regions with higher particle concentrations. When photo-inactive silica particles are mixed with the AgCl particles, they respond to the ion release by swimming towards and surrounding individual AgCl particles, see Fig. 4(c)–(f). Collective chemical oscillations were also observed in suspension of AgCl particles in the presence of UV light in a dilute hydrogen peroxide solution [90]. Both single-particle and collective, multi-particle response, were observed due to an oscillatory, reversible conversion of AgCl to silver metal at the particle surface. These light-powered microswimmers self-organize into clusters of oscillators with significant spatiotemporal correlations in the clusters.

Two-dimensional "living crystals", which form, break, and reform, were observed in suspensions of active bi-material (Janus) particles [91]. These particles exhibit self-propelled motion at the experimental cell surface (e.g. glass slide) when illuminated by blue light in H_2O_2 solution.

4. Conclusions and perspectives

A rapidly expanding and developing field of out-of-equilibrium colloids promises many scientific and technological breakthroughs. One of the intriguing research directions is a collective behavior of shape-anisotropic colloids, like rods, platelets, or more complex shapes, e.g. chiral. Similar to liquid crystals [6], where a variety of non-trivial phases arises due to shape anisotropy of the molecules, we can anticipate a rich spectrum of non-trivial dynamic states when suspensions of anisotropic particles are energized by electric or magnetic fields. These states, showing unique optical and mechanical properties, will play a bigger role in reconfigurable smart materials for emerging technologies based on self-assembly, from photonics [13,92], to microfluidic machines [59] and robotics [33,93].

Artificial swimmers is another promising research area. Technical progress in the design of swimmers, optimization of their swimming abilities, and functionalization for specific applications will be likely one of the major research thrusts. Currently most of the swimmers' designs are based on the hydrogen peroxide propulsion, that makes them not suitable for the most of medical applications related to targeted drug delivery. The use of the biologically common fuels, such as glucose, is highly desirable.

A significant fidelity increase of the numerical algorithms for active colloidal suspensions is anticipated due to revolutionary increase in computing power. The majority of current theoretical approaches is based on direct discrete particles simulations with stochastic forces due to thermal fluctuations, whereas hydrodynamic interactions are included on a highly simplified level, mostly via viscous friction acting on the particle, see e.g. [58,67,94,95]. More adequate treatment of the hydrodynamic forces between colloidal particles is highly desirable, but accurate modeling of the fluid flow generated by multiple arbitrary shaped particles is still computationally prohibitive. In addition to discrete particle simulations, a number of continuum phenomenological approaches was proposed to describe dynamic states, like rotating binary clusters and self-propelled magnetic snakes [30,52]. Other theoretical approaches are based on phenomenological hydrodynamic-type equations for out-of-equilibrium systems (active gels) [96,97]. Such generalized hydrodynamic approaches, while rather powerful in principle, have some limitations when applied to active colloidal systems. Namely, formal absence of the scale separation between the size of mesoscopic colloidal particles and emerging large-scale pattern diminishes the validity of the hydrodynamic approaches. Thus, further progress is also expected in the development of coarse-grained description for the dynamic states in colloidal suspension.

From a theoretical perspective, there is constantly growing interest to the analysis of generic dynamic and statistical properties of collective behavior exhibited by self-propelled particles with various kind of interactions, from point-like particles to rigid rods, mostly near the threshold of some symmetry-breaking instability, see e.g. [98–103]. A variety of dynamic phases was predicted, both analytically and numerically, from moving clusters, bands, to various swarming states. However, to date, the connection between these simulations and experimentally observed dynamics of self-propelled colloids remains not fully satisfactory. Future more refined mathematical models will likely incorporate on a more realistic level the multitude of forces acting on self-propelled colloidal particles.

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