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Synthetic electrically driven colloids: A platform for understanding collective behavior in soft matter

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Abstract

The collective motion of synthetic active colloids is an emerging area of research in soft matter physics and is important both as a platform for fundamental studies ranging from non-equilibrium statistical mechanics to the basic principles of self-organization, emergent phenomena, and assembly underlying life, as well as applications in biomedicine and metamaterials. The potentially transformative nature of the field over the next decade and beyond is a topic of critical research importance. Electrokinetic active colloids represent an extremely flexible platform for the investigation and modulation of collective behavior in active matter. Here, we review progress in the past five years in electrokinetic active systems and related topics in active matter with important fundamental research and applicative potential to be investigated using electrokinetic systems.

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Abbreviations

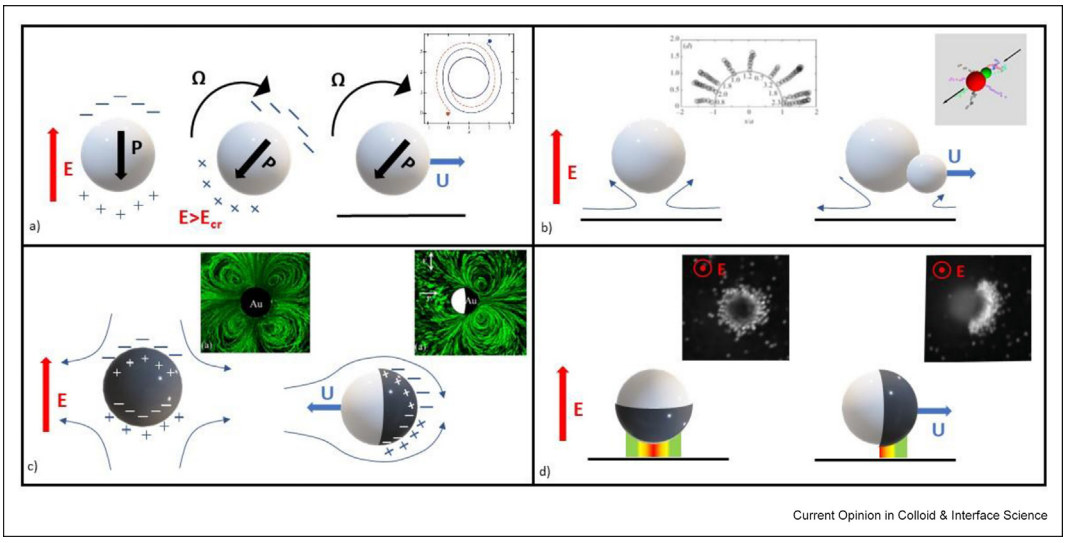
EHD, Electrohydrodynamic; DEP, Dielectrophoresis; pDEP, Positive dielectrophoresis; nDEP, Negative dielectrophoresis; ICEO, Induced-charge electroosmosis; ICEP, Induced-charge electrophoresis; sDEP, Self-dielectrophoresis; JP, Janus particle; EK, Electrokinetic; EDL, Electric double layer; TRS, Time-reversal symmetry; ABP, Active Brownian particle; AOUP, Active Ornstein-Uhlenbeck particle; MIPS, Motility-induced phase separation.

Introduction

To define collective motion, it is instructive to look to nature. Consider a single bird, which either flits along random pathways or moves deterministically to achieve a goal such as feeding. However, when several birds congregate, they move in specific macroscale patterns — flocking, swarming, etc. These patterns are examples of collective motion, and their manifestation in both living and synthetic systems confirms that often the motion is not driven by any form of consciousness but rather arises from the interactions of multiple bodies and the fields surrounding them. The onset of these patterns is a defining characteristic of both biological and synthetic active matter and is traditionally associated with organisms/particles that self-propel, i.e., the energy source comes from within the particle itself (e.g., biochemical for organisms and magnesium/zinc core for active particles) or is external (e.g., chemical fuel in the solution) to it and harvested at its surface (e.g., catalytic reactions). Active motion can also arise in response to external forces (e.g., electric/optic/magnetic/acoustic fields), provided that the propulsive mechanism—hence, direction and magnitude of the active velocity component—is produced by symmetry breaking at the particle level. This distinguishes such systems from the "phoretic" linear response driven by external gradients in which all colloids move along the field gradient (e.g., electrophoresis, magnetophoresis, and diffusiophoresis). Pattern formation and long-range spatiotemporal order in the collective motion of active matter are ubiquitous across diverse systems and scales. Thus, robust, reproducible, and tunable experimental active systems are invaluable tools in the study of emergent phenomena.

Within this review, we focus on electric fields as the external energy source for powering self-propulsion and illustrate their unique advantage in terms of the range of flexibility and control offered by manipulation of the field properties—signal, amplitude, and frequency—to modulate independent particle mobility and multibody interactions. In addition, the electric field is a facile energy source that does not suffer from biocompatibility and finite life of chemical fuels. There are four primary physical mechanisms that are used to drive active particles in electric fields (Figure 1). (a) Beyond a critical voltage, weakly conducting dielectric particles undergo spontaneous rotation due to the Quincke instability yielding a translational component when adjacent to a wall [1,2]. (b) The proximity of a particle

Figure 1



Active particle self-propulsion driving mechanisms: (**a**) Quincke effect: Dielectric particles spin when the field exceeds a critical value; translating when adjacent to a wall. Inset: Simulated path lines of two Quincke rollers [1]. (**b**) Asymmetric electrohydrodynamic flow between the particle and electrode substrate yields translation. Inset: Path lines of tracer particles around a dielectric sphere [3] and asymmetric doublet [4]. (**c**) Breaking the symmetry of quadrupolar induced-charge electroosmotic (ICEO) flow leads to the translation of a metallodielectric Janus sphere: Inset: uPIV images of ICEO flow around a stagnant gold and Janus particle [8]. (**d**) Non-uniform gradients below a metallodielectric Janus particle cause the particle to move under self-dielectrophoresis. Inset: Tracers are attracted to regions of high field strength under positive dielectrophoresis.

to an electrode also distorts the local electric field to produce electrohydrodynamic (EHD) flow at the electrode surface [3]; breaking the symmetry of this flow through the particle properties will cause the particle to translate [4]. (c) Polarization of a conducting particle subject to a uniform field is countered through the development of an induced electric double layer (EDL) at the particle surface. The field acting on the diffuse charge it induced within the EDL results in non-linear (quadratic) electroconvection in the form of a quadrupolar hydrodynamic flow termed induced-charge electroosmosis (ICEO) [5]; breaking the symmetry of this flow at the particle surface causes the particle to translate under induced-charge electrophoresis (ICEP) [6]. (d) Finally, non-uniform gradients induced by particle-wall proximity can also cause translation under electrostatic self-dielectrophoresis (sDEP) when the symmetry is broken [7].

Beyond these mechanisms, random thermal fluctuations, particle–particle, and/or particle–wall interactions (which may also be functions of the electric field) conspire to provide local mechanisms for the emergent correlation of the active particles' velocities, giving rise to the ubiquitous patterns of collective active matter. Modeling the response is complex and requires a symbiosis between theory, numeric, and experiment and interdisciplinary knowledge that is not always readily available. Several theoretical paradigms are currently being employed to analyze and predict these patterns, ranging from multi-particle Langevin equations to effective thermodynamic continuum approaches, using both "classical" numerical methods and machine-learning approaches to obtain solutions.

Focusing on advances in the past five years, we review the current state of the art in experiment, theory, and simulation of active electric-field-driven colloids and related systems, concluding with a discussion of applications and perspectives on open questions and future directions.

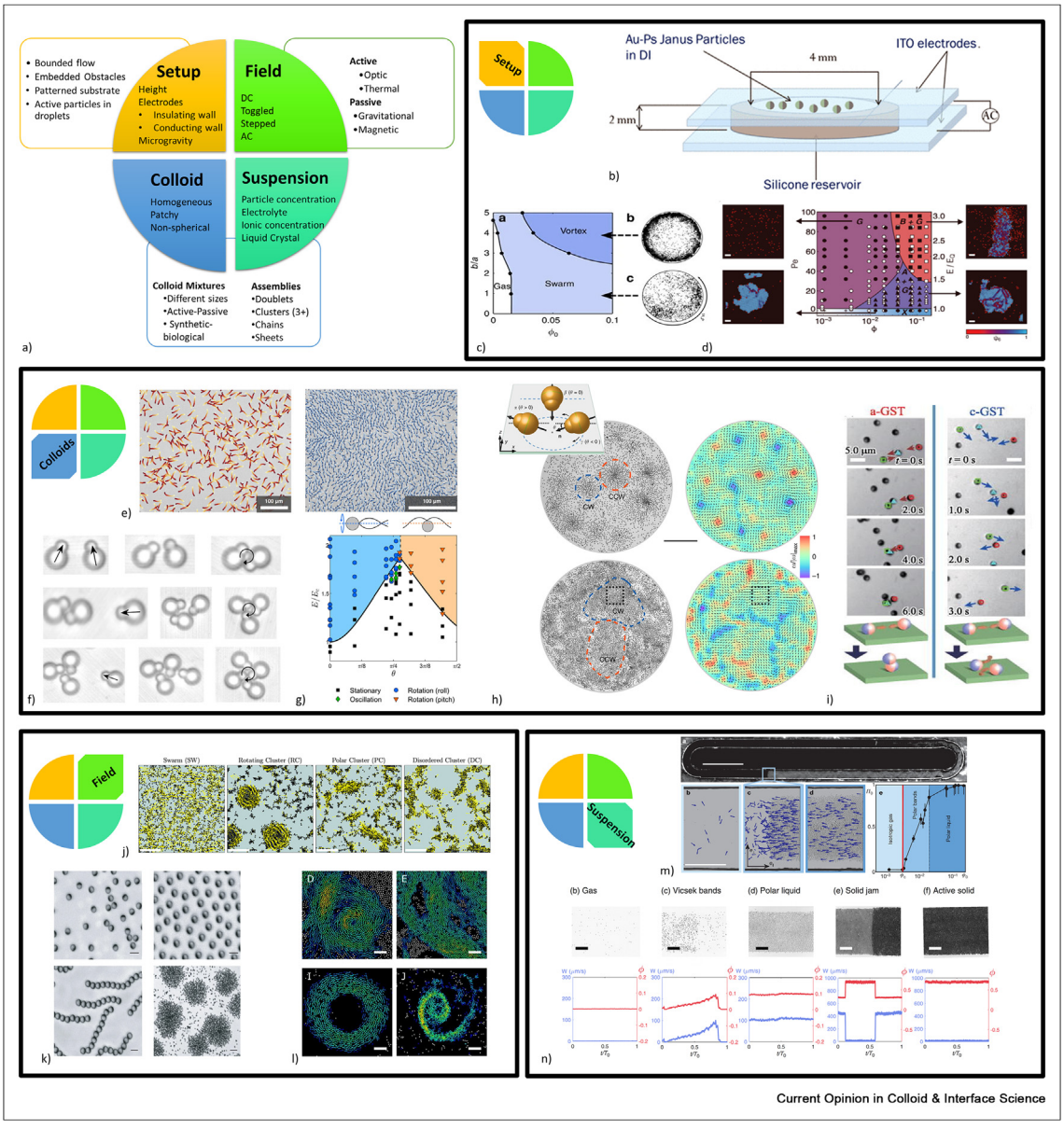
Experimental colloidal systems

The scallop theorem [9] states that bodies which deform in a reciprocal manner (have one degree of freedom) will not move in a viscous environment (corresponding to low Reynolds numbers). In nature, critical symmetry breaking [10] is commonly achieved through shape distortion or flagellar propulsion. In microscale synthetic systems, which are also characterized by low Reynolds, asymmetry is most commonly introduced through the geometry or material properties of the active colloid [11] but can also arise from asymmetry in the fluid or the system geometry; all potentially affecting the self-propulsion force. Accordingly, the basic experimental setup of a colloidal system is informed by the choice of applied field + colloid + suspending medium (Figure 2). In the ensuing section, we consider each of these factors separately and then discuss systems in which multiple factors are employed simultaneously.

Role of the applied field

Active colloids can be driven by both direct current (DC) and alternating current (AC) electric fields with the resultant motion—both individual and collective—strongly dependent on the field properties. As the field can be externally controlled, this characteristic underlies the key advantage of electrokinetically driven systems—dynamic modulation in situ and

Figure 2



Key components of active matter experimental systems: (**a**) Schematic of key components of experimental systems. (**b–d**) **Setup:** (**b**) Sample experimental setup where electric field is applied perpendicular to ITO substrate, (**c**) transition from gas/swarm/vortex in a confined circular chamber depends on volume fraction [44], (**d**) near-wall proximity causes dielectric spheres to interact through long-range EHD induced by near-wall proximity and the Quincke effect resulting in various phases which depend on the voltage of the applied field [12]. (**e–i**) **Colloid composition:** (**e**) Path lines of Janus particles traveling in disordered and ordered states under AC fields [21], (**f**) assembly of dimers into chiral structures depends on the direction of EHD flow at each lobe [4], (**g**) state diagram of biflagellated Quincke rollers [27], (**h**) clockwise and counterclockwise vortices coexist in the suspension of Quincke dimers with the size dependent on volume fraction [26], (**i**) particle–particle interactions of GST-coated Janus spheres depend on whether GST is in amorphous (a-GST) or crystalline (c-GST) phase. (**j–l**) **Field effects:** (**j**) Stepped DC fields with different relaxation times result in different modes of collective motion [14], (**k**) variation of frequency of AC field causes particles to transition from swarms to chains to clusters [16], (**l**) collective motion of Quincke rollers (freestanding donut and spiral swarm) form at subcritical electrical fields [13]. (**m–n**) **Concentration and constitution of suspension:** (**m**) Quincke rollers transition from directed to collective motion as volume fraction increases [2], (**n**) Quincke rollers transition between directed motion, flocking, and jamming states depending on local volume fraction [45].

the realization of multiple modes of behavior in a single platform.

The most common DC-driven active systems are Quincke rollers (Figure 1a) [2]. Since the instability only emerges above a critical threshold electric field value, these systems readily transition from passive to active [12], although collective motion may be observed before individual motion [13]. The magnitude of the field and the volume fraction governs the transition between modes, including the formation of passive crystals and polar bands of Quincke rollers propagating through an "active colloidal gas" [12]. Square wave electric fields can drive random walk of Quincke rollers when the frequency is greater than the Maxwell–Wagner charge relaxation [14]; toggled fields can modulate particle–particle interactions and resultant assemblies [15].

In AC fields, the individual particle mobility and collective motion strongly depend on the applied field frequency which dictates the driving mechanism—EHD (Figure 1b), ICEO (Figure 1c), and sDEP (Figure 1d)—and electrostatic interactions contributing to collective motion [16] and assembly [17]. At low frequencies, on the order of the inverse of the electrode charge relaxation time, long-range EHD flow induced by particle-wall proximity [3] (Figure 1b) controls particle aggregation and assembly [18] and drives geometrically asymmetric particles [19]. At frequencies on the scale of the inverse of the charge relaxation time around the particle, metallodielectric (in which one hemisphere is conducting and the other dielectric) Janus particles (JP) translate with the dielectric hemisphere forward due to ICEP [6,20] (Figure 1c), reversing direction to travel with the metallic hemisphere forward at higher frequency sDEP [7] (Figure 1d). The combination of altered swimming direction and electrostatic interactions affects the modes of collective behavior which transition from swarming to clustering and can be either rotating, polar, or disordered [21]. The field frequency also affects the dynamic assembly of active colloids into hierarchical structures, such as JP doublets [22] and chains [23], through DEP.

Colloid design

The Quincke effect is unique in that active motion can be attained with dielectric spheres that are homogeneous and symmetric [2]. The introduction of geometric asymmetry introduces a rotational component to the velocity; dimers and trimers travel in circular orbits and flip-flop, respectively [24], while the handedness of pear-shaped colloids [25,26] yields chiral flow at the macroscale. Biflagellated Quincke rotors self-oscillate due to elastohydrodynamic-EHD instability [27].

For the EHD, ICEP, and sDEP mechanisms, symmetry is commonly broken through the material properties (and in some cases, also the geometry). Patchy colloids are a general class where multiple properties—material, electrical, and chemical—are integrated into one particle. The simplest version is a Janus sphere where each hemisphere has different properties. For EK-driven systems, the contrast between hemispheres usually relates to the frequency-dependent relative permittivity and conductivity of the materials. Metallodielectric JPs—where one hemisphere is conducting, and the other hemisphere is dielectric—represent the maximum asymmetric contrast, and both the mobility of individual particles [7] and their collective motion [16,21] vary as a function of voltage and frequency. The weight of the metallic coating generally causes particles to sediment to the bottom of the chamber resulting in two-dimensional (2D) planar systems, with a notable exception being the work of Sakaï et al. [28] where the thin coating and addition of glycerol to counteract buoyancy allowed for three-dimensional (3D) assembly. Coating thickness also determines the induced dipole moment [29,30] and stable orientation [30,31]. Adding dielectric surface coatings, e.g., SiO_2, modifies individual polarizability and dipolar particle–particle and particle–wall interactions [16]. Although the limiting contrast of metallodielectric spheres will yield maximum ICEP velocities [32], net motion can be achieved with any contrast as evidenced in Ref. [33] where particles coated in dielectric, amorphous phase chalcogenide GeSbTe (aGST) exhibited similar ICEP mobility to Au-coated particles. Notably, significant differences in the collective motion of aGST and the crystalline-metal phase cGST coated particles were observed since even at high frequency as aGST—in contrast to cGST—did not exhibit repulsive interactions but rather stacked vertically due to similarity of dipole moments in the two hemispheres.

As with the Quincke rollers, particles that are **non-spherical** (ellipsoids [34,35], dimers [19], and doublets [22]) can be designed to interact and translate in complex paths, including rotation in addition to linear translation. Geometric asymmetry is also key to active motion at low frequencies due to asymmetric EHD between the particle and substrate [4,19] (an electrostatically asymmetric metallodielectric JP will remain stagnant in this frequency domain as the Debye layer screens the metallic hemisphere, such that particle-induced EHD flow mirrors symmetric flow around an dielectric sphere [3,36]). Complex shapes can either be fabricated a priori [25,37], aggregate from electrostatic interactions [24], or form dynamically in situ due to EHD [18], dipolar interactions [38], or dielectrophoretic trapping [39]. Collisions can lead to hierarchical self-assembly of more complex structures [24]. Colloid design for experimental systems could be optimized through numerical simulations, particularly in looking toward the attainment of 3D motion. For example, it has

been proposed that Quincke-driven helical particles could translate even in the absence of a proximate boundary [40] while electrokinetically driven colloids with multiple metallic patches could move against gravity out of the confining 2D plane [41].

Finally, we note that although our focus here is on synthetic active colloids, we note that AC fields have also been used for dynamic assembly of biological systems such as bacteria [42] and E-coli through the control of polarized interactions and EHD flow [43,169].

Effects of suspension properties

The third key component of the experimental system is the suspension, which is defined here to include the colloids and the suspending fluid. The **concentration of active colloids** will dictate the importance of short-range electrostatic interactions, pair alignment [46], polar order [21], and the frequency of particle−particle collisions which contribute to collective motion. As the volume fraction increases, particles transition from random walk to collective motion [2]. At very high concentrations, solutions become jammed [45] or crystallized and cease to be active. The aggregation will also affect motility as the effective friction or Stokes' drag varies [39,47]. Focusing on the suspending fluid, **increasing conductivity** has been shown to affect EHD [48] and ICEP due to effects such as the Stern layer capacitance and enhanced surface conductance [49]. The addition of salt also tunes the dipolar reactions as the relaxation time of the induced-charge shifts to higher frequencies, can alter the formation of chains and their mobility [23], or introduce new structures not evident in deionized (DI) water [16]. Varying the fluid density via the addition of glycerol has been used to density match active colloids [28] allowing for 3D activity. It has been proposed that **asymmetric electrolytes** could be used to control the direction of induced-charge electroosmotic flow [50]. In **liquid crystals**, the mobility of the active colloid depends on the direction of the applied field and the local orientation of the direction [29,51−53]. A challenge with many complex and non-Newtonian fluids, including biologically relevant fluids such as blood, is the high electric conductivity which impedes motion driven by electric fields.

Multiple symmetry breakers

Increasing the complexity of experimental systems yields phenomena otherwise unobservable. **Integration of a secondary field** can induce a second active or passive (linear) response to imbue additional functionality. Magnetic fields direct motion through dictating interface orientation [39,54,55] and by extension particle−particle interactions. Optical fields control the mobility of active droplets filled with bacteria [53], rectify independent [56], and collective motion of EHD-driven colloids [18], as well as increase the mobility of photocatalytic JPs through alignment [57]. Acoustic fields were used to counteract gravity and levitate 2D colloidal sheets which then interacted in three dimensions under an AC field [58]. Gold@-TiO2−SiO2 Janus "nanotrees" can be driven by one or a combination of optical, thermal, and/or electrical fields [59]. Additionally, it is important to note that in most systems, inherent **secondary fields** exist that can cause departure from theoretical models of EK phenomenon in isolation. Two common examples are laser thermal heating which can result in self-thermophoresis [60−62] and gravity which causes the commonly used metal-coated JPs to orient with metal side down [63] and aggregated flocks of colloids to settle [64]. Experimental isolation of these phenomena is key to understand individual and collective motion suggesting the necessity of creative solutions such as microgravity experiments [65], much like the situation for passive colloids [66,67].

The interaction of active colloids with boundaries is another important factor affecting both the individual mobility and phase transition. Unconfined Quincke rollers form colliding band-shaped swarms [2] but would display a single vortex pattern [2,44] if bounded. The presence of embedded obstacles or an external flow would alter their dynamic steady states, such as the frequency of oscillating chiral flow for the former [69] and the appearance of a hysteretic response in confined active flocks for the latter [70]. Substrate patterning via chemical functionalization [47] or physical surface roughness will further modulate both individual and collective motion. For a metallodielectric JP, near-wall proximity can drive mobility reversal [7] and dielectrophoretic assembly of complex colloids [71]. Additionally, active colloids bounded in liquid droplets deform the boundary as they move collectively, with the resultant shape fluctuation exhibiting a power spectrum that correlates to the particle-interface collision [72].

Perhaps, the richest array of phenomena is obtained by introducing multiple species of active or passive matter into a single system, e.g., mixtures of **different dimensions** or material properties, **mobile and stagnant active particles**, **active-passive** mixtures, and **synthetic-biological** mixtures. The interaction of active and passive colloids depends on the driving mechanisms [73−76] and relative proportions of the mixture. From a purely hydrodynamic perspective, the direction of flow around the active colloid dictates whether nearby particles accumulate (inward flow) or are depleted (pushing/repulsive flow) [4,73,74,77]. Dynamic assembly in electric field-driven systems has been demonstrated for mixtures of different-sized passive homogeneous spheres resulting in active aggregates [19]. More complex structures can form in active-passive mixtures

[17,38,39,76] and synthetic active colloids with biological cargo [17,51,53,78]. In EK systems, accumulation can also arise from EHD flow between the particle and wall and/or dielectrophoretic trapping due to localized non-uniformity of the electric field [39,71] arising from particle-wall proximity [71] or the asymmetry of the active colloid [39]. The mobility of the aggregate structure varies due to changes in active surface area and effective radius [39,47]. The resultant structures can interact with each other, and other particles suspended in the medium to enhance stability [76] or even exchange cargo [39].

Fundamental mechanisms contributing to collective behavior

Critical open questions concern how particles interact with each other, external fields, the surrounding bath, and confining walls or boundaries [79,80] to yield a diversity of emergent, collective behaviors with a variety of long-range structures. Also, there is the practical question of how best to incorporate EK and other many-body interactions into a tractable theoretical/computational framework for a specific problem.

For active and passive charged colloids alike, screened electrostatic interactions provide intermediate−range interactions between particles, walls, ions, and external fields, coupling to the hydrodynamics through EK interactions (e.g., Maxwell and osmotic stresses). Thus, many of the same considerations regarding EK interactions apply to both active and passive colloids. While mean-field EK theory is well-developed for isolated particles in simple, binary electrolytes, effort must be taken to evaluate potentially subtle effects which alter the usual Debye layer picture and may become especially important for dense suspensions of active colloids. In addition to linear EK effects, largely responsible for passive colloid electrokinetics (i.e., electrophoresis and colloid stability), the EDL, Stern layer [48], and surface conductance [81] mediate nonlinear EK interactions, such as DEP motion of a polarizable body in a non-uniform electric field [82], EHD [4], ICEO [5,83], and ICEP [6]. So, for example, non-negligible dependence on ion size [49,84] can introduce corrections to the charge relaxation frequency of the particles, electrodes, and suspending solution. Colloidal stability involves contributions to the free energy from the bulk electrolyte and the colloid−Debye layer electrolyte interactions in addition to the screened pair potentials [85]; the electric double layer in high-density colloids is no longer in equilibrium with an infinite ionic reservoir. Electrostatic imbalance between the induced dipoles within the opposite hemispheres of the JPs has been shown to determine the stability and interactions of active colloids [16,33]. Likewise, the EK response of active particles with broken electrostatic [32] and/or hydrodynamic symmetry [35] may resemble that of non-spherical passive colloids [86]. Many of these considerations remain insufficiently explored to be implemented in a fully many-particle theoretical framework.

The Debye-shielded electrostatic interactions will generally be shorter ranged than hydrodynamic interactions, and long-range effects are mediated through EK coupling. Active colloids near interfaces can experience Marangoni-like flows due to gradients [87]. Different modes of propulsion (e.g., “pullers” vs. “pushers”) can induce different hydrodynamic interactions [73,77,88]. Active EK colloids can function in both modes [4], thus providing a means of modulating hydrodynamic interactions. The proximity of active colloids to a conducting wall will result in EHD [3] which can drive active motion [19], distort ICEP flow [36], and dictate long-range interactions [4]. Particle−wall interactions typically include multiple forces, e.g., both hydrodynamic and electrostatic contributions leading to differences in the bulk vs. confined stability of passive colloids [85]. Competition between many-body hydrodynamic interactions and other (many-body) phoretic interactions can influence the local slip velocity of an individual particle. Near a wall with the irrotational flow, the particle−wall interaction permits the derivation of non-equilibrium pair potentials, the gradient of which yields a force [89]. The particle−wall and confining field interactions for surface anisotropic particles, like active EK colloids, can be utilized in assembly [79]. Moreover, these near−wall interactions lead to strong, non-uniform field gradients which can modulate the mobility [7,90,91], modulate particle−particle interactions via DEP [39,71], lead to anomalous diffusion [92], modulation of collective behavior [93], and, in general, play a critical role in the collective (bulk) behavior of active matter [80,94]. On Earth, gravity is the chief confining field, leading to sedimentation effects for aggregates of passive colloids [64] and difficulty in observing very large colloidal crystals; the effect on active colloids is to confine them to 2D, and distinct behavior is expected in 3D [28]. This motivates several questions about the role of hydrodynamic effects from hard boundaries versus confining fields, whether sedimentation can be mitigated by external fields, and how such collective motion differs from unconfined, 3D motion as approximated by, for example, microgravity experiments.

Owing to low-Reynolds numbers, active colloidal motion is often modeled as overdamped driven Brownian motion, and the inertial terms are neglected. While accurate for a single, isolated particle at long times and/or sufficiently low driving frequencies, inertia may not always be negligible for EK-driven colloids, particularly for collective or confined motion. The overdamped models nonetheless capture several interesting effects,

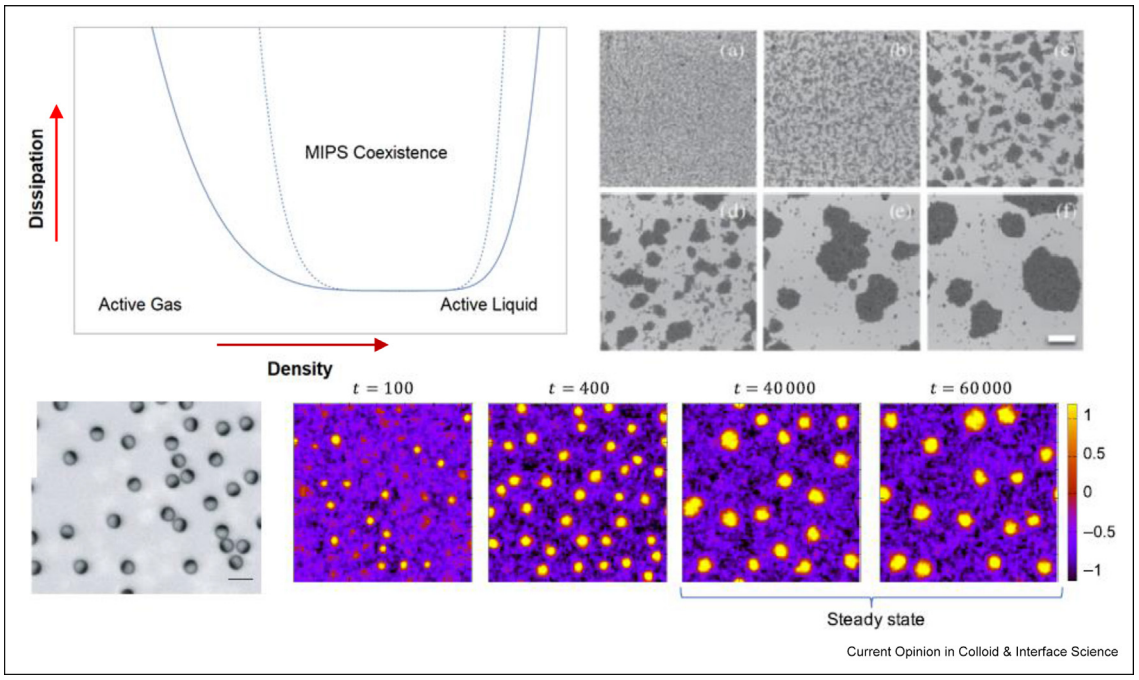
including phase transitions and anomalous diffusion [97—99]. However, for stochastic systems undergoing strong interactions in both active colloidal [101—105], and the general case [100], inertial corrections can play a critical role in long-time/ensemble dynamics. Coupling of the momenta exerted on EK active particles and the fluid motion via inertia may be important in dense/confined systems with strong collective response, especially because the system is in contact with a finite bath. Accounting for inertia in rotational diffusion, particularly in dense/confined collective motion, can lead to diffusion enhancement and a modified persistence length [101]. Coupling between the rotational and translational degrees of freedom leads to anomalous diffusion, directed transport, and localization phenomena [102]. Such considerations have led to a microscopic derivation of hydrodynamic equations for active suspensions [103], novel predicted behaviors such as acoustic filtering via active fluids [104], and modified active (effective equilibrium) temperature [105]. Inertia has also been shown to introduce hydrodynamic response and can affect the flocking transition [106]. Beyond inertia, other effects could contribute to modified diffusive transport for EK-active Brownian particles (ABPs), such as modified Brownian motion in the presence of gradients [107,108], memory friction [109,110], or even Casimir effects [111]. While some effects may be subtle, there is merit from a fundamental point of view of having a complex model system that permits experimental access to the detailed stochastic dynamics underlying emergent behaviors. A key question is at what level in terms of the collective behavior do such effects play an important role in determining dynamic structure? The EK active colloids provide a means to control and probe interparticle, particle—wall interactions in a system for which, at least in principle, simulations and experiments can have essentially the same resolution, i.e., at the individual particle-bath level of a thermodynamically significant ensemble.

These interactions lead to a rich variety of collective phenomena in the dynamics of active matter, such as motility-induced phase separation (MIPS). Here, active particles only undergoing repulsive interactions spontaneously separate into dense and dilute phases, driven by a velocity slow-down in the dense phase [95,97—99,105,112—114,114—123]. Phase separation has been predicted and observed in 2D terrestrial experiments. 2D molecular dynamics simulations have predicted transitions between fluid-like and crystalline phases, displaying anomalous diffusive behavior [97]. Velocity-aligned domains, vortex-like defects [113], bifurcations between different phase separation regimes [115], and multiple regimes of domain separation kinetics and domain size [117] have all been predicted. MIPS-induced collective trapping and smectic defects in the presence of an obstacle have been observed in active rods [116]. Experiments with EK JPs demonstrate velocity alignment resulting in the arrest of MIPS-domain formation in active particles with short-range repulsion [95]. Tuning of phase segregation can be achieved via electrostatic imbalance for active particles with surface charge [16], and hydrodynamic interactions have been demonstrated to counteract MIPS [118]. For inertial ABPs, the phase transitions are shown to be discontinuous, and cluster coarsening is suppressed by inertia, leading to a nucleation-like phase separation [119] and corresponding to the region between binodal and spinodal lines on the schematic in the top of Figure 3. Similarly, the arrest of cluster coarsening has been observed experimentally to occur due to orientational interactions [95], and continuum predictions have also attributed the arrest of cluster coarsening to the local breaking of TRS [96].

These phenomena occur in both 2D and 3D [98,114], but distinctions emerge regarding domain size and kinetics. Moreover, active particles in external fields, both gravitational [124] and the general case [125], can exhibit effective temperature. Comparison between equilibrium and non-equilibrium phase transitions [126] is further supported by the prediction of a 3D phase diagram demonstrating liquid, liquid—gas coexistence, solid—liquid coexistence regimes, and metastability in the form of a kinetic barrier to crystal formation [99]. 2D MIPS has been shown to belong to the Ising universality class for non-inertial ABPs [120] as well as active Ornstein-Uhlenbeck particles (AOUPs) [121]. A general condition for active systems to have an effective equilibrium regime, hence a well-defined effective temperature, is that they observe a time-reversal symmetry (TRS) at the particle-level in the sense that time-forward and time-reversed paths are statistically equally likely [127]. Open questions remain concerning how these results translate to 3D [98], how confining walls affect hydrodynamic interactions, and to what extent inertia can play a role, particularly in dynamic phase transitions. EK active colloids, with tunable interactions, provide an ideal probe with which to address such questions.

There are two main theoretical lenses through which to view the problem, microscopic particle dynamics, or continuum hydrodynamics. The most fundamental pragmatic approach is coarse-grained molecular dynamics, i.e., using multiparticle Langevin equations for modelling particle trajectories. While “all-atom” molecular dynamics is certainly possible, it is perhaps more appropriate to model active systems like motor proteins [128], the most basic point of view considers microscopic equations of motion for individual particle trajectories, typically taking the form of Langevin equations. Improved Brownian simulations consider multipolar interactions [129], lubrication between particle—particle and particle—wall [130]. Langevin

Figure 3



Top left figure shows a schematic phase diagram for an active Brownian particle (ABP) with binodal (solid) and spinodal (dashed) lines shown; additional interactions can lead to more complicated phase behavior (see main text.) Colloids have been observed to follow both nucleation and decomposition-based phase separation. Top right figure is adapted from Ref. [95], showing the time evolution of motility-induced phase separation (MIPS), domain coarsening, and arrest of phase separation in EK JPs attributed to orientational interactions. Scale bar is 100 μm. Bottom right shows the simulation of qualitatively similar arrest of domain separation in a continuum ABP model with local TRS violation [96]. Bottom left shows active gas [16]; the active liquid, or "swarm" state is shown in Figure 1 panel b [16].

simulations of oppositely charged particles investigate the frequency and field-dependent transition between disordered states, formation of lanes, and jamming in 3D [131]. In general, the models may be either of the ABP [117,119,120,132] or AOUP [121] type; the practical distinction being whether the thermal fluctuations are taken to directly affect the orientation of the particle, hence only the direction of propulsive velocity (ABPs), or its magnitude and direction via a fluctuating force (AOUPs.) In the detailed study of a specific system such as synthetic EK colloids, it is not clear a priori that one method is universally preferable. The AOUP approach may permit a more physically insightful means to capture changes in the particle velocity based on explicit interactions, e.g., with other particles, walls, and (dynamic) changes in the surrounding electrolyte properties. However, the essential collective behaviors can be qualitatively captured by the ABP approach; even purely phenomenological "agent" models, such as the Vicsek model or explicit Toner–Tu equations, [120] can describe phenomena such as MIPS in electrophoretic JP [95]. Of course, the detailed phase diagram will depend upon the model [117,119–121]. Continuum phase-field theories offer another practical alternative for studying collective behavior. Indeed, microscopic approaches like ABP have been used to study interfacial behavior in MIPS [132], demonstrating interfacial pressure and curvature phenomena, even leading to a Gibbs–Thomson relationship. Hydrodynamic equations may be derived from these microscopic equations of motion [95,103]. These approaches can be used to investigate the stability of active collective states [133] and have phenomenological appeal; in special cases, one can assume an effective equilibrium, with Cahn–Hilliard-like equations governing the particle density near the MIPS transition [134,135]. While it is possible to introduce a generalized chemical potential and generalized thermodynamics [136], in some cases, due to underlying anisotropy in density fluctuations introduced by the non-equilibrium driving, it is generally not possible to obtain this from the minimization of an effective free energy functional [126]. Nonetheless, a recent result by Markovich et al. [137] adopts a linear irreversible thermodynamics approach and rigorously includes the coupling between dissipated heat and the driving fields, serving as an explicit case of the more general coupling between work and heat in such systems [138]. Consistently and accurately representing the cornucopia of many-body and mean–field

interactions in a single, theoretical framework remains a challenge.

While machine learning (ML) has been fruitfully applied for pattern recognition [139,140] in active matter, recent progress in hidden-layer network operator learning methods to simulate nanoscale liquid–vapor phase change [141] offers promise for studying MIPS and related phenomena. The accessibility of these mesoscale active systems will facilitate the development of ML as a predictive, physics-based tool, and ML techniques may offer a third path forward, e.g., in parametrizing pairwise and three-body interactions for many-body models, or even obliviating the need for many-body simulations in some cases.

Outlook

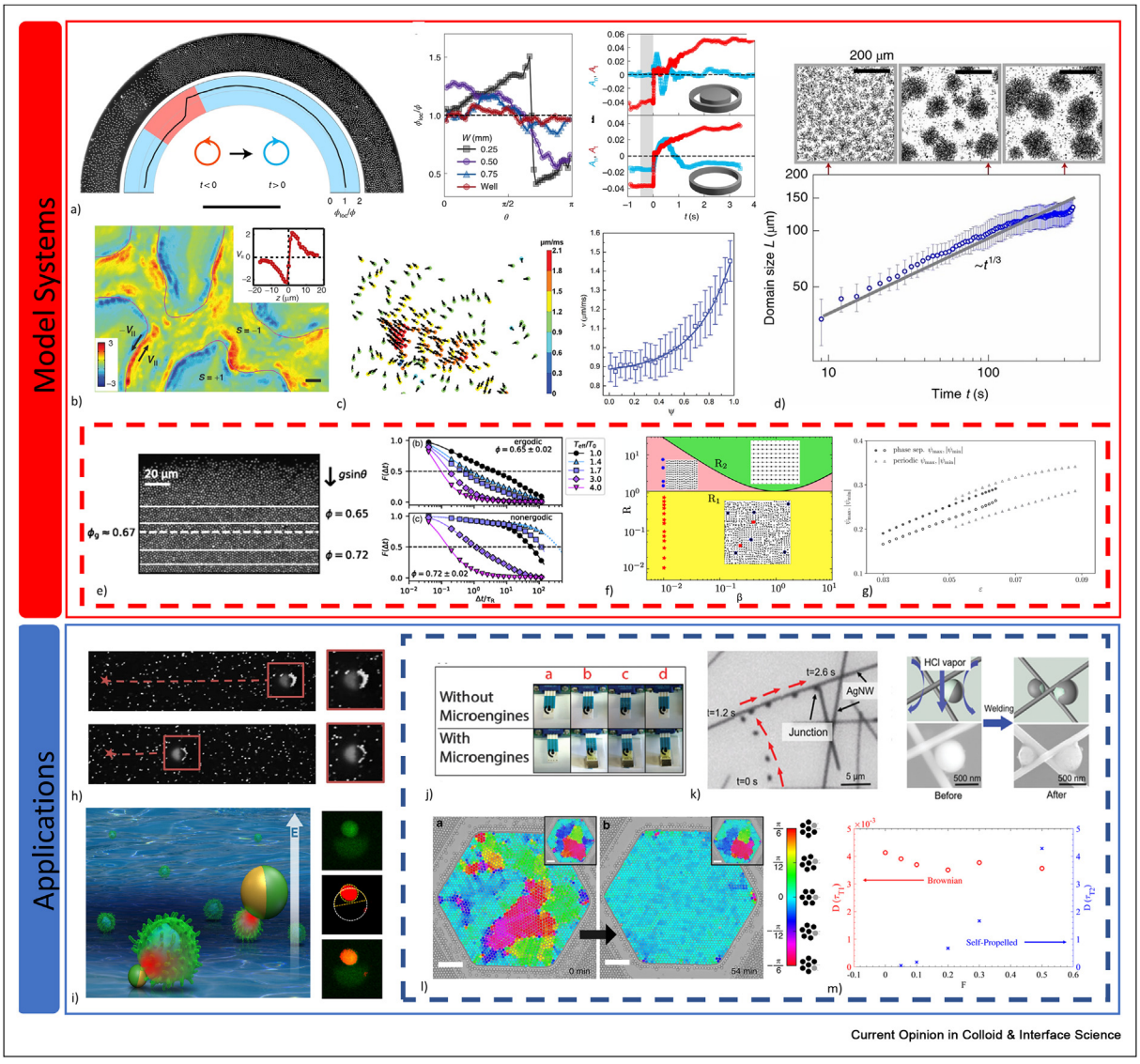
The applications of soft matter research may be divided into two categories—advancing fundamental phenomenological understanding and the commercial potential and realization of these advances (Figure 4). One of the most intellectually appealing features of synthetic active matter in general and EK active colloids specifically lies in the ability to serve as a highly tunable, and facile experimental platform for the investigation of emergent phenomena appearing in a wide array of complex systems [142–144]. Understanding of interactions between particles and obstacles on emergent phases in experimentally accessible non-equilibrium systems maps onto natural and biological systems across scales. As a customizable proxy for other mesoscale active matter, experimental observation and numerical predictions form a synergistic feedback loop that can determine the limitations of present theory and knowledge. For example, the observation of 3D sheets in Sakaï et al. [28] cannot be understood based on dipolar interactions informing the development of secondary models.

The key advantage of utilizing E-fields lies in the degree of external control over the colloid driving mechanism (Quincke instability, EHD flow, ICEP, and sDEP) which are also fine-tuned through the field properties—voltage, frequency, and waveform—allowing evaluation of the relative roles of short-range particle–particle interactions and long-range hydrodynamics underlying collective motion [145]. Thus, a single experimental setup can probe the transition between modes and phases, including active and passive [12], random walk to collective motion [16,26], and assembly. Toggled fields can modulate particle–particle interactions and resultant assemblies [15], as well as test memory effects and the related timescales [14,146]. Mesoscale systems provide accessible experimental platforms to test the foundations of statistical and thermal physics, e.g., the nature of non-equilibrium states, the approach to equilibrium, TRS, and the role of boundary conditions. Anomalous thermodynamic effects can occur for effective equilibria, e.g., the presence of a motility-induced temperature difference in coexisting phases of active inertial particles [112] and of an odd viscosity for actively torqued systems due to breaking time-reversal symmetry caused by inertial coupling between spin and velocity [147]. Nonetheless, the effective temperature concept has been examined for active mixtures, with good agreement between theory and experiment for quasi-2D systems [148]. Active-passive mixtures can undergo similar phase separation and assembly [122], with effective interactions between the passive particles that strongly depend on their interaction with the active particles [149]. The tunability of EK interactions permits tailored particle interactions, resulting in pattern formation [150] and progress further toward controlled assembly and field-modulated active structures [123]. This tunability also offers an advantage in the exploration of fundamental physical phenomena currently emerging in other active systems such as the ergodicity breaking and glass transition demonstrated by catalytic JPs [151], as well as behaviors predicted by theory/simulations like inertial contributions to flocking transitions [106] or a secondary bifurcation in active phase separation [115]. Competing effects could be further isolated and fundamental mechanisms better understood through creative experimental design which removes inherent secondary forces, such as the use of ISS to perform microgravity experiments [65].

We next focus on potential commercial applications of EK-driven systems and also draw on examples driven by other mechanisms that could be compatible with EK forcing. We note that the primary constraint is the need for external electrodes which limits biocompatibility in vivo and may also be the reason that EK applications are relatively undeveloped. In vitro, electrically powered active colloids can advance "lab-on-a-chip" capabilities. Non-uniform electric field gradients adjacent to the JP can be used to separate a target via DEP [71], transfect genes through localized electroporation [152], stretch DNA [153], or build a programmable reconfigurable micromachine [154] whose efficiency would be enhanced through optimizing the micro-robotic swarm [155,156]. Measurements of changes in individual active colloid mobility have widely been suggested as an analytical tool [157]; changes in collective mobility could be more robust than single-particle measurement and allow detection at a lower resolution for easier integration to portable instrumentation. From a bulk perspective, active particles can be added to heating/cooling fluids to increase mass [158] and heat transfer [159,160] or alter the rheology; in Ref. [161], polar and antiparallel alignment of JPs traveling under ICEP lead to an effective negative viscosity of active suspension and

emergence of active turbulence [162]. Another important application is the development of multifunctional, dynamically responsive materials where the assembly can be dictated by the external electric field. Catalytic active colloids can mend wires [163] and strengthen [164] and anneal [165] colloidal crystals, where in contrast to equilibrium systems, the order may increase with the number of "defects" [166]. The efficiency of these applications could increase with the intelligent actualization of collective motion. The integration of automated feedback loops [55] enhanced by ML [167] to read and control colloid mobility and collective motion can be used to automate the reconfiguration and analysis.

Figure 4



Potential use of EK active particles collective behavior: (**a–e**) Electrokinetically driven active colloids inform fundamental physical models (dotted line encloses important studies in non-E-field-driven systems whose results could be further explored using electric-driven colloids). (**a**) Quincke rollers demonstrate state memory by consistently reversing vortex direction when the field is toggled [146]. (**b**) Discontinuities at domain boundaries in electrokinetically driven systems suggest limitations to the effective temperature concept [148]. (**c**) Strength of applied field can probe correlation between pair alignment, particle velocity, and density [46]. (**d**) Characterization of phase separation in active fluids through monitoring cluster growth upon field activation [168]. (**e**) Ergodicity breaking and glass transition were observed with catalytic JPs [151]. (**f**) Inertial contributions to flocking transitions [106]. (**g**) A secondary bifurcation in active phase separation [115]. (**h-m**) Active colloid applications whose efficiency may be increased through collective motion: (dotted line encloses non-EK-driven but potentially compatible systems). (**h**)Accumulation, transport, and release of cargo by EK-driven JP [71]. (**i**) Localized gradients adjacent to active colloids enable electroporation for gene transfection [152]. (**j-m**) (**j**) Microengines accelerate chemical reactions and electrochemical detection [158], (k) active colloids locate flaws and self-repair as "mini welds" [163], (**l**) active colloids anneal crystal [165], (**m**) active colloids enhance heat transfer up to three orders of magnitude [160].

Electrokinetic actuation enables dynamic control of active systems to produce a rich array of phenomena that can be tuned through the electric field properties which dictate both the active driving mechanisms and modulates particle−particle interactions. This flexibility allows for the isolation of competing short-range interactions and long-range hydrodynamics and the probing of fundamental questions of non-equilibrium soft-matter and other physical systems. We note that the applications for synthetic EK colloids lag when compared to other active mechanisms leaving room for the significant potential for growth and development in which tunable collective motion could enhance efficiency and reconfigurability.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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* of special interest
** of outstanding interest

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