

Lifting active matter into three dimensions using buoyancy control and external alignment

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Sedimentation and gyrotaxis from metallic coatings in synthetic active matter confine particles to quasi-two-dimensional, wall-dominated states. To sustain a true three-dimensional distribution, both effects must be mitigated. Here, density matching in a glycerol/deionized water mixture (counteracting sedimentation) is combined with a uniform ac electric field that aligns and propels particles, minimizing gyrotaxis. Measurement of near-wall and bulk mobilities is now possible, revealing frequency-dependent particle-wall interactions consistent with theory. The method can be extended to other active systems to probe confinement effects and nonequilibrium behavior.

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Synthetic three-dimensional active systems provide a uniquely accessible experimental model with which to understand nonequilibrium statistical mechanics and complex systems [1,2], biological interactions [3,4], near and long-range wall effects [5–9], and the optimization of diffusion-driven applications (e.g., thermal transport [10]). However, the experimental realization of such systems remains a significant practical challenge due to sedimentation and gyrotaxis. Here, we address this challenge by density matching active colloids with the suspending solution while simultaneously restricting out-of-plane motion by controlling the alignment of the particle with an external electric field. We characterize the timescales associated with persistent motion in the bulk versus long-term wall accumulation. The setup is then used to measure the particle mobility at varying distances from the channel walls providing unique insight into the limitations and assumptions of existing theoretical models and simulations, as well as a clear pathway to the application of this approach to other active systems.

In accordance with the scallop theorem [11], a key requirement for the translation of microscale active particles is symmetry breaking. As in nature, this can be achieved through geometry (e.g., helical flagella [12]) but is more commonly attained by breaking material symmetry with a “Janus particle” (JP) [13] configuration, usually in the form of a polystyrene bead partially coated with a metal—the choice of which depends on the propulsive mechanism [14]. The high density of the metal causes particles to rotate the metallic hemisphere downward and sediment to the lower boundary [15–18]. If the colloidal activity is sufficient to counteract

gravitational torque and realign the metallodielectric interface with a component perpendicular to the wall, colloids exhibit stuck, oscillatory, or sliding states at the lower boundary [16,17,19–21] [Fig. 1(a)]. If the interface remains parallel to the wall, colloids may migrate to the upper boundary under negative gravitaxis [22], exhibiting the aforementioned states there [Fig. 1(b)]. In both cases, the experimental system is quasi-two-dimensional (quasi-2D).

Once at the wall, the particle mobility, equilibrium orientation and height are dictated by the complex interplay of short-range particle-wall interactions, including increased Stokes drag [19,23,24], electrostatic forces [19], phoretic interaction [25], electrohydrodynamic flow [26,27], and amplified electrical or chemical [28,29] gradients.

The predominance of wall effects and lack of means to directly measure bulk mobility experimentally create a conundrum for the validation of theoretical models of the propulsive mechanism, generally derived for a single particle translating in the bulk (e.g., Refs. [30–32]). Despite laying the foundational theory that can be extended to account for wall effects in accordance with experimental observation (e.g., Refs. [5,16,19,21]), these models cannot be tested independently within the quasi-2D paradigm. Moreover, other effects, such as the attraction of swimmers to out-of-plane boundaries [8,33,34], cannot be observed at all in synthetic systems. Although such effects have been measured experimentally with biological swimmers, these systems have limited control when compared to synthetic actuation, restricting the parameter space available for exploration and validation of molecular dynamics simulations. The use of external trapping (e.g., magnetic [35], optical [34,36]) mechanisms to lift particles from the wall suffers from the difficulty that the trapping mechanism may interfere with the interaction being tested. Mitigating gravitational effects by conducting experiments in microgravity [37,38] is a potential solution but is both technically challenging [39] and cost prohibitive, as the length of a typical experiment requires long-term microgravity such as on the International Space Station.

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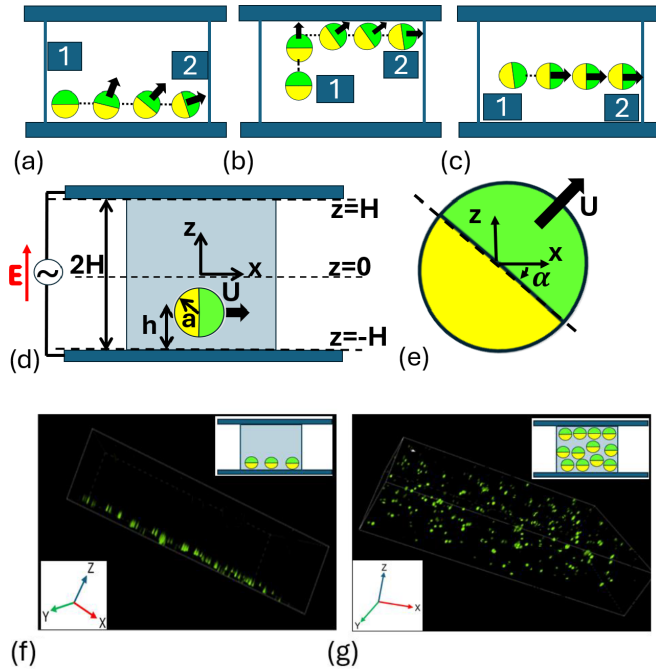


FIG. 1. (a)–(c) Timelapse of active colloids in differing configurations. “1” indicates initial equilibrium position (no forcing) and “2” represents final equilibrium position. (a) Sedimentation dominated. (b) Density matched but no external alignment. (c) Density matched with external alignment. (d) Schematic of electric-field-driven experimental setup. (e) Schematic of Janus particle with equilibrium orientation angle α determined by balance of gravitational torque and external alignment. z stack of JP distribution in (f) pure DI water (sedimentation dominated) and (g) 70% w/w glycerol/DI (density matched).

A more straightforward approach to counteract sedimentation is to increase the density of the suspending solution so that the JPs are neutrally buoyant. However, without also counteracting the gravitational torque, bulk mobility measurements are not possible; particles that escape the lower substrate will rapidly translate upward and be trapped at the upper boundary [16] [Fig. 1(b)]. Thus, to measure mobility in the bulk, one must both match the density and align the interface perpendicular to the wall [Fig. 1(c)].

In the present work, this is achieved by density matching gold-polystyrene Janus particles in a 0.70% w/w

glycerol/water solution (see Sec. S1 of the Supplemental Material [40]) and subjecting them to a uniform ac electric field, which is applied perpendicular to the upper and lower boundaries [Fig. 1(d)]. Previously, this setup was used to map the phase space of a concentrated suspension of density-matched metallodielectric JPs subject to ac electric fields [41]. Here, we use a relatively low volume fraction of Janus particles ($\sim 0.78\%$) to limit particle-particle interactions (see Sec. S2 of the Supplemental Material [40]). Under the applied field, the metallic hemisphere of the Janus particle becomes polarized, inducing migration of ions within the fluid to preserve net neutrality and creating an induced electric double layer (EDL). The applied field acts on this EDL, producing hydrodynamic flow, which then propels the Janus particle with the dielectric (fluorescent) hemisphere facing forward under induced charge electrophoresis (ICEP) [30,42,43]. The JP can then be considered a pusher-type swimmer [5], where we note that for the classical squirmer (swimmer) problem (e.g., Refs. [44,45]), the tangential or normal surface velocities of the colloid are prescribed; in the present case, surface velocities are defined by the Helmholtz-Smoluchowski slip velocity.

Following Ref. [30] (see also Appendix), in the absence of wall effects, the dimensionless theoretical ICEP force (normalized by $\varepsilon\varepsilon_0a^2E_0^2$, where ε is the relative permittivity of the fluid, ε_0 the permittivity of a vacuum, E_0 the amplitude of the applied electric field, and a the particle radius) for a single particle translating in the bulk is given by

$$F_{\text{ICEP}}^{(1)} = \frac{27\pi}{16} \left[\frac{\Omega_H^2}{1 + \Omega_H^2} \right] \left[\frac{1}{4 + \Omega^2} \right], \quad (1)$$

where the factor of $1/(4 + \Omega^2)$ (where $\Omega = \omega a \lambda_0 / D$, ω is the frequency of the applied field, λ_0 is the Debye length, and D is the diffusivity of a z : z symmetric solute) accounts for the high-frequency screening due to relaxation of the EDL at the metallic hemisphere, while $\Omega_H^2/(1 + \Omega_H^2)$ (where $\Omega_H = \omega H \lambda_0 / D$, $2H$ is the chamber height) accounts for the low-frequency decay due to screening of the electrodes.

The modification of the ICEP force due to wall proximity can be obtained following the asymptotic approach of Refs. [24,46], where we have extended our solution presented in Ref. [43] to the fifth order in a/h (see Appendix). Additionally, a higher-order correction accounting for the conducting nature of the wall/electrode is included following Ref. [26] (see Appendix) such that the ratio of the velocity near the wall relative to the bulk is a function of frequency:

$$F_{\text{ICEP}}^{(1)} = \frac{27\pi}{16} \left(\frac{\Omega_H^2}{1 + \Omega_H^2} \right) \times \left[\frac{\left[1 - \frac{9}{16} \left(\frac{a}{h} \right) + \frac{1}{8} \left(\frac{a}{h} \right)^3 - \frac{45}{256} \left(\frac{a}{h} \right)^4 - \frac{1}{16} \left(\frac{a}{h} \right)^5 \right]^{-1}}{4 - \frac{1}{2} \left(\frac{a}{h} \right)^3 + \frac{1}{16} \left(\frac{a}{h} \right)^5 + \Omega^2 \left(1 - \frac{1}{4} \left(\frac{a}{h} \right)^3 - \frac{1}{8} \left(\frac{a}{h} \right)^5 \right)} \right]. \quad (2)$$

We note that Eq. (2) reduces to Eq. (1) in the remote limit, namely, when $a/h \rightarrow 0$. The resultant expression indicates that the ICEP force increases with proximity to the channel wall and as a function of the frequency of the applied field. The corresponding velocity U for a freely suspended particle translating in the x direction is derived by equating $F_{\text{ICEP}}^{(1)}$ [Eq. (2)] with the Stokes drag defined according to Ref. [23], such that

$$U = \frac{F_{\text{ICEP}}^{(1)}}{6\pi f},$$

$$f = 1 - \frac{9}{16} \left(\frac{a}{h} \right) + \frac{1}{8} \left(\frac{a}{h} \right)^3 - \frac{45}{256} \left(\frac{a}{h} \right)^4 - \frac{1}{16} \left(\frac{a}{h} \right)^5. \quad (3)$$

The particles are fabricated from polystyrene spheres (radius $a = 2.5 \mu\text{m}$, Fluro-Max, Thermo Scientific) on which a thin 15 nm Ti/15 nm Au hemispherical coating is deposited by E-beam [46] (see Sec. S2.1 of the Supplemental Material [40]). The particles are density matched at ambient lab temperature of 25 °C in a 0.70% w/w glycerol (Sigma-Aldrich, $\rho_g = 1.26 \text{ g/cm}^3$ [47], 92.09 g/mol, $\text{C}_3\text{H}_8\text{O}_3$) to deionized water ($\rho_w = 0.997 \text{ g/cm}^3$) mixture with density $\bar{\rho}_f = 1.17 \text{ g/cm}^3$ [47] (see Sec. S2.2 of the Supplemental Material [40]). In Figs. 1(f) and 1(g), a z stack imaged with a spinning disc confocal microscope (Nikon CSU-X1 mounted on Nikon TI2-E) and motorized stage (Ti2-S-SE-E) with a 20 $\times$ objective illustrates the difference in distribution of particles suspended in DI water [Fig. 1(f)], which is sedimentation dominated, and the 0.70% w/w glycerol/DI mixture [Fig. 1(g)]. The density-matched solution is placed inside single ($2H = 120 \mu\text{m}$) or stacked ($2H = 240 \mu\text{m}$) silicon reservoirs (Secure Seal Space) that are sandwiched between an indium tin oxide (ITO)-coated glass slide (Delta Technologies) and ITO-coated cover slip (SPI) [Fig. 1(d)]. To minimize particle sticking to the ITO [43,48,49], a 20 nm SiO_2 layer is thermally deposited by E-beam on the ITO-coated side of both the glass slide and cover slip (see Sec. S2.3 of the Supplemental Material [40]). The ITO-coated substrates serve as both channel walls and electrodes, which are connected to an ac function generator (Keysight, 33210A) for application of voltages between 3.3 and 16 V_{pp} and frequencies ranging from 0.2 to 5 kHz. Image capture is facilitated by a high-resolution (ORCA-Fusion C14440-20UP) camera at a rate of 4 fps. Mobility analysis was conducted using ImageJ with the speckle tracker plugin [50] and postprocessed with in-house Python code [51] (see Sec. S2.4 of the Supplemental Material [40]).

Given the strongly temperature-dependent properties of glycerol [47,52], a key concern with the application of the electric field is Joule heating causing density and viscosity gradients as well as electrothermal flow [53]. Here, we aim to minimize Joule heating and electrothermal effects by using deionized water with the glycerol, which yields a solution conductivity less than $1 \mu\text{S/cm}$ [54]. To evaluate the presence of temperature gradients, the temperature of the device under a continuous applied field is measured externally with an infrared camera and internally with temperature-sensitive fluorescent dyes ([54,55]) over a period of 10 min with little variation observed (see Secs. S3.1 and S3.2 of the Supplemental Material [40] and Ref. [56]); similarly, there is no marked increase in JP velocity over time, which would correlate with reduced viscosity (see Sec. S3.3 of the Supplemental Material [40]). Microparticle image velocimetry is used to check for buoyant or electrothermal background flow (see Sec. S3.4 of the Supplemental Material [40] and Ref. [57]). No significant difference is observed between the control and the chamber subject to an applied field. Combined, these observations suggest that any Joule heating effects are secondary to the dominant mechanisms described below. Finally, we validate that the driving mechanism is ICEP by verifying the velocity scales quadratically with the applied field (versus electrothermal flow, which scales to the fourth power [53,54]) and the frequency dispersion is well modeled by Eq. (3) when the

Stern layer capacitance is accounted for (see Sec. S4 of the Supplemental Material [40] and Refs. [58–61]).

In order to measure the bulk mobility, it is critical that the velocity is predominantly in-plane and particles remain in the bulk for the duration of the experiment. A 60 $\times$ /1.40 oil immersion objective (Nikon) is used to reduce the focal plane facilitating extraction of the out-of-plane velocity component (in the z direction). The objective is initially focused in bulk ($z = 0$) and individual particles are tracked. When the particle leaves the focal plane, the microscope is refocused with a motorized stage (Ti2-S-SE-E) to extract the out-of-plane displacement. A sample timelapse is presented in Fig. 3(a). The in- and out-of-plane velocities averaged for three particles at a range of voltages between 6 and 18 V are provided in Fig. 3(b). Approximately 98% of particles translated upward as expected for the gravitational-dominated orientation [Fig. 1(e)]. The ratio of these velocities is found to be $0.088 < U_z/U_x < 0.18$, corresponding to a calculated range of α° , $80 < \alpha^\circ < 88$, where $\alpha^\circ = 90 - \tan^{-1}(U_z/U_x)$ [Fig. 3(b), secondary axis]. Although these values are larger than those reported in Ref. [43], this is consistent with the reduced thickness of the metallic coating and corresponding reduction in the gravitational torque.

Characterization of the out-of-plane velocity establishes the maximum timescale for the operation of a single chamber for bulk mobility measurements based on the time taken for particles to travel to the boundary. Thus, to define the bulk JP residence time for a chamber of $H = 120 \mu\text{m}$, we consider the time (τ_0) necessary for a particle to travel from $z = -25$ to $z = 25$, i.e., $\tau_0(\text{min}) = (50/U_z)/60$. Accordingly, in Fig. 3(c), we measure the distribution of the particles throughout the chamber as a function of time. The fraction of particles $p(z)$ in an imaged x - y layer (index i) at a given time t is calculated as $p(z) = N_{z=i} / \sum_{i=1}^n N_i$, where N_i is the number of particles in the i th layer and n is the number of layers. The chamber is divided into five equal layers, where $i = 1$ is the bottom of the chamber ($z = -H$) and $i = 5$ is the top layer ($z = H$). With an applied field of 8 V_{pp} and 1 kHz, where particles have a mobility of $U_x = 0.53 \pm 0.05 \mu\text{m/s}$ and $U_z = 0.05 \pm 0.03 \mu\text{m/s}$ [Fig. 3(b)], the time for a particle moving with average velocity to leave the bulk will be $\tau_0 \sim (50/0.05)/60 = 17 \text{ min}$ [dashed line in Fig. 2(c)], while particle travelling at plus/minus one standard deviation will leave between 10 and 42 min, respectively. Indeed, we observe that at $t = 45 \text{ min}$, less than 3% of particles remain in the bulk [Fig. 2(c)].

Interestingly, these measurements also show that although the measured $U_z > 0$ for 98% of particles, a significant number of particles accumulate at the lower boundary. Most of these particles appear to originate from the closest layer as can be observed in Fig. 2(c), where at short times, $p(i_2)$ decreases while $p(i_3)$ and $p(i_4)$ increase. This short-time accumulation in the bulk and the top bias in the saturation values are likely due to gyrotaxis resulting in a velocity vector with an upward component [Fig. 1(d)]. However, the accumulation at the lower boundary above 40% indicates the presence of long-range effects that draw particles farther from the wall as well as short-range forces preventing the particles from escaping. Previously, such phenomena have been reported in biological systems [4], examined theoretically and numerically in the

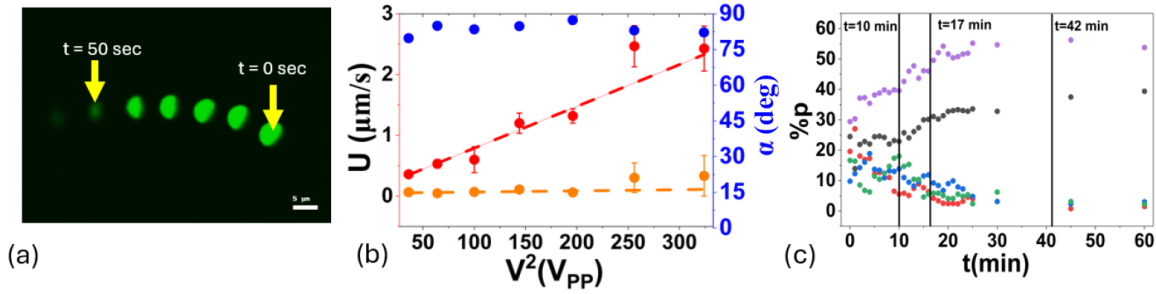


FIG. 2. (a) Timelapse image of Janus particle ($8 V_{pp}$, 1 kHz) located in the bulk ($i = 3$) of a chamber $2H = 120 \mu\text{m}$. (b) Averaged velocities in the x (red) and z (orange) directions. The dotted lines (red and orange) represent quadratic scaling expected in ICEP. Blue circles show extracted value of α (secondary axis). (c) Depletion of the bulk and accumulation near the walls over long times; colors correspond to varying heights: black ($i = 1$), red ($i = 2$), blue ($i = 3$), green ($i = 4$), and purple ($i = 5$). The dashed line represents the time taken for a particle with average speed to leave the bulk; the solid and dotted lines indicate the times taken for a particle with speeds of plus/minus one standard deviation to leave the bulk.

context of active Brownian particles (e.g., Refs. [5,16,19,21]), but experiments in synthetic systems were prevented by their quasi-2D nature. Identification of these long-range mechanisms, validation of these simulations, and exploration of the broader parameter space made possible by controllability of active colloids is another exciting direction for application of the present experimental setup, left here for future work.

While the time for the bulk to deplete sets the upper bound limit for these experiments, the lower bound is dictated by minimum time necessary to measure the activity of the particles, which may be quantified by the mean square displacement (MSD). At short times, active particles exhibit ballistic motion, while over long times, the motion is diffusive. At low particle concentrations, the transition for observation of ballistic (short timescale) to diffusive (long timescale) motion is governed by the timescale for rotational diffusion of a single Brownian particle in a viscous bath: $\tau_r = 8\pi\eta a^3/(2k_B T)$, where k_B is the Boltzmann constant, T the temperature, and η the viscosity. For $a = 25 \mu\text{m}$, $T = 298 \text{ K}$, $\eta = 17 \text{ cP}$ (see Sec. S5 of the Supplemental Material [40]), $\tau_r \sim 835 \text{ s}$. Thus, only ballistic motion may be observed in the bulk and one may equivalently extract the velocity using the straight line speed as $U = \Delta x/\Delta t$, where Δx refers to the distance between the initial and final positions of the particle and Δt is

the run-time of a single mobility measurement. We validate that the obtained velocities correspond with values obtained from the asymptotic expression for the MSD, valid in the limit $\tau \ll \tau_r$: $\text{MSD} = 4D\tau + U_x^2 \tau^2$ (see Sec. S2.4 of the Supplemental Material [40] and Ref. [62]), where $D = k_B T/6\pi\eta a$ is diffusivity coefficient and τ = number of frames/frame rate. We note that based on the ratio of U_z/U_x obtained above, the error in estimating the velocity based solely on the in-plane straight line speed is within 10%, since for a maximum value of $U_z = 0.18U_x$, $U_t = \sqrt{U_x^2 + 0.18U_x^2} = 1.1U_x$.

In Fig. 3, we compare the velocity of the Janus particles for varying values of h in chambers of $2H = 120 \mu\text{m}$ [Fig. 3(a)] and $2H = 240 \mu\text{m}$ [(Fig. 3(b)] at a constant electric field $E_0 = 24 \text{ kV/m}$. The velocity at each plane ($U|_{z=h}$) is normalized by the mobility at the channel centerline ($U|_{z=0}$) [Fig. 3(a)].

The standard deviation is calculated using the first-order nonlinear error propagation (see Sec. S6 of the Supplemental Material [40]). The maxima in Fig. 3(a)—compared to the expected plateau observed in Fig. 3(b)—raise the question as to whether confinement restricts the bulk mobility even for electrode spacing an order of magnitude larger than the radius $H = 12a$. Replotting the normalized velocities as a function of the height scaled with the radius, h/a , it is observed that a constant velocity is approached at $h/a > 20$, suggesting that the peak in Fig. 3(a) does reflect the maximum bulk

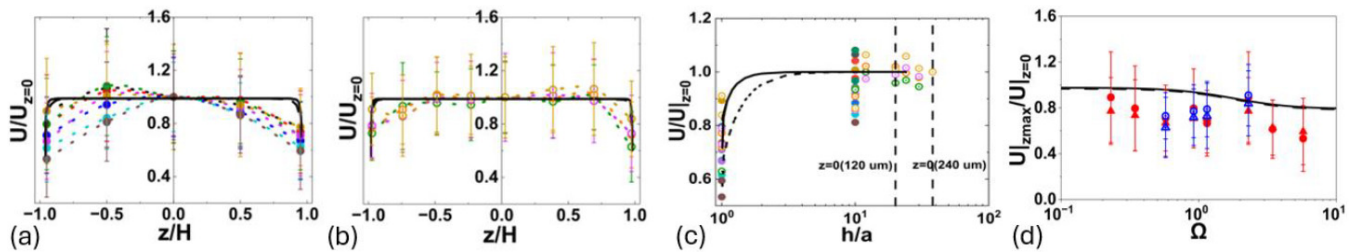


FIG. 3. Variation of mobility normalized by the midplane velocity ($U_{z=0}$) as a function of chamber height: (a) $2H = 120 \mu\text{m}$ and (b) $2H = 240 \mu\text{m}$. Variation of the ratio between near-wall and bulk mobilities as a function of (c) h/a and (d) Ω . Solid circles correspond to $2H = 120 \mu\text{m}$; open circles correspond to $2H = 240 \mu\text{m}$. (d) Variation of ratio between near-wall and bulk mobilities as a function of Ω . Red circles and triangles correspond to $2H = 120 \mu\text{m}$ for the bottom and top walls, respectively, and open blue circle and triangle correspond to $2H = 240 \mu\text{m}$ for the bottom and top walls, respectively. Colors denote frequencies according to black (0.2 kHz), red (0.3 kHz), blue (0.4 kHz), green (0.5 kHz), orange (0.8 kHz), pink (1 kHz), mustard (2 kHz), cyan (3 kHz), and brown (5 kHz). Lines correspond to Eq. (3) for $\Omega = 0.1$ (solid) and $\Omega = 10$ (dashed).

mobility [Fig. 3(c)]. The large spread of the velocities at $h/a = 10$ suggests that the velocity of particles captured in the focal plane is varying significantly in this domain, in accordance with the theoretical model, wherein the velocity decays rapidly near the wall.

Near the wall, particles are generally observed to exist in a monolayer due to short-range wall attraction [5]; here, the spread of the velocities at $h/a \rightarrow 1$ suggests the presence of frequency-dependent particle wall interaction, which may also modulate the equilibrium height [63]. Accordingly, in Fig. 3(d) we plot the frequency dependence of the ratio $U|_{z_{\max}}/U|_{z=0}$ [where $z_{\max} = \pm(H-h)$, $i = 1, i = 5$] and compare to the theoretical model [Eq. (3)] in Fig. 3(d). The model qualitatively captures the overall decrease of $U|_{z_{\max}}/U|_{z=0}$ with frequency but does not predict the increase at moderate frequencies. It is most likely that this is due to the limitations of the theoretical model (which is essentially a modified far-field approach using the method of successive images [24]) that cannot capture effects in the near-contact limit, including expected frequency-dependent variations in equilibrium height [63], or the frequency shift between near-wall and bulk mobilities previously predicted by numerical simulations [46]. That the near-contact limit is likely reached is supported by Fig. S8(c) in Sec. S4 of the Supplemental Material [40], where it is observed that quantitatively the theory evaluated at $h/a = 1.05$ overpredicts the measured near-wall velocity. Extension of the present model to the near-contact limit is left for future work, while it is noted that at such distances one would need to account for EDL overlap and local field distortions. The current experimental setup may uniquely provide a pathway for validation of such models.

In this vein, we reemphasize that the measurements described in this Letter are uniquely facilitated by combining both density matching and external alignment to counteract gravitational torque. Electrokinetic actuation offers two important advantages over other common active propulsion mechanisms (e.g., self-diffusiophoresis, self-electrophoresis) in this context: (1) The velocity scales linearly with the particle radius [42] such that larger, more buoyant particles may be used (see Sec. S1 of the Supplemental Material [40]) and (2) the electric field aligns the induced dipole within the polarizable surface so that the particle velocity is predominantly in the x - y plane [Fig. 1(c)]. Extension of the approach to other active systems that do not inherently constrain the interface orientation (e.g., catalytic JPs) will require the addition of an external aligning force (e.g., magnetic). Such

experiments could provide deeper insight into short-range particle-wall interactions and the role of local chemical field gradients in modulating mobility. One can also envision a system where these properties are exploited to actively regulate localized particle volume fractions and orchestrate collective motion within active colloidal assemblies. Understanding and harnessing these capabilities are essential for enhancing the stability and performance of active colloidal dispersions or emulsions, thereby advancing practical applications in materials science and soft matter technologies.

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Data availability. The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

Appendix. The theoretical model is based upon the weak-field formulation [19] wherein the governing electrostatic and hydrodynamic equations are linearized and decoupled. This technique is applicable when the electric potential ϕ is consider small with respect to thermal scale, $\phi_t = k_B T / ze$, where k_B is the Boltzmann constant, T the temperature, z the valence, and e the elementary charge. Previously, this model has been used to effectively model the ICEP velocity of metallodielectric Janus particles. Here, following Ristenpart *et al.* [26], we extend our previous work in Ref. [46] to account for the constant potential prevailing at the electrode/channel wall, which yields a higher-order, frequency-dependent correction to the ICEP mobility in accordance with experimental observations.

Consider two electrodes (wall) where the potential on the upper wall ($z = H$) is $\phi(H) = 0$ and that on the lower wall ($z = H$) is $\phi(H) = \Delta\phi_0$.

Following Eq. (26) of Ref. [26], the ambient electric potential in the absence of the particle can be expressed as

$$\phi(z) = \frac{\Delta\phi_0}{2} \left[1 - \frac{\sinh(\gamma z/H) \operatorname{cosec}(\gamma) - i(z/H)\gamma v \coth(\gamma)}{1 - i\gamma v^2 \coth(\gamma)} \right], \quad (\text{A1})$$

where $v^2 = \omega\lambda_0^2/D$ and $\gamma^2 = (H/\lambda_0)^2 - i(\omega H^2/D)$.

For a thin EDL, $\gamma \sim H/\lambda_0 \gg 1$, and thus near the upper wall, $z \sim H$ such that

$$\phi(z) = \frac{\Delta\phi_0}{2} \left[1 - \frac{1 - i(z/H)\gamma v^2}{1 - i\gamma v^2} \right]. \quad (\text{A2})$$

Next, consider a Janus particle lying at a distance h from the upper wall. A Cartesian coordinate system attached to the JP (x_1, y_1, z_1) admits $x_1 = x$, $y_1 = y$, $z_1 = -H + h + z$.

Thus, in the absence of the JP, Eq. (A2) can be written as

$$\phi(z_1) \simeq \frac{\Delta\phi_0}{2} \frac{\Omega_H}{\Omega_H + i} \left[\frac{h}{H} - \frac{z_1}{H} \right], \quad (\text{A3})$$

where $\Omega_H = \gamma v^2 \sim \omega \lambda_0 H/D$.

Using the approximation (including its first wall image),

$$\phi(z_1) \simeq \frac{\Delta\phi_0}{2} \frac{\Omega_H}{\Omega_H + i} \times \left[\frac{h}{H} - \frac{1}{H} \left(z_1 + \frac{a^3 \mathbb{D} z_1}{R_1^3} + \frac{a^3 \mathbb{D} z_2}{R_2^3} \right) \right] \quad (\text{A4})$$

such that $\mathbb{D}$ denotes the corresponding dipole strength and $x_2 = x_1$, $y_2 = y_1$, $z_2 = z_1 - 2h$, $R_1^2 = x_1^2 + y_1^2 + z_1^2$, $R_2^2 = x_1^2 + y_1^2 + (z_1 - 2h)^2 = R_1^2 - 4z_1 h + 4h^2$. Note that $\phi(z_1 = h) = \phi(z = H) = 0$. Next, using the following asymptotic expansion in $a/h \leq 1$: $1/R_2^3 \sim 1/(2h)^3 [1 - \frac{3}{2}(a/2h)^2 + \dots]$, one gets

$$\phi(z_1) \simeq \frac{\Delta\phi_0}{2} \frac{\Omega_H}{\Omega_H + i} \times \left[\frac{h}{H} + \frac{a^3 \mathbb{D}}{H(2h)^2} \left[1 - \frac{3}{2}(a/2h)^3 + \dots \right] - \frac{1}{H} \left(z_1 + \frac{a^3 \mathbb{D} z_1}{R_1^3} + \frac{a^3 \mathbb{D} z_1}{(2h)^3} \left(1 - \frac{3}{2}(a/2h)^3 + \dots \right) \right) \right]. \quad (\text{A5})$$

At the surface of the Janus particle, we impose the common resistance-capacitance boundary condition [46] for ac fields:

$$a \frac{\partial \phi}{\partial R_1} \Big|_{R_1=a} = -i\Omega(\phi - \phi_0), \quad \Omega = \frac{\omega \lambda_0 a}{D}, \quad (\text{A6})$$

which when expressed in JP-attached spherical coordinates (R_1, θ, φ) renders

$$\phi - \phi_0 \simeq -\frac{\Delta\phi_0}{2} \frac{\Omega_H}{\Omega_H + i} \left[R_1 + \frac{a^3 \mathbb{D}}{R_1^2} + \frac{a^3 \mathbb{D} R_1}{(2h)^3} \left(1 - \frac{3}{2}(a/2h)^2 + \dots \right) \right] \sin \theta \cos \varphi, \quad (\text{A7})$$

which yields

$$\mathbb{D} \simeq \left(\frac{2 - i\Omega}{1 + i\Omega} - \left(\frac{a}{2h} \right)^3 \left[1 - \frac{3}{2} \left(\frac{a}{2h} \right)^2 \right] \right)^{-1}. \quad (\text{A8})$$

Note that for $a/h \rightarrow \infty$ and $\Omega = 0$, one gets $\mathbb{D} = 1/2$ in accordance with Ref. [46].

To evaluate the ICEP force exerted on a polarizable colloid (Q and χ represent the induced-charge and ambient potential), we use the Teubner formulation [64], which for a thin EDL renders [46]

$$F_1^{\text{ICEP}} = \frac{3T}{16} \left(\frac{\lambda}{\lambda_0^2} \right) \int_S (\delta_{ij} - n_i n_j) Q^* \frac{\partial \chi}{\partial x_j} dS. \quad (\text{A9})$$

Here, S denotes the surface of the metallic hemisphere, $\mathbf{n}$ is its normal vector, and $*$ represents the complex conjugate. The geometrical parameter T in Eq. (A9) accounts for the proximity effect of the nearby no-slip wall, which following Davis and Crowdy [24] can be estimated to fifth order in wall spacing as

$$T \sim \left[1 - \frac{9}{16} \left(\frac{a}{h} \right) + \frac{1}{4} \left(\frac{a}{h} \right)^3 - \frac{63}{256} \left(\frac{a}{h} \right)^4 - \frac{1}{16} \left(\frac{a}{h} \right)^5 \right]^{-1}. \quad (\text{A10})$$

Taking advantage of the axial symmetry around x_1 of the potential $\chi(r, \theta)$, one gets

$$\frac{\partial \chi}{\partial x_1} - n_1 \frac{\partial \chi}{\partial r} = \frac{\partial \chi}{\partial \theta} \frac{\partial \theta}{\partial x_1} \quad (\text{A11})$$

and

$$\frac{\partial \theta}{\partial x_1} = -\frac{\sin \theta}{a}. \quad (\text{A12})$$

Finally, noting that at the metallic hemisphere S , $\phi = 0$ and thus $0 = -(\lambda/\lambda_0)^2 Q + \chi$ [64], we obtain

$$F_1^{\text{ICEP}} = \frac{3}{16a} T \int_S \chi^* \frac{\partial \chi}{\partial r} \sin \theta dS. \quad (\text{A13})$$

Recalling next that $\chi(r, \theta) = \phi(z_1) - \phi_0$, one readily finds from Eq. (A7) that

$$\begin{aligned} \frac{1}{2} \chi(a, \theta) &\simeq -\frac{\Delta\phi_0}{2H} \left(\frac{\Omega_H}{\Omega_H + i} \right) a \\ &\times \left[1 + \mathbb{D} \left(1 + \left(\frac{a}{2h} \right)^3 + \dots \right) \sin \theta \cos \varphi \right] \end{aligned} \quad (\text{A14})$$

and

$$\begin{aligned} \frac{1}{2} \frac{\partial \chi}{\partial \theta}(a, \theta) &\simeq -\frac{\Delta\phi_0}{2H} \frac{\Omega_H}{\Omega_H + i} a \\ &\times \left[1 + \mathbb{D} \left(1 + \left(\frac{a}{2h} \right)^3 + \dots \right) \cos \theta \cos \varphi \right]. \end{aligned} \quad (\text{A15})$$

Substituting the above in Eq. (A13) yields

$$\begin{aligned} F_1^{\text{ICEP}} &\simeq \left(\frac{\Delta\phi_0}{2H} \right)^2 \frac{\Omega_H^2}{\Omega_H^2 + 1} T \left| 1 + \mathbb{D} \left(1 + \left(\frac{a}{2h} \right)^3 + \dots \right) \right|^2 \\ &\times \frac{3}{4} a^3 \int_0^{2\pi} \cos^2 \varphi d\varphi \int_0^{\pi/2} \sin^3 \theta \cos \theta d\theta \end{aligned} \quad (\text{A16})$$

or

$$\begin{aligned} F_1^{\text{ICEP}} &\simeq \frac{3\pi}{16} a^3 \left(\frac{\Delta\phi_0}{2H} \right)^2 \frac{\Omega_H^2}{\Omega_H^2 + 1} \\ &\times T \left| 1 + \mathbb{D} \left(1 + \frac{1}{8} \left(\frac{a}{h} \right)^3 + \dots \right) \right|^2. \end{aligned} \quad (\text{A17})$$

Finally, substitution of Eqs. (A7) and (A9) into Eq. (A16) and gathering terms to $(a/h)^5$, we finally obtain

$$\begin{aligned} F_1^{\text{ICEP}} &= \frac{27\pi}{16} a^3 \left(\frac{\Delta\phi_0}{2H} \right)^2 \frac{\Omega_H^2}{\Omega_H^2 + 1} \\ &\times \frac{T}{4 - \frac{1}{2} \left(\frac{a}{h} \right)^3 + \frac{1}{16} \left(\frac{a}{h} \right)^5 + \Omega^2 \left(1 + \frac{1}{4} \left(\frac{a}{h} \right)^3 - \frac{1}{8} \left(\frac{a}{h} \right)^5 \right)}. \end{aligned} \quad (\text{A18})$$

Note that for the unbounded case, $(a/h) \rightarrow 0$ and $T \rightarrow 1$, F_1^{ICEP} reduces to

$$F_1^{\text{ICEP}} = \frac{27\pi}{16} a^3 \left(\frac{\Delta\phi_0}{2H} \right)^2 \frac{\Omega_H^2}{\Omega_H^2 + 1} \times \frac{1}{4 + \Omega^2}, \quad (\text{A18})$$

which is identical to Eq. (15) in Ref. [46], where $\Delta\phi_0/2H \sim E_0$ and $a = 1$.

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