Supplementary Information (SI) for Soft Matter. This journal is © The Royal Society of Chemistry 2025

# Electronic Supplementary Information: Tunable Assembly of Confined Janus Microswimmers in Sub-kHz Electric Fields under Gravity

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#### S1. Vertical density profiles

At f=800 Hz, our system shows features analogous to the active gas-like state observed for suspension of Pt-capped catalytically active Janus particles under a gravity field<sup>1–3</sup>. Figure S1 shows normalized average density profiles as a function of height for different frequencies of the AC electric field. Blue dotted lines show fits to the ideal gas law,  $\rho = \rho_0 e^{-z/\delta_{eff}}$ , where  $\delta_{eff} = \frac{k_B T_{eff}}{m^* gsin\theta}$  corresponds to the effective sedimentation length, with  $T_{eff}$  the effective temperature,  $m^*$  the buoyant mass of a single microswimmer, and  $gsin\alpha$  the effective gravitational constant at the angle  $\alpha = 45^\circ$ . At f=800 Hz the data follow the shape of the ideal gas law in the lower part of the trap. However, the effective temperature we extract from the sedimentation length is slightly lower than the effective temperature extracted from the effective diffusion coefficient of the microswimmers (~  $10^5$  K versus ~  $10^6$  K, respectively), suggesting the importance of other factors such as inter-particle interactions. In contrast, at higher f the fit is not an accurate description of the data.



Figure S1: Vertical density profile of Janus microswimmers at different frequencies. Average number density profile as a function of height of Janus microswimmers at different frequencies and  $V_{pp} = 4$  V, taken from the 400  $\mu$ m center of the particle trap after 9 min. of equilibration. Blue dotted lines indicates best fits to ideal gas law,  $\rho = \rho_0 e^{-z/\delta_{eff}}$ .

S2. Condensation of microswimmers under gravity by switching the frequency of the AC electric field.



Figure S2: Condensation of microswimmers under gravity by switching the frequency of the AC electric field. High-magnification snapshots of the system described in the main text, captured at different times after switching the frequency from 800 Hz to 200 Hz. Panels (a–c) show raw images, while panels (d–f) display the corresponding annotated images. Regardless of the frequency, the caps of the microswimmers remain clearly visible. In the annotated images, particles are color-coded based on their cap orientation, with the caps artificially highlighted. Black arrows indicate the velocity vector direction for each particle. These images correspond to Figure 4e in the main text.

### S3. Comparison of Pd capped and bare $SiO_2$ spheres



800 Hz - t = 9 min. 500 Hz - t = 9 min. 300 Hz - t = 9 min. 200 Hz - t = 9 min.

Figure S2: Comparison of Pd capped and bare SiO<sub>2</sub> spheres. Snapshots of the system described in the main text after 9 min. of equilibration at different AC electric field frequencies at a peak-to-peak voltage of  $V_{pp} = 4$  V for (a-d) active Janus particles (SiO<sub>2</sub> spheres with a Pd cap) and (e-h) passive particles (uncoated SiO<sub>2</sub> spheres). Scale bars: 50  $\mu$ m.

#### S4. Time scale estimation

The relaxation time scales of ions in solution under the effect of an AC electric field have been extensively studied by Squires, Bazant, Ristenpart and Delgado in previous works<sup>4–6</sup>. To calculate the range of frequencies at which electrohydrodynamics (EHD) flows take place we have taken into account different parameters. In our solution, the conductivity and dielectric constant of the media are  $\sigma_{\rm m} = 1.5 \times 10^{-5} \, {\rm Sm}^{-1}$  and  $\epsilon_{\rm m} = 78 \, (25^{\circ}C)$ , respectively. Thus, the Debye length  $\kappa^{-1}$  is estimated as

$$\kappa^{-1} = \sqrt{\frac{\epsilon_{\rm m} \epsilon_0 D}{\sigma_{\rm m}}} \approx 300 \rm{nm}, \tag{1}$$

with  $\epsilon_0 = 8.854 \times 10^{-12} \text{Fm}^{-1}$ ,  $D = 2 \times 10^{-9} \text{m}^2 \text{s}^{-1}$  considering a general value for ions in the media to be able to do the calculation. Using the calculated Debye length, we estimate the charge relaxation time on the particle surface as

$$\tau_{\rm p} = \kappa^{-1} \mathrm{R/D} \approx 0.000225 \mathrm{s},\tag{2}$$

with  $R=1.5 \ \mu m$  the particle radius. This value corresponds to a frequency of  $f \approx 4.4 k H z$ . We furthermore estimate the charge relaxation time on the electrode surface as

$$\tau_{\rm p} = \kappa^{-1} 2\mathrm{h/D} \approx 0.0045\mathrm{s},\tag{3}$$

with  $2h=30 \ \mu m$  the separation between the two electrode. This value corresponds to a frequency of  $f \approx 200 Hz$ .

A summary of all the parameters used in the calculations in the main text and supplementary information are depicted in Table.S1

Parameters	Values
2h	$3.0 \times 10^{-5} \text{ [m]}$
R	$1.5 \times 10^{-6} [m]$
$\sigma_{ m m}$	$1.5 \times 10^{-5} [\text{Sm}^{-1}]$
$\epsilon_{ m m}$	78
$\epsilon_{0}$	$8.854 \times 10^{-12} [Fm^{-1}]$
D	$2 \times 10^{-9} [m^2 s^{-1}]$

Table S1: Parameters used in the calculation of the various timescales and other quantities in this system.

## **List of Supplementary Videos**

- Video S1: Video of the metallo-dielectric Janus particles confined in a rectangular trap under gravity after 9 minutes of equilibration under and AC electric field with frequency of 800 Hz and a peak-to-peak voltage of  $V_{pp} = 4$  V. Video corresponds to the snapshot in Figure 1e in the main text. The video is sped up 2 times.
- Video S2: Video of the metallo-dielectric Janus particles confined in a rectangular trap under gravity after 9 minutes of equilibration under and AC electric field with frequency of 500 Hz and a peak-to-peak voltage of  $V_{pp} = 4$  V. Video corresponds to the snapshot in Figure 1f in the main text. The video is sped up 2 times.
- Video S3: Video of the metallo-dielectric Janus particles confined in a rectangular trap under gravity after 9 minutes of equilibration under and AC electric field with frequency

of 300 Hz and a peak-to-peak voltage of  $V_{pp} = 4$  V. Video corresponds to the snapshot in Figure 1g in the main text. The video is sped up 2 times.

- Video S4: Video of the metallo-dielectric Janus particles confined in a rectangular trap under gravity after 9 minutes of equilibration under and AC electric field with frequency of 200 Hz and a peak-to-peak voltage of  $V_{pp} = 4$  V. Video corresponds to the snapshot in Figure 1h in the main text. The video is sped up 2 times.
- Video S5: Condensation of microswimmers under gravity by switching the frequency of the AC electric field from 800 Hz to 200 Hz at a fixed peak-to-peak voltage of  $V_{pp} = 4$  V. Microswimmers are color-coded according to their cap orientation. Black arrow indicated their direction of motion extracted from their velocity vector. Video corresponds to the snapshots in Figure 4e in the main text. The video is real time.

## References

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