Supporting Information

Two-dimensional glass-transition-like behavior of Janus particle-laden interface

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Images of the monolayer formed by Janus particles after deposition onto the air-water interface

The following micrographs were generated with A NIMA Langmuir trough (Biolin Scientific) equipped with an imaging window machined in the center mounted on top of an inverted microscope (IX73 Olympus). The imaging of the particle-laden interface was carried out using a 20x objective (6.6-7.8 WD, 0.45 NA). The trough is supported by a moving stage (Applied Scientific Instrumentation) and thus the trough position can be controlled precisely. Therefore, it is possible to image the microstructure at different locations throughout the monolayer for a better representation of the surface monolayer as shown in Figure S1.

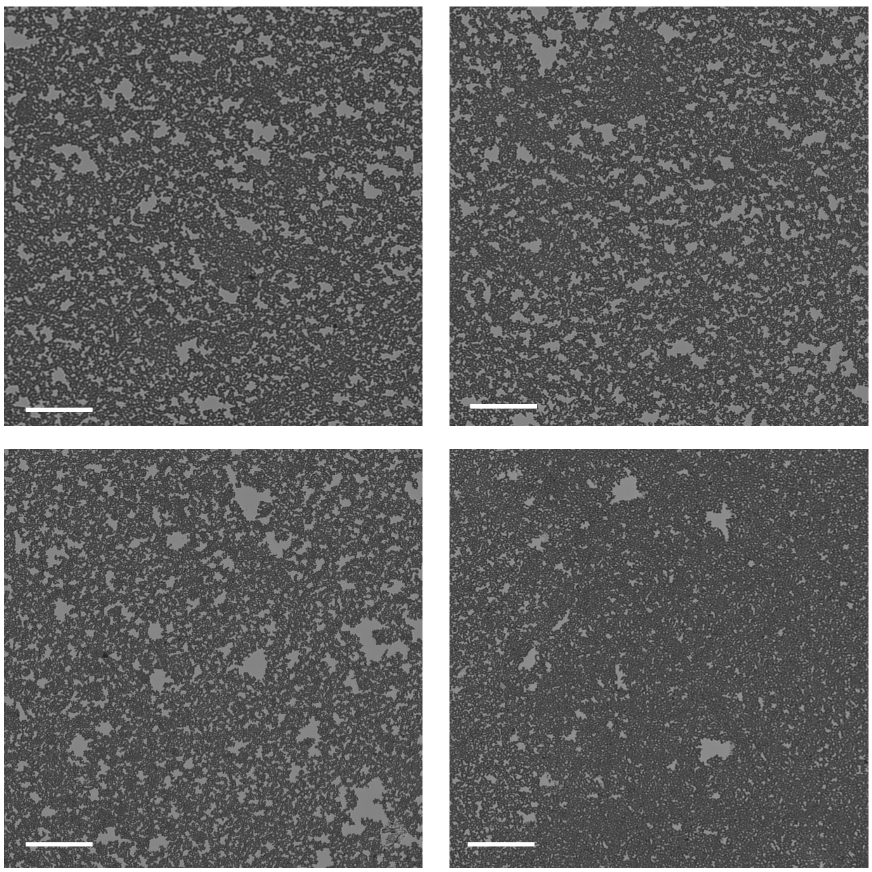

Figure S1 – Micrographs of the interfacial monolayer formed by Janus particles at the air-water interface sampled at different locations within the interface. Scale bar is 10 µm.

SEM images of Janus particles transferred from the air-water interface onto a silicon wafer

Scanning Electron Microscope (SEM) images were collected for particles deposited on a substrate examples of which are provided in Figure S2. The SEM utilized was a Field Emission environmental SEM from Thermo. For the image acquisition, we utilized an accelerating voltage of 10.00 kV and a working distance of 9.7 mm.

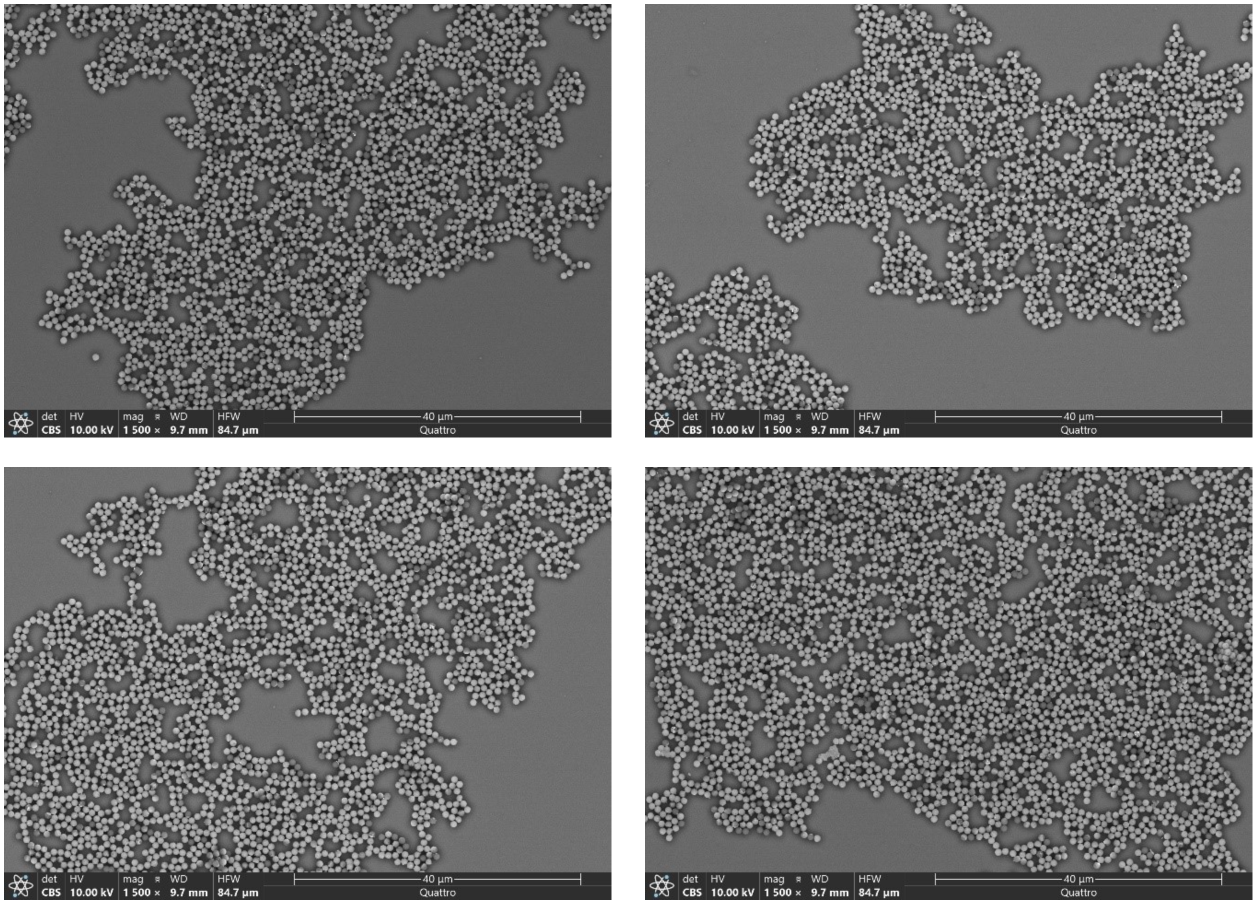

Figure S2 – SEM pictures of interfacial monolayer of Janus particles formed at the air-water interface imaged after transferring to a silicon wafer from the air-water interface.

From the captured SEM images, examples of which are shown in Figure S2, we calculated the crystallinity order parameter C_6 (Juárez and Bevan 2012), which is defined as the number of hexagonally packed neighbors of a specific particle. The number of neighbors (N_c^k) to the particle k are all particles j within a coordination radius (r_c). Identification of crystalline neighbors for particle k is based on a six-fold orientation order parameter for that particle, (ψ_6^k), which is calculated as follows:

$$\psi_6^k = \frac{1}{N_c^k}\sum_{j=1}^{N_c^k}\left[e^{i6\theta_{kj}} \qquad r_{kj} \leq r_c\right]$$

Next, we can calculate the crystalline connectivity (χ_6^{kj}) between particles k and j.

$$\chi_6^{kj} = \frac{\left|Re\left[\psi_6^k \psi_6^{j*}\right]\right|}{\left|\psi_6^k \psi_6^{j*}\right|}$$

where ψ_6^{j*} is the complex conjugate of ψ_6^j. The number of crystalline near neighbors (C_6^k) for particle k is as follows:

$$C_6^k = \sum_{j=1}^{N_c^k} \left[\begin{vmatrix} 1 & \chi_6^{kj} \geq 0.32 \\ 0 & \chi_6^{kj} < 0.32 \end{vmatrix} \right]$$

Which is based on a criterion that a connection between particles k and j only to be crystalline for $\chi_6^{kj} \geq 0.32$. Finally, we can calculate the ensemble average over all particles ($\langle C_6 \rangle$) as:

$$\langle C_6 \rangle = \frac{1}{N} \sum_{k=1}^{N} C_6^k$$

Following this procedure, and using ~ 50,000 particles imaged via SEM in this analysis, we estimated the $\langle C_6 \rangle$ of our particle system to be ~ 2.0 ± 0.1. Our data analysis was done on a modified version of the software HEXI (Domonkos, Jackivová, and Pathó 2022). In short, HEXI identifies individual particles and determines their position. With that information one can calculate the beforementioned properties.

References

Domonkos, Mária, Rajisa Jackivová, and Andor Pathó. 2022. 'Image analysis algorithm for the verification of hexagonal symmetry in spherical nanostructures', *Microelectronic Engineering*, 251: 111635.

Juárez, Jaime J, and Michael A Bevan. 2012. 'Feedback controlled colloidal self-assembly', *Advanced Functional Materials*, 22: 3833-39.