Electronic Supplementary Information

Translational and rotational dynamics in dense suspensions of smooth and rough colloids

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Validation of the MSD and MSAD measurements for smooth and rough colloids

As part of the process to validate the MSD and MSAD measurements in the colloidal suspensions, we plot the experimental data along with theoretical predictions for hard spheres available from the literature. Figure S1 shows that our measurements of the diffusivity for smooth colloids are in relatively good agreement with the Stokesian Dynamics work of Brady and co-workers [1, 2]. Deviations at high ϕ can be attributed to the finite repulsion between colloids in our system.

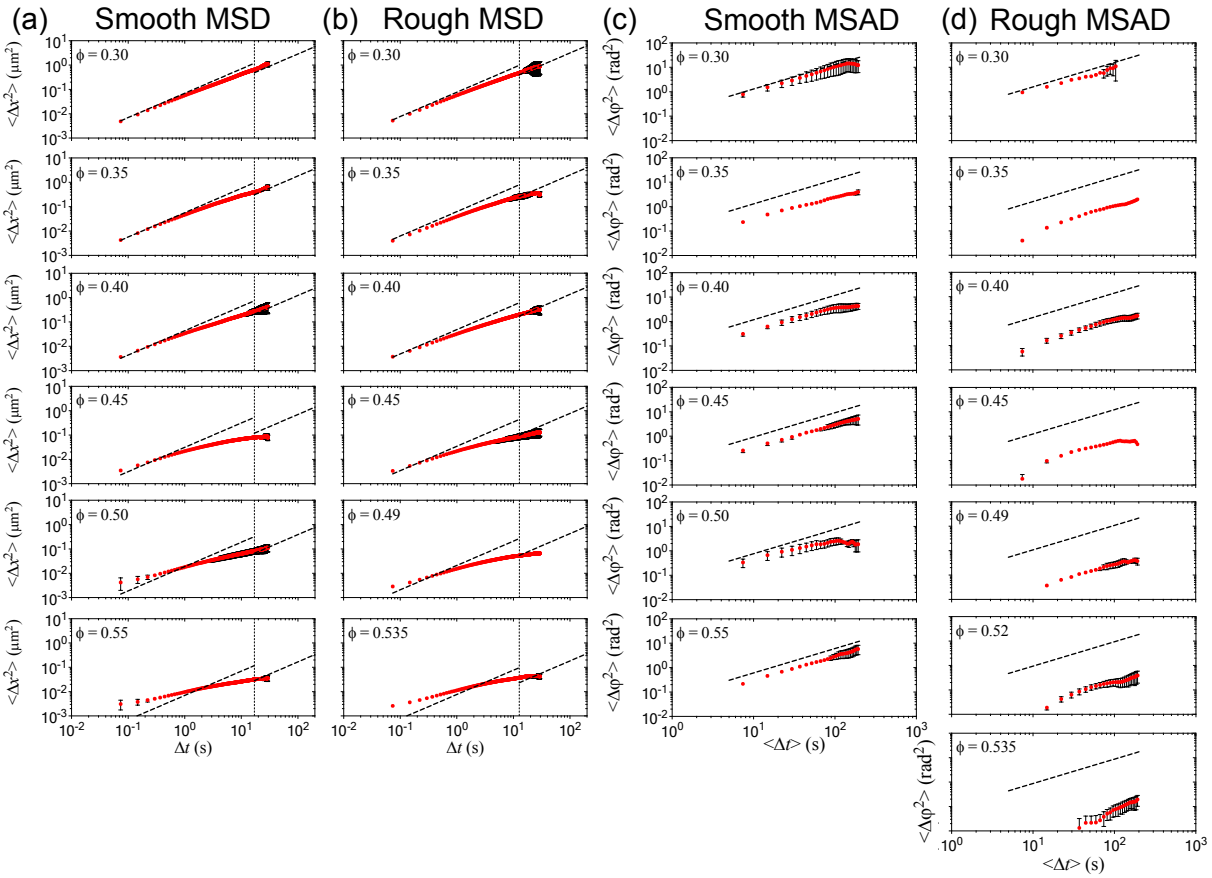

Figure S1. (a, b) The noise-corrected MSD and (c, d) noise-corrected MSAD measurements for smooth and rough colloids. Dashed lines in in (a, b) are short- and long-time translational diffusivity predictions and in (c, d) are short-time rotational diffusivity predictions from Stokesian Dynamics simulations of hard spheres [1, 2]. In (a, b), the transition between short time and long time diffusion is marked by the diffusive time scale, which refers to the average time it takes a particle to translate its own radius. Error bars represent standard deviation from independent measurements, where available.

Videos of Janus tracer particles in suspensions

Supplementary Video SV1 is a real-time 2D movie of a representative smooth Janus tracer in a suspension of smooth colloids ($\phi = 0.50$), while Supplementary Video SV2 is a real-time 2D movie of a representative rough Janus tracer in a suspension of rough colloids ($\phi = 0.50$). The translational and rotational dynamics of the suspensions are observable in these videos.

References

[1] R. Phillips, J. Brady, and G. Bossis. Hydrodynamic transport properties of hard‐sphere dispersions. I. Suspensions of freely mobile particles. *The Physics of Fluids* **31**, 3462-3472 (1988).

[2] J. F. Brady. The long-time self-diffusivity in concentrated colloidal dispersions. *Journal of Fluid Mechanics* **272**, 109-134 (1994).