Reply to the comment from Ikeda, Berthier, and Sollich (IBS) [1]

We thank IBS for their comments which question our interpretation of the universal viscosity divergence near the flow-arrest transition in constant stress and pressure rheology of hard-sphere colloidal suspensions [2]. IBS introduced two Péclet numbers: $\mathrm{Pe}_0 = \dot{\gamma}a^2/d_0$ and $\mathrm{Pe} = \dot{\gamma}a^2/d(\phi)$, with $\dot{\gamma}$ the strain rate, a the particle size, d_0 the isolated single-particle diffusivity and $d(\phi)$ the long-time at-rest self-diffusivity, and considered three regimes: (i) $\mathrm{Pe}_0 < \mathrm{Pe} \ll 1$, (ii) $\mathrm{Pe}_0 \ll 1 \ll \mathrm{Pe}$, and (iii) $1 \ll \mathrm{Pe}_0 < \mathrm{Pe}$.

IBS's claim that "only Pe is considered in [2]" is not true. The stress Péclet number $\mathrm{Pe}_\sigma = \sigma a^2/(\eta_0 d_0)$, with σ the imposed stress and η_0 the solvent viscosity, is a primitive input to our simulations. It compares the magnitude of the imposed stress relative to the particle thermal fluctuations, and is trivially connected to Pe_0 through $\mathrm{Pe}_\sigma = \eta \mathrm{Pe}_0$, with η the dimensionless shear viscosity.

Near the flow-arrest transition, Pe_0 is of little relevance to suspension dynamics. What drives an otherwise arrested suspension to flow are internal structural rearrangements, which are characterized by $d(\phi)$, not by the local "in cage" thermal fluctuations described by d_0. Near athermal jamming, i.e., close to the point $(\phi_{\mathrm{SAP}}, \mu_{\mathrm{SAP}})$ in Fig. 1, the condition $\mathrm{Pe}_0 \gg 1$ is not satisfied. Here, the imposed pressure $\bar{\Pi} = \mathrm{Pe}_\sigma/(6\pi\mu_{\mathrm{SAP}})$ satisfies $\bar{\Pi} \sim (\phi_{\mathrm{SAP}} - \phi)^{-\delta}$ with $\delta = 1$ near jamming [3]. Meanwhile, the universal viscosity divergence suggests $\mathrm{Pe}_0 \sim \mathrm{Pe}_\sigma(\phi_{\mathrm{SAP}} - \phi)^\gamma$ with $\gamma \approx 2$, which leads to $\mathrm{Pe}_0 \sim \mu_{\mathrm{SAP}}(\phi_{\mathrm{SAP}} - \phi)^{\gamma-\delta}$, independent of Pe_σ and $\bar{\Pi}$. Thus, $\mathrm{Pe}_0 \ll 1$ for $\gamma > \delta$, which is the case for hard-sphere suspensions when $(\mathrm{Pe}_\sigma, \phi) \to (\infty, \phi_{\mathrm{SAP}})$. IBS's distinction between regimes (ii) and (iii) is therefore unnecessary, and Pe alone is sufficient. This is also reflected in recent experiments [4] which show that suspensions enter the non-Brownian regime sooner, i.e., at lower Pe_0, with increasing ϕ—the shear stresses where the shear thinning regime ends are the same over a wide range of ϕ.

In regime (i), linear response theory requires $\Pi(\phi, \dot{\gamma}) = \Pi^{\mathrm{eq}}(\phi) + \Delta\Pi(\phi)\dot{\gamma}^2$ and $\sigma(\phi, \dot{\gamma}) = \eta_T(\phi)\dot{\gamma}$. Due to the different $\dot{\gamma}$ dependences, one can always evaluate $\eta_T(\phi)$ at sufficiently small $\dot{\gamma}$ with $\Pi \approx \Pi^{\mathrm{eq}}(\phi)$. In the low μ limit, constant Π and constant ϕ results are equivalent. This is shown in Fig. 1: Far from the glass transition ϕ_g, the contour at constant Π_1 asymptotes to the contour at constant ϕ_1 at a low but finite μ. Near ϕ_g, the contours at Π_2 and ϕ_2 approach each other as $\mu \to 0$. Therefore, by construction, our approach can probe the glass transition. On the other hand, the viscosity divergences observed along constant-ϕ and constant-Π contours may be different due to the different approaches to the arrested region [2], as illustrated by the viscosity contours in Fig. 1. Furthermore, it is still an open question whether the product $\eta_T(\phi)d(\phi)$ remains constant near

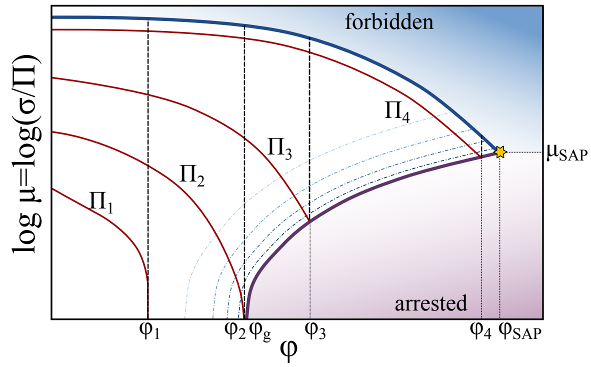

Figure 1. Sketch of the μ-ϕ flow map based on [2]. The thick curves enclose the flowing region, with the lower curve outlining the arrested region, and the upper curve outlining the non-Brownian limiting behavior. The two curves intersect at the Shear Arrest Point $(\mu_{\mathrm{SAP}}, \phi_{\mathrm{SAP}})$. The solid lines represent constant-Π contours at pressures $\Pi_1 < \Pi_2 < \Pi_3 < \Pi_4$. The dashed lines show the constant-ϕ contours at the corresponding at-rest volume fraction. The dash-dotted lines are the constant-viscosity contours.

ϕ_g, and, consequently, simulations and experiments of the relaxation time [5] cannot infer the viscosity divergence [1].

When the at-rest volume fraction is above ϕ_g, the diffusivity $d(\phi) \to 0$ and the suspension has a yield stress. This corresponds to IBS's regimes (ii) and (iii). Here, the viscosity is inherently non-Newtonian regardless of Pe_0, and exhibits universal divergences at constant Π. IBS's interpretation using a Herschel-Bulkey model for the pressure nicely complements our work. Our study is for true hard spheres whose behavior can be fundamentally different from soft-particle systems, even when the stiffness of the potential is increased [6] or the confining pressure is reduced [7] to eliminate particle overlaps. For example, in the non-Brownian limit, the singular hard-sphere potential leads to a finite shear viscosity despite the stress's thermal origin [8]. In the same limit, the viscosity from a soft potential (no matter how stiff) approaches zero.

Finally, we agree with IBS that in their regime (iii), our data are sparse since ϕ_{SAP} can only be approached from below in our simulations. However, as we have already pointed out, $\mathrm{Pe}_0 \gg 1$ cannot be achieved near athermal jamming, and our results agree with the viscosity divergence found in non-Brownian experiments [9].

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[1] A. Ikeda, L. Berthier, and P. Sollich, arXiv:1602.04796 (2016).
[2] M. Wang and J. F. Brady, Phys. Rev. Lett. **115**, 158301 (2015).
[3] M. D. Rintoul and S. Torquato, Physical Review Letters **77**, 4198 (1996).
[4] B. M. Guy, M. Hermes, and W. C. K. Poon, Phys. Rev. Lett. **115**, 088304 (2015).
[5] G. Brambilla, D. El Masri, M. Pierno, L. Berthier, L. Cipelletti, G. Petekidis, and A. Schofield, Phys. Rev. Lett. **102**, 085703 (2009).
[6] A. Ikeda, L. Berthier, and P. Sollich, Phys. Rev. Lett. **109**, 018301 (2012).
[7] T. Kawasaki, D. Coslovich, A. Ikeda, and L. Berthier, Phys. Rev. E **91**, 012203 (2015).
[8] J. F. Brady and J. F. Morris, J. Fluid Mech. **348**, 103 (1997); J. Bergenholtz, J. F. Brady, and M. Vivic, *ibid.* **456**, 239 (2002).
[9] F. Boyer, E. Guazzelli, and O. Pouliquen, Phys. Rev. Lett. **107**, 188301 (2011).