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: $\text{Pe}_0 = \gamma a^2/d_0$ and $\text{Pe} = \gamma a^2/d(\phi)$, with $\gamma$ 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) $\text{Pe}_0 < \text{Pe} \ll 1$, (ii) $\text{Pe}_0 \ll 1 \ll \text{Pe}$, and (iii) $1 \ll \text{Pe}_0 < \text{Pe}$.

IBS’s claim that “only Pe is considered in [2]” is not true. The stress Péclet number $\text{Pe}_\sigma = \sigma a^2/(\eta_0 d_0)$, with $\sigma$ the imposed stress and $\eta_0$ 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 $\text{Pe}_0$ through $\text{Pe}_\sigma = \eta_0 \text{Pe}_0$, with $\eta_0$ the dimensionless shear viscosity.

Near the flow-arrest transition, $\text{Pe}_0$ 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 $(\phi_{\text{SAP}}, \mu_{\text{SAP}})$ in Fig. 1, the condition $\text{Pe}_0 \gg 1$ is not satisfied. Here, the imposed pressure $\Pi = \text{Pe}_\sigma/(6\eta_0 \mu_{\text{SAP}})$ satisfies $\Pi \sim (\phi_{\text{SAP}} - \phi)^{-\delta}$ with $\delta = 1$ near jamming [3]. Meanwhile, the universal viscosity divergence suggests $\text{Pe}_0 \sim \text{Pe}_\sigma (\phi_{\text{SAP}} - \phi)^\gamma$ with $\gamma \approx 2$, which leads to $\text{Pe}_0 \sim \mu_{\text{SAP}} (\phi_{\text{SAP}} - \phi)^{\gamma-\delta}$, independent of $\text{Pe}_\sigma$ and $\Pi$. Thus, $\text{Pe}_0 \ll 1$ for $\gamma > \delta$, which is the case for hard-sphere suspensions when $(\text{Pe}_\sigma, \phi) \to (\infty, \phi_{\text{SAP}})$. IBS’s distinction between regimes (ii) and (iii) is therefore unnecessary, and $\text{Pe}_0$ 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 $\text{Pe}_0$, with increasing $\phi$—the shear stresses where the shear thickening regime ends are the same over a wide range of $\phi$.

In regime (i), linear response theory requires $\Pi(\phi, \dot{\gamma}) = \Pi(\phi) + \Delta \Pi(\phi) \dot{\gamma}^2$ 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 $\Pi \approx \Pi(\phi)$. In the low $\mu$ limit, constant $\Pi$ and constant $\phi$ results are equivalent. This is shown in Fig. 1: Far from the glass transition $\phi_g$, the contour at constant $\Pi$ asymptotes to the contour at constant $\phi_1$ at a low but finite $\mu$. Near $\phi_g$, the contours at $\Pi_2$ and $\phi_2$ 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-$\phi$ and constant-$\Pi$ 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 $\phi_g$, and, consequently, simulations and experiments of the relaxation time [5] cannot infer the viscosity divergence [1].

When the at-rest volume fraction is above $\phi_g$, 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 $\text{Pe}_0$, and exhibits universal divergences at constant $\Pi$. 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 $\phi_{\text{SAP}}$ can only be approached from below in our simulations. However, as we have already pointed out, $\text{Pe}_0 \gg 1$ cannot be achieved near athermal jamming, and our results agree with the viscosity divergence found in non-Brownian experiments [9].

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