Discontinuous Shear Thickening of Frictional Hard-Sphere Suspensions

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Discontinuous shear thickening (DST) observed in many dense athermal suspensions has proven difficult to understand and to reproduce by numerical simulation. By introducing a numerical scheme including both relevant hydrodynamic interactions and granularlike contacts, we show that contact friction is essential for having DST. Above a critical volume fraction, we observe the existence of two states: a low viscosity, contactless (hence, frictionless) state, and a high viscosity frictional shear jammed state. These two states are separated by a critical shear stress, associated with a critical shear rate where DST occurs. The shear jammed state is reminiscent of the jamming phase of granular matter. Continuous shear thickening is seen as a lower volume fraction vestige of the jamming transition.

Suspensions of particles at high volume fraction of solid, often termed dense suspensions, have a rich non-Newtonian rheology. This is particularly striking for the simple system of nearly rigid particles in a Newtonian fluid, which exhibits shear thinning, shear thickening, and normal stresses, the last associated with strong microstructural distortion, despite the dominant influence played in such mixtures by viscous (Stokes-flow) fluid mechanics [1]. The phenomenon of discontinuous shear thickening (DST) (see [2–5] and references therein) is especially fascinating. Suspensions exhibiting DST flow relatively easily with slow stirring, but become highly viscous or even seemingly solid if one tries to stir them rapidly. In a rheometer, the transition is seen at a critical shear rate for a given volume fraction. It is often found that DST is completely reversible [6]. DST typically occurs for a volume fraction that exceeds a threshold value \( \phi_c \), which depends on the nature of the suspended particles: increased nonsphericity or surface roughness seem to lower \( \phi_c \). Continuous shear thickening (CST) is observed below \( \phi_c \), and becomes weaker with decreasing volume fraction. Although counterintuitive, the abrupt or discontinuous increase of viscosity with increase of shear rate is a generic feature of dense suspensions [3, 7], occurring in both Brownian (colloidal) and non-Brownian suspensions. This ubiquity suggests the possibility of a single mechanistic basis applicable to the various types of suspension. DST has yet to be reproduced by a simulation method which can unambiguously point to the essential physical features necessary for its observation. This Letter presents a novel method able to identify these features.

Several possible mechanisms have been proposed as the origin of DST. An order-disorder mechanism [8–10] describes a low shear rate ordered flow with few interactions between particles that becomes unstable at high shear rates and evolves to a disordered, highly interacting viscous flow. A hydroclustering [6, 11–15] or (hydro)contact network [16, 17] mechanism attributes the thickening to clusters of particles “glued” together by the lubrication singularity. The competition between a force (Brownian or interparticle) tending to keep particles apart and the imposed shear strain, which tends to push particles together along the compressional axis, results in narrower interparticle gaps as the shear rate increases. The resulting clusters of particles move more rigidly, effectively increasing the viscosity. Neither of these scenarios makes a distinction between CST and DST, and the development of hydroclusters oriented with their dominant principal axis in the compressional quadrant in Brownian hard-sphere suspensions leads only to CST [18, 19] even at volume fractions as large as \( \phi = 0.58 \) [20]. A theoretical approach based on an ad hoc mode-coupling theory attempts to describe DST as a shear-induced glass transition [21–24]. Another suggested mechanism [5, 25–27] explicitly relates DST to the existence of an underlying jamming transition due to the frustration of the granularlike dilatancy by the confining stress.

The appropriate mechanism has been difficult to ascertain. Most of the mechanisms noted predict the shear rate above which shear thickening happens [5, 28, 29]. Experimentally, the order-disorder transition seems unnecessary [30], at least with a strictly ordered state [10]. Simulations based on purely hydrodynamic modeling, such as Stokesian Dynamics [12], show that hydroclusters appear in the semidilute regime and networks in the concentrated regime (\( \phi \gtrsim 0.5 \)), where they produce a (weak) CST [13, 16, 31, 32]. DST has never been reproduced by those models.

A key mechanical issue left largely unconsidered in previous simulation efforts is the occurrence of contacts, and, in particular, frictional contacts between particles. It is known that, despite the lubrication force, particle roughness can lead to contacts, resulting in qualitative changes from the expected behavior of ideal smooth hard particles [33, 34]. One consequence of surface contact is an increase of viscosity with increasing surface friction [35]. In a colloidal silica suspension exhibiting DST, increased particle roughness has been shown to lead to a smaller critical shear rate [36, 37]. Even for ideally
smooth spheres, such issues as finite particle deformability may play a role for the large stresses that arise at small interparticle gaps. Such small gaps, dropping to subnanometer scale even for noncolloidal particles, lead us to question the relevant physics of close particle interactions. The experiments cited above, as well as physical intuition, suggest that contact between particles is an essential ingredient of the mechanics of flow of highly concentrated suspensions.

In this Letter we introduce a numerical model merging hydrodynamics and features of granular physics. The model permits contacts between particles by assuming a cutoff in the singular resistance due to lubrication for a small interparticle gap in the Stokes regime. These contacts are treated with a model adopted from granular intuition, suggesting that contact between particles is an essential ingredient of the mechanics of flow of highly concentrated suspensions.

![Figure 1](image)

**FIG. 1.** (color online) (a), (b) Shear rate and stress dependence of the relative viscosity $\eta$, respectively. $\Gamma$ is the dimensionless shear rate. The open and filled symbols indicate the results for $n = 512$ and 2048, and the volume fractions are shown in the graphs. The friction coefficient is $\mu = 1$ except for the dashed and dotted blue lines, for which $\mu = 0.1$ and 0, respectively. Red symbols show the results with 1.5 times stiffer particles. (c) DST (red line) and CST (dashed line) are shown in the phase diagram. The former is expected to reach the jamming point $\phi_J$ for $\Gamma \to 0$, which is not seen because of log scale. The contour lines for $\phi < 0.56$ are labeled by the relative viscosity, $\eta(\phi, \Gamma)$. Before jamming (black domain), the shear jammed states (gray domain) exist. Observed flowing and jammed states at $\phi = 0.58$ are shown by circles and crosses, respectively.

For concentrated suspensions, the resistance matrix can be approximately obtained by neglecting the far-field or many-body effects and taking the leading terms of the pair hydrodynamic interactions [38]. In the simulations, we use the leading terms from the exact solution for two particles [39, 40] in order to handle bidisperse systems, but the following explanation assumes a monodisperse system for simplicity. There is a singular factor $1/h_{ij}$ in the resistance to relative motion of particles $i$ and $j$, where $h_{ij}$ is the interparticle gap. We argue that it is appropriate, in seeking to represent real suspensions, to relax the idealization to represent factors such as the finite roughness of particle surfaces. We regularize the lubrication by inserting a small length $\delta$ to prevent divergence at contact $h_{ij} = 0$ as in [41] ($\delta = 10^{-3}a$ is used, where $a$ is the particle radius). The squeezing mode of the lubrication force is written as

$$F_{\text{lub}}^{(i,j)} = -\alpha(h_{ij})\left((U^{(i)} - U^{(j)}) \cdot n^{(i,j)} r^{(i,j)}\right).$$

Here $\alpha(h) = 3\pi \eta a^2/(2h + \delta)$, where $\eta$ is the viscosity of the suspending fluid. $n^{(i,j)}$ is the unit vector along the line of centers from particle $i$ to $j$. Thus, the hydrodynamic force acting on a particle is approximately given as the sum of the regularized lubrication force and the Stokes drag $F_{\text{Stokes}}^{(i)} = -6\pi \eta a (U^{(i)} - U^{\infty}(r^{(i)}))$. The hydrodynamic forces scale with shear rate $\dot{\gamma}$, and hence there is no essential shear-rate dependence.

The contact force $F_C$ is activated for $h_{ij} < 0$. A simple spring-and-dashpot contact model [41–43] is employed to mimic frictional hard spheres; the normal force is proportional to the overlap $-h_{ij}$:

$$F_{\text{C,nor}}^{(i,j)} = k_n h_{ij} n^{(i,j)};$$

and to the tangential spring displacement $\xi^{(i,j)}$:

$$F_{\text{C,tan}}^{(i,j)} = k_t \xi^{(i,j)}$$

and $T_C^{(i,j)} = k_t a n_{ij} \times \xi^{(i,j)}$ (see [43] for details), where $k_t$ is the tangential spring constant. The tangential force is subject to Coulomb’s law.
Even with contact forces, ideal hard-sphere suspensions should be Newtonian, because different \( \dot{\gamma} \) result in the same particle trajectories, but with different time (\( \sim 1/\dot{\gamma} \)) and force (\( \sim \dot{\gamma} \)) scales. When trying to mimic hard spheres with linear springs, we should avoid introducing an artificial shear-rate dependence. We therefore choose \( k_n \) and \( k_t \sim \dot{\gamma} \), and turn the dashpot resistance to keep a short contact relaxation time (= \( 10^{-4}/\dot{\gamma} \)), in contrast to [16].

The shear-rate dependence is introduced by another force that is not scaled with \( \dot{\gamma} \), which we take as an electrostatic double-layer force [44]. The approximate form \( F^{i,j}_R = -C_\text{ae}\kappa h^{i,j} n^{i,j} \) is used for \( h^{i,j} > 0 \), with \( 1/\kappa = 0.05a \). The repulsive force acts to keep particle gaps wider, and is more effective at small shear rate, i.e., where the shear rate \( 1/\dot{\gamma} \) is longer. We thus introduce a dimensionless shear rate as a ratio of two force scales: \( \dot{\gamma} \equiv 6\pi\eta_0a^2\dot{\gamma}/F_R(h = 0) \), which is analogous to the Péclet number for Brownian suspensions.

Simulations are performed using Lees-Edwards boundary conditions. The simulation boxes are cubes for \( n = 512 \) particles and rectangular parallelipipeds (the shear plane is square, and the depth is one half of the other dimensions) for \( n = 2048 \). The influence of particle migration, as previously discussed [45], can be ruled out here since the system is homogeneous owing to the boundary conditions. A bidisperse system is investigated to avoid small shear rates, i.e., where the shear time \( 1/\dot{\gamma} \) is longer. We thus introduce a dimensionless shear rate as a ratio of two force scales: \( \dot{\gamma} \equiv 6\pi\eta_0a^2\dot{\gamma}/F_R(h = 0) \), which is analogous to the Péclet number for Brownian suspensions.

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DST is observed when the percolating network can elastically sustain an applied stress. This is only possible for a minimum volume fraction \( \phi \geq \phi_c \), where there are enough constraints to “lock” or “jam” the structure. The critical volume fraction can be identified as the shear jamming [46]: \( \phi_c = \phi_S(\mu) \). It is close to the values observed for the static jamming of frictional spheres, but it need not be the same [46]. When the suspension is forced to flow at high shear rate in a strain-controlled experiment, the viscosity is dominated by the yield stress of the solid network, which is itself influenced by the confinement. The network of contacts is constantly broken and reformed, going from one transient solid configuration to another.

For \( \phi < \phi_S(\mu) \), the CST is a vestige of this jamming transition. Even when the applied shear stress is larger than \( \sigma_{on} \), no strictly jammed contact network can form. Only underconstrained structures appear for \( \sigma > \sigma_{on} \) (or equivalently \( \dot{\Gamma} > \dot{\Gamma}_c \)), reminiscent of the jammed states seen for \( \phi > \phi_S(\mu) \). These structures still require a large applied stress to flow, as they are only deformable via collective rearrangements. Upon decrease of \( \phi \), these networks are increasingly underconstrained, and the high viscosity phase fades away. It is worth noticing that the low viscosity phase, essentially frictionless (as there are fewer frictional contacts), has similar behavior, forming force networks increasingly constrained as the volume fraction increases [49, 50]. It is indeed seen in FIG. 1 (a) that the viscosity also increases with \( \phi \) in this phase. However, the point where solid frictionless structures appear is only reached for \( \phi = \phi_J \), which is much larger than \( \phi_S(\mu) \). The fact that these two divergences occur at two different volume fractions is the cause for the blowup of the difference of viscosity between the low shear rate frictionless state and the high shear rate frictional state as \( \phi \to \phi_S(\mu) \).

The above results lead us to propose a schematic phase diagram for the shear thickening of athermal suspensions, represented in the \( \phi-\dot{\Gamma} \) plane in FIG. 1 (c). DST, denoted by a solid (red) line, occurs in the range \( \phi_S(\mu) < \phi < \phi_J \) for a critical shear stress \( \sigma_{on} \). Asymptotically, in the low viscosity frictionless phase, \( \eta \propto (1 - \phi/\phi_J)^{-q} \) with \( q = 2 \) [49], which gives for the critical shear rate \( \dot{\Gamma}_c \propto \sigma_{on} (1 - \phi/\phi_J)^{3} \). This scaling is, however, difficult to observe in our data range, as we are still rather far from the divergence. Above the red line, the shear stress is a yield stress of the shear jammed state, proportional to the pressure. For ideal hard spheres sheared at constant volume, this region would simply be inaccessible, as the yield stress would be infinite. Below \( \phi_S(\mu) \), CST occurs around an isostress dashed black line, which appears as the continuation of the DST red line. The stress is domi-
nated by the proximity of shear jammed states above this dashed line, and gives a viscosity $\eta \propto (1 - \phi/\phi_S(\mu))^{-q'}$ with an estimated $q' \approx 1.5$.

This phase diagram may well be valid even in the case of Brownian suspensions, where Brownian motion may play a role similar to the double-layer force, namely preventing contacts and reopening gaps at low shear rate.

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