# Shear flow of non-Brownian suspensions close to jamming

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The dynamical mechanisms controlling the rheology of dense suspensions close to jamming are investigated numerically, using a simplified model for the relevant dissipative forces. We show that the velocity fluctuations, which control the dissipation rate and therefore the effective viscosity of the suspension, are the same in quasi-static simulations as for the actual dissipative dynamics. We conclude that the statistical properties of grain trajectories – in particular the critical exponent of velocity fluctuations with respect to volume fraction $\phi$ –are largely independent of the dissipation mechanism. Rather they are determined by steric effects, which are the main driving forces in the quasistatic simulations. The critical exponent of the suspension viscosity with respect to $\phi$ can then be deduced, and is consistent with experimental data.

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Athermal disordered systems such as foams [1], emulsions [2], suspensions [3] or granular materials [4] exhibit a critical phase transition between a liquid-like and a solid-like mechanical behaviour, when the particle volume fraction $\phi$ crosses the jamming point $\phi_c$. For $\phi > \phi_c$, these amorphous systems can resist shear. The elastic shear modulus vanishes at $\phi_c$ with a critical exponent different from the mean field one [5, 6]. Above a yield stress $\sigma_Y$, vanishing at $\phi_c$, they present a non-Newtonian rheology, which has received contradictory explanations based on (i) the glassy dynamics of a system presenting scale-free energy distributions [7], (ii) self-activated plastic events [8, 9], (iii) the critical scaling laws of the shear modulus and of the coordination number [10]. Together with conventional molecular dynamics simulations (MD), quasistatic methods (QS) have been applied to study the plastic flow of these amorphous solids at the yield-stress $\sigma_Y$ [11–15]. It is generally assumed that QS accurately describe the dynamics of the true system in the limit of asymptotically small shear rate $\dot{\gamma}$. However, the existence of a proper quasistatic limit remains controversial, and there is growing evidence that quasistatic flows actually correspond to a finite-size dominated regime, with a correlation length that saturates at the system size [9, 15].

Symmetrically, for $\phi < \phi_c$, amorphous materials can flow under an infinitesimal shear stress $\sigma$ and present a viscosity $\eta$ diverging at $\phi_c$ like $\eta \propto (\phi_c - \phi)^{-\alpha}$. In the case of a suspension of non-Brownian particles, the best fit of recent experimental results give a critical exponent of $\alpha = 2.4$ for volume control experiments [16] and of $\alpha = 1.9$ for pressure control experiments [3] (inset of Fig. 1). By contrast, a mean field argument based on the average gap between particles, and which assumes that dissipation mostly takes place in the lubricated film separating particles, predicts an exponent $\alpha = 1$ [17].

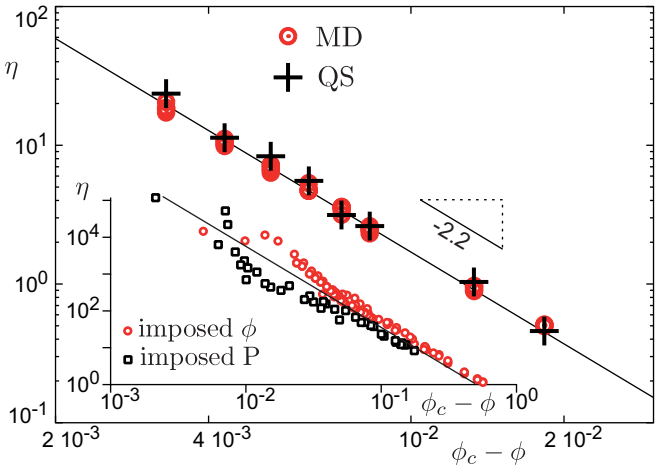


FIG. 1. Viscosity $\eta$ as a function of volume fraction $\phi$ measured from molecular dynamics (MD) and quasi static simulations (QS). Inset: compilation of experimental data available in the literature at imposed pressure $P$ (with $\phi_c = 0.587$) and imposed volume fraction $\phi$ (with $\phi_c = 0.615$).

The explanation of the critical exponent as well as the underlying mechanisms of the flow arrest have remained open and controversial questions up to now.

Here, we present simulation results for the viscous flow of a simplified model system in the vicinity of the close-packed state. We identify a *dynamical contribution* to the divergence of the viscosity, which has its origin in the singularity of velocity fluctuations. By comparing dissipative molecular dynamics simulations with a quasistatic energy minimization method, we obtain important clues on how universal aspects of the jamming transition are

interrelated with system-specific properties that depend on details of the particle interactions. More specifically, our results imply a decoupling scenario between particle trajectories and dissipative mechanism. In this picture, the statistical properties of trajectories are governed by the structural singularity of random close packing and the lack of available space for particle motion. On the other hand, "system-specific" dissipation mechanisms affect the rheological properties, via the dissipated energy along the given predetermined trajectories.

*Numerical set-up* – We consider a two-dimensional system constituted by $N$ soft spherical particles of unit mass, $N/2$ of unit diameter and $N/2$ of diameter 1.4. The particle volume fraction is defined as $\phi = \sum_{i=1}^{N} \pi r_i^2/L^2$, where $L$ is the size of the simulation box. Periodic (Lee-Edwards) boundary conditions are used in both directions. Two particles $i, j$ interact when their distance $r$ is smaller than the sum of their radii $r_i + r_j$, with a repulsive potential $E(r) = (1 - r/(r_i + r_j))^2$ whose unit multiplicative factor fixes the energy unit. We compare the divergence of viscosity for $\phi < \phi_c = 0.843$ [14, 18] using two different dynamics: non-equilibrium dissipative molecular dynamics (MD) and quasistatic simulations (QS).

*MD simulations* – In the MD simulations, the system is sheared at a shear rate $\dot{\gamma}$. Newton's equations of motion are integrated with elastic contact forces and a viscous drag force

$$\vec{F}^{\rm visc}(\vec{v}_i) = -\zeta \delta v_i = -\zeta \left[ (\vec{v}_i - \vec{v}_{\rm flow}(\vec{r}_i)) \right] . \qquad (1)$$

proportional to the velocity difference $\delta v_i$ between the particle velocity $\vec{v}_i$ and the flow velocity $\vec{v}_{\rm flow}(\vec{r}_i) = \hat{e}_x \dot{\gamma} y$, whose fluctuations are neglected [19–22]. $\zeta$ is a multiplicative factor proportional to the fluid viscosity that compares viscous to elastic effects. Thermal and lubrication forces are not accounted for. Unlike in granular systems, the particle-particle collisions are elastic and the only dissipation is due to viscous losses associated to the fluctuations of the particle velocity field. The shear stress $\sigma$ is calculated from the particle positions $\vec{r}_i = (x_i, y_i)$ and the forces $\vec{F}_i = (F_{ix}, F_{iy})$ acting on them as

$$\sigma = L^{-2} \sum_{i=1}^{N} x_i F_{iy} . \qquad (2)$$

The dominant contribution comes from the elastic forces that result from particle overlaps. The resulting relation between the shear stress $\sigma$ and the shear rate $\dot{\gamma}$ is shown in Fig. 2. For small strain rates the stress grows linearly with strainrate, $\sigma = \eta \dot{\gamma}$, characteristic for a simple Newtonian fluid with viscosity $\eta$. At larger strainrates and for small dissipative coefficients $\zeta$ the stress grows faster than linearly. This shear-thickening behavior is the signature of inertial effects. A shear-thinning regime (not shown here) appears at a volume fraction $\phi$ closer to $\phi_c$ [22].

*QS simulations* – Quasistatic simulations consist of successively applying small steps of shear and minimizing the total potential energy. By construction, they generate particle trajectories at $\dot{\gamma} \to 0$. An elementary strain step of $\gamma_0 = 5 \cdot 10^{-5}$ is used. After each change in boundary conditions the particles are moved affinely to define the starting configuration for the minimization, which is performed using conjugate gradient techniques [23]. The minimization is stopped when the nearest energy minimum is found. As no static, force-balanced state exists below the jamming point ($\phi < \phi_c$), the inter-particle forces at the minimum are strictly zero; the particles can always arrange in such a way as to avoid mutual overlaps. Thus, each minimized configuration corresponds to a true hard-sphere state and the resulting particle trajectories can be viewed as a sequence of snapshots of a dynamically evolving hard-sphere system. Particle motion in such a system is driven by steric exclusion and the lack of free volume. In particular, particles have to move ever larger distances, when the jamming point is approached, before they are able to find new overlap-free states compatible with the imposed shear [24].

We now determine the shear stress and the viscosity from the QS trajectories. Of course, during the QS simulation, no particle overlap occurs so that all contact forces are strictly zero. Still, we can determine the power $\Gamma$ per unit surface that would be dissipated along the QS trajectories, if the dissipation mechanism of Eq. (1) was present. $\Gamma$ is equal to the power injected per unit volume in the system, $\sigma\dot{\gamma}$, and can be expressed as:

$$\Gamma = L^{-2} \left\langle \sum_i \vec{F}^{visc}(\vec{v}_{i,\rm qs}) \cdot (\vec{v}_{i,\rm qs} - \vec{v}_{\rm flow}(\vec{r}_i)) \right\rangle$$

From this expression, we deduce the viscosity:

$$\eta = \frac{\Gamma}{\dot{\gamma}^2} = -\zeta \frac{N}{L^2} \frac{\langle \delta v^2 \rangle}{\dot{\gamma}^2} = -\zeta \frac{N}{L^2} \int \Delta^2 P(\Delta) d\Delta . \qquad (3)$$

where $P(\Delta)$ is the probability distribution function of the particle velocity rescaled by the shear rate: $\Delta = \delta v/\dot{\gamma}$. As particle coordinates in the QS simulation are only available at discrete steps, one has to define an effective particle velocity $\vec{v}_{\rm qs} = \dot{\gamma} \delta\vec{r}/\gamma_0$ from the particle displacement $\delta\vec{r}$ during such a single step. Therefore, $\Delta$ is also the displacement rescaled by the strain interval $\gamma$ – $P(\Delta)$ is the van Hove function. Note that the viscosity is related to the second moment of the velocity fluctuations, and thus of $P(\Delta)$ (Eq. 3).

*Statistical equivalence of MD and QS trajectories* – To characterise statistically the trajectories we look at the probability distribution for particle velocities, $P(\Delta)$. We concentrate on the velocity component in the gradient direction ($y$-component), which automatically eliminates trivial particle motion due to the average flow field. In order to highlight the contributions to the viscosity, we have chosen to plot the quantity $\Delta^2 \mathcal{P}(\Delta)$ in Fig. 3.

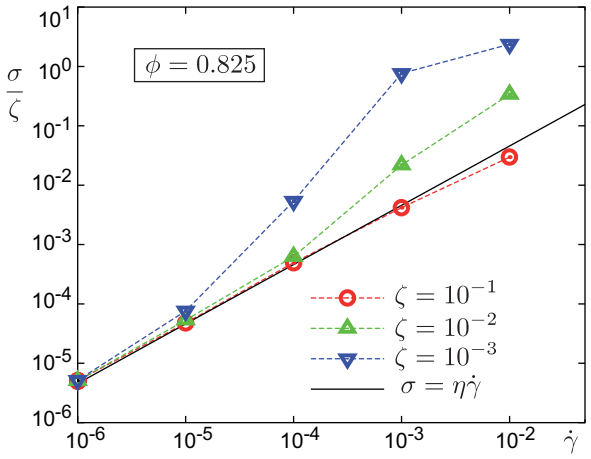


FIG. 2. Relation between stress $\sigma$ and strain rate $\dot\gamma$ obtained from MD simulations for different $\zeta$, for the same volume fraction $\phi = 0.825$. The solid line shows the value of the viscosity $\eta$ determined from the quasi-static simulation.

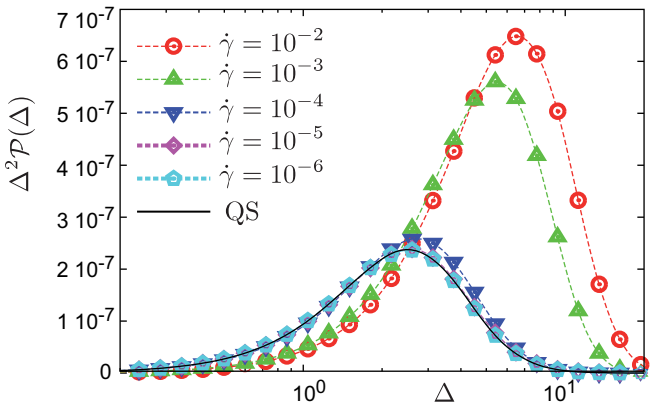


FIG. 3. Probability distribution function $P(\Delta)$ of the rescaled velocity fluctuations $\Delta = \delta v/\dot\gamma$. It is determined from the Van Hove function measured for $\gamma = 5\,10^{-4} = 10\,\gamma_0$, for $\zeta = 10^{-2}$ and different strain-rates. Volume fraction $\phi = 0.825$.

When the strain rate is small enough, $P(\Delta)$ asymptotes to a limiting form. In this limit, particle velocities simply scale linearly with the strain rate, $\delta v \propto \dot\gamma$, which is directly related to the Newtonian flow regime of Fig. 2. Whenever the rheology deviates from Newtonian behaviour (shear-thickenning or shear-thinning), the distribution function deviates from its asymptotic form.

Beside, we also find essentially the same functional form in the QS simulation. Therefore, the real trajectories of the MD simulation in the Newtonian regime are statistically identical to the trajectories of the QS simulation. The Newtonian flow regime can therefore be considered as a true quasi-static limit, which is by no means obvious. In fact, the QS limit seems much better defined here ($\phi < \phi_c$) than in the plastic flow regime ($\phi > \phi_c$), where QS simulations have usually been applied. The most important consequence is that the QS trajectories can be used to determine the flow rheology, and to determine the dynamical mechanism controlling it. As has been said above, the QS trajectories correspond to the motion of effectively impenetrable, hard sphere particles. Therefore, it seems that steric exclusion and the lack of available space close to the jamming transition are the dominant driving mechanisms for the nontrivial particle motion that is observed. The precise dissipative process, on the other hand, only plays a minor role, as shown by the equivalence between a real dissipative force (MD) and an unphysical conjugate gradient minimization (QS). One can therefore conclude that particle motion (trajectories) and dissipation mechanism are uncoupled. The trajectories are determined by steric effects independently of the specific dissipation mechanism and dissipation only becomes important for the rheological properties.

*Viscosity exponent close to jamming* – Fig. 2 compares the rheology obtained using molecular dynamics (data points) and quasi-static simulations (straight line). They nicely collapse on each other when MD simulations are considered in the limit of small shear rate. For MD, we used in practice $\dot\gamma = 10^{-6}$ to determine the viscosity $\eta$. Fig.1 shows the viscosity $\eta$ determined from both simulations, as a function of volume fraction $\phi$. Beyond noting the quality of the collapse, one observes that the viscosity diverges with $\phi_c - \phi$, with a scaling exponent $\simeq 2.2$ consistent with the value measured experimentally.

To further evidence the suggested decoupling between particle motion and dissipation we have conducted additional simulations where a modified dissipative force

$$\vec F^{\rm visc}(\vec v_i) = -\zeta\delta v\,|\delta\vec v|^{\nu-1}\,, \qquad (4)$$

with a variable exponent $\nu$ is used. In addition to the value of $\nu = 1$, which gives back Eq. (1), the two different values of $\nu = 2/3$ and $\nu = 4/3$ have been tested (Fig.4).

As a result we find that the rheological properties do indeed depend on the value of the exponent $\nu$, with a small-strainrate regime where $\sigma = \hat\eta\dot\gamma^\nu$. By way of contrast, the velocity distribution function $P(\Delta)$ is nearly independent of the choice of $\nu$. In particular, the maximum of $\Delta^2 P(\Delta)$ remains located at the same place as in the QS simulation. The statistical occurence of rare events, which make up the tail of $P(\Delta)$, slightly depend on the value of $\nu$; smaller values of $\nu$ lead to a stronger tail (and therefore to a reduced weight in the peak). Nevertheless, a calculation similar to Eq. (3) can again be used to accurately predict the shear stress/shear rate relation for any value of $\nu$, starting from one and the same QS simulation (black lines in Fig.4a). It turns out that the details of the tails of $P(\Delta)$ do not affect the average

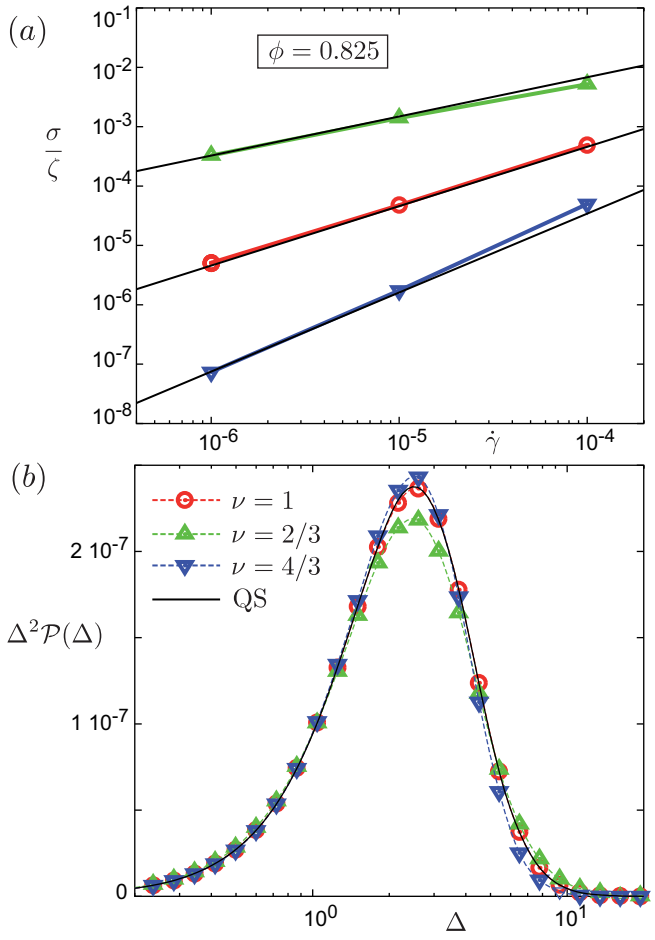


FIG. 4. (a) Relation between shear stress $\sigma$ and strain rate $\dot{\gamma}$ for different values of $\nu = 2/3, 1, 4/3$ ($\phi = 0.825$, $\zeta = 0.1$). The solid (black) lines are the prediction from the QS simulation along the lines of Eq. (3). (b) Probability distribution function $P(\Delta)$ of the rescaled velocity fluctuations $\Delta$ for the same systems at $\dot{\gamma} = 10^{-6}$.

rheological properties (the integral of $\Delta^2 P(\Delta)$).

Eq. (4), giving the power dissipated per unit volume, leads to the scaling law $\eta \sim \delta v^{1+\nu}$, which connects the divergence of the macroscopic viscosity to the scaling law followed by the microscopic particle motion $\delta v \sim (\phi_c - \phi)^{-\beta}$. Thus, the seemingly harmless power balance turns into a relation between the exponents controlling the divergence of velocity fluctuations and that of viscosity: $\alpha = \beta(1+\nu)$. We have recently shown that $\beta \approx 1.1$ [24], which gives $\alpha \simeq 2.2$, consistent with the exponent extracted from the MD data in Fig. 1. Note however, that subdominant corrections can lead to apparent exponents that may not reflect the true asymptotic behavior[18, 25].

In conclusion, we wish to emphasize that the singular nature of the velocity fluctuations, $\delta v \sim (\phi_c - \phi)^{-\beta}$ results into a *dynamical contribution* to the divergence of the viscosity, which had not previously been identified. For example, short-range lubrication forces have been argued to lead to a divergence as $\eta \sim (\phi_c - \phi)^{-1}$, due to the average gap separating densely packed particles. If the observed decoupling between particle motion and dissipation mechanism also holds for the lubrication force between particles, the dynamical contribution due to the singular velocity fluctuations would just superimpose on this divergence, effectively leading to a higher exponent $\alpha = 1 + 2\beta$.


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