Microfluidic Rheology of Soft Colloids above and below Jamming

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The rheology near jamming of a suspension of soft colloidal spheres is studied using a custom microfluidic rheometer that provides the stress versus strain rate over many decades. We find non-Newtonian behavior below the jamming concentration and yield-stress behavior above it. The data may be collapsed onto two branches with critical scaling exponents that agree with expectations based on Hertzian contacts and viscous drag. These results support the conclusion that jamming is similar to a critical phase transition, but with interaction-dependent exponents.

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The steady shear flow rheology is measured with the microfluidic device sketched in Fig. 2(a). It consists of a rectangular PDMS channel, 25 μm wide × 100 μm deep × L = 2 cm long, fabricated with standard soft lithography [14] and bonded to a glass microscope slide. The fluid is forced through the channel using pressurized air and inlet/outlet tubing of sufficient diameter that the imposed pressure drop \( P \) occurs only along the length \( L \) of the channel. Force balance therefore allows the shear stress at distance \( y \) from the center of the channel to be computed as \( \sigma(y) = (\Delta P/L)y \). The corresponding strain rate at \( y \) is found by numerical differentiation of the velocity profile, \( \dot{\gamma}(y) = dv_x(y)/dy \) [15]. For this, we collect video data with a Phantom CMOS camera (1–10 000 fps) connected to a Zeiss Axiovert 200 microscope with 100x objective focused at midheight. Since the channel is tall, the observed flow is equivalent to that between parallel plates [15]. An objective-cooling collar (Bioptechs) and cooling plate above the sample are controlled to about 0.1°C in order to vary the volume fraction. An example video frame in Fig. 2(c) displays bead-scale intensity variations, so that particle image velocimetry may be implemented with custom LABVIEW code. Example velocity profiles are superposed on the still image of Fig. 2(c). Altogether, for a single pressure drop, the \( \sigma(y) \) and \( \dot{\gamma}(y) \) data may thus be combined to give stress vs strain rate shear rheology. The dynamic range is typically two decades in \( \dot{\gamma} \), and may be extended by varying the imposed pressure drop.

This rheology concept has been realized previously [16,17], and is related to experiments [18–20] where the shape of a velocity profile is used to characterize shear rheology. Microfluidics is an ideal platform, since the channels are long compared to width so that entrance or exit effects are easily avoided. And owing to the small scale, high strain rates may be achieved at low Reynolds numbers, so that inertial flow instabilities are avoided. Furthermore, the local strain rate is directly measured, and hence no problems arise from wall slip or shear banding as typically hamper use of conventional rheometers for materials with a yield stress.

Results for the stress vs strain rate are collected in Fig. 3. Good agreement is found for multiple pressure drops at the same volume fraction. This demonstrates the reproducibility and level of uncertainty in our data; it also implies the absence of nonlocal effects, by contrast with Refs. [17,20]. Note that the data show a clear distinction in functional form above and below \( \phi_c \). For low \( \phi \), the stress tends towards zero at low strain rates. For higher \( \phi \), the stress extrapolates toward a nonzero yield stress \( \sigma_y \). We find qualitatively similar results using a conventional rheometer. To analyze the flow curves, we first fit the stress data to the phenomenological Herschel-Bulkley form:

\[
\sigma = \sigma_y \left[ 1 + (\dot{\gamma})^\delta \right]
\]
where $\beta$ is the shear-thinning exponent, $\tau$ is a time constant, and $K$ is called the consistency. The quality of the fits is satisfactory, as shown by the dashed curves in Fig. 3 for $\phi > \phi_c$. The results for $\beta$ displayed in Fig. 4(a) exhibit no apparent dependence on $\phi_c$ and have average and standard deviation $0.48 \pm 0.03$. For other microgel systems, Ref. [21] discusses yield-stress behavior and Ref. [22] finds $\beta = 0.45$, while Refs. [23–26] fit to forms that cross between different limiting viscosities at low and high strain rates. The value $\beta = 1/2$ is predicted near jamming for viscously-interacting athermal particles [10]. For simplicity, and so that $K$ has constant units, we henceforth fix $\beta = 1/2$ and repeat the fits.

The fitting parameters $\sigma_\gamma$ and $\tau$ are collected in Figs. 4(b) and 4(c) as a function of $\phi$. Both the yield stress and the time constant have been rendered dimensionless by appropriate factors of the elasticity $E$ of the particulate material and the viscosity $\eta_0$ of the suspending fluid. This also serves to eliminate the spurious $\phi$ dependence originating from the variation of $E$ with particle swelling. While $K$ is always well defined, $\sigma_\gamma$ and $\tau$ exist only above jamming and, respectively, appear to vanish and diverge on approach to $\phi_c$. As shown in the main plots, the results may be fitted to power-law forms $\sigma_\gamma/E \sim (\phi - \phi_c)^\alpha$ and $\tau E/\eta_0 \sim 1/(\phi - \phi_c)^\Gamma$, giving $\{ \phi_c = 0.633 \pm 0.002, \Delta = 2.2 \pm 0.4 \}$ and $\{ \phi_c = 0.637 \pm 0.002, \Gamma = 3.8 \pm 0.6 \}$. The two values for $\phi_c$ are in agreement and average to $0.635 \pm 0.003$, consistent with random close packing of spheres. Fixing $\phi_c$ to this value, we plot $\sigma_\gamma/E$ and $\tau E/\eta_0$ vs $\phi - \phi_c$ on log-log axes as insets in Figs. 4(b) and 4(c). These demonstrate power-law behavior, and give the final refined scaling exponents as $\Delta = 2.1 \pm 0.2$ and $\Gamma = 4.1 \pm 0.3$. However, we will conservatively take the final statistical uncertainties to be twice as large, as given by fits where $\phi_c$ floats. The systematic errors based on the allowed range of $\beta$ are 0.1 and 0.4 for $\Delta$ and $\Gamma$, respectively. Note that $\Delta = \beta\Gamma$ holds within uncertainty, which is required so that $K$ remains finite and nonzero at $\phi_c$ and so that at high strain rates the stress scales as $(\eta_0 \dot{\gamma})^\beta E^{(1 - \beta)}$ independent of $\phi$. Also, the very same exponents are found within experimental uncertainty for NIPA particles about 8 times less massive [15].

Our experimental value of $\Delta$ agrees with that simulated in Ref. [8], and our full suite of $\{\beta, \Delta, \Gamma\}$ values are in remarkably good agreement with those predicted in Ref. [10]. The observed value of the yield-stress exponent may be understood physically in terms of the scaling of the shear modulus $G$ and the yield strain $\gamma_\gamma$. For repulsive particles with interaction energy proportional to overlap raised to the power $\alpha$, numerical simulations find $G \sim (\phi - \phi_c)^{\alpha - 3/2}$; this differs from the naive expectation $\alpha - 2$ due to $\phi$-dependent nonaffine motion [2,4]. If the yield strain scales as $\gamma_\gamma \sim (\phi - \phi_c)^{\alpha/2}$, then $\Delta = \alpha - 1/2$ [10]. For Hertzian elastic particles, $\alpha = 5/2$, this predicts $\Delta = 2$ and $\Gamma = \Delta/\beta = 4$ as seen here.

The “distance” $\phi - \phi_c$ to jamming thus controls the yield stress $\sigma_\gamma$ and the time constant $\tau$ appearing in the Herschel-Bulkley form of stress vs strain rate, Eq. (1), according to respective scaling exponents $\Delta$ and $\Gamma$. Therefore, for volume fractions above $\phi_c$, the shear rheology data should all collapse onto a single master curve when plotted dimensionlessly as $\sigma_\gamma/E(\phi - \phi_c)^\alpha$ vs $\eta_0 \dot{\gamma}/(E(\phi - \phi_c)^{\Gamma})$. This construction and the required collapse for $\phi > \phi_c$ are demonstrated in Fig. 5. A noteworthy feature of this plot is that collapse also occurs for all data below jamming, for $\phi < \phi_c$, onto a distinct branch. Note that the two branches merge close to where the dimensionless scaled stress and strain rate are both near 1, which is reassuring. The collapse along a second branch need not have happened, and serves to emphasize that behavior is controlled by distance to point $J$—just as second order phase transitions are controlled by distance to criticality. The quality of the collapse is only slightly better using

![Graph showing fitting parameters vs volume fraction](image-url)
\begin{align*}
\beta &= 0.48 \pm 0.03 \\
\Delta &= 2.1 \pm 0.4 \pm 0.1 \\
\Gamma &= 4.1 \pm 0.6 \pm 0.4
\end{align*}

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