First publ. in: Physical Review Letters 89 (2002), Article 248304

Theory of Nonlinear Rheology and Yielding of Dense Colloidal Suspensions

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(Received 14 March 2002; published 25 November 2002)

A first-principles approach to the nonlinear flow of dense suspensions is presented which captures shear thinning of colloidal fluids and dynamical yielding of colloidal glasses. The advection of density fluctuations plays a central role, suppressing the caging of particles and speeding up structural relaxation. A mode coupling approach is developed to explore these effects.

PACS numbers: 82.70.Dd, 64.70.Pf, 83.60.Df, 83.50.Ax

The properties of dispersions under flow are central to their processing and technological use [1,2]. But especially the nonlinear rheology is not yet well understood. For the simplest case of steady shearing, the low density behavior is known [3], but upon increasing the density the growing importance of particle interactions requires theoretical approximation [4,5], hinders simulations [6], and calls for studies of model systems, e.g., [7,8]. Of major interest is the arrest of the structural relaxation when approaching solidification for higher densities, which raises the question of how the imposition of steady shearing might interfere with glass formation. The linear phenomenology is familiar: a colloidal fluid possesses a viscosity and flows, while a colloidal glass characterized by elastic constants only distorts under strain [1,2]. But the nonlinear rheology of glassy colloids, which exhibit a continuous slowing down of the structural relaxation due to particle blocking (the "cage effect") [1], is less clear. While the mode coupling theory (MCT) recovers the linear phenomenology of this fluid-to-glass transition from microscopic starting points [9], a nonlinear external driving introduces new time scales whose influence on (non)equilibration is not understood, and has been addressed only in minimal models [10] or mean-field approaches [11]. Moreover, as the true nature of the glass is still uncertain, its behavior under shearing may provide broader new insights (as suggested by recent simulation studies [12,13]).

Here we develop a first-principles approach for the simplest case of a disordered colloidal suspension under steady imposed shear, neglecting both many-body hydrodynamics and the resulting velocity fluctuations. We first identify some generic features in the yield properties of glass; approximations suggested by the MCT are then introduced in order to derive quantitative predictions.

The system consists of N spherical particles (diameter d) dispersed in a volume V of solvent with imposed flow profile $\mathbf{v}(\mathbf{r}) = \boldsymbol{\kappa}\mathbf{r}$, where for simple shear with velocity along the x axis and its gradient along the y axis, the shear rate tensor is $\kappa_{ij} = \dot\gamma\,\delta_{ix}\delta_{jy}$. The effect of the shear rate $\dot\gamma$ on the particle dynamics is measured by the Peclet number [2], $\mathrm{Pe}_0 = \dot\gamma d^2/D_0$, formed with the bare diffusion coefficient D_0 of a particle. Dimensionless units are obtained by setting $d = D_0 = k_BT = 1$. The evolution of the distribution function Ψ of the particle positions, $\mathbf{r}_i$, $i = 1, \ldots, N$, under internal forces $\mathbf{F}_i$ and shearing, but neglecting hydrodynamic interactions, is given by the Smoluchowski equation [2,14]:

$$\partial_t\Psi = \Omega^{(\dot\gamma)}\Psi,$$
$$\text{where } \Omega^{(\dot\gamma)} = \sum_i \boldsymbol{\partial}_i \cdot (\boldsymbol{\partial}_i - \mathbf{F}_i - \boldsymbol{\kappa}\mathbf{r}_i). \qquad (1)$$

The system is taken to be in quiescent equilibrium ($\dot\gamma = 0$) at $t \le 0$ when averages $\langle\cdots\rangle^{(\dot\gamma=0)}$ are the canonical equilibrium ones. Then at $t = 0^+$, the velocity profile is switched on instantaneously, so that the steady-state distribution function Ψ_s, which satisfies $\Omega^{(\dot\gamma)}\Psi_s = 0$, will be approached at long times, $t \to \infty$. If Ψ_s was known, the steady-state average $\sigma = \langle\sigma_{xy}\rangle^{(\dot\gamma)}$ of the (thermodynamic) shear stress [15] could be found. From this the shear viscosity would follow as $\eta(\dot\gamma) = \eta_\infty + \sigma/\dot\gamma$ (where the solvent viscosity is denoted η_∞). But the rapid increase of $\eta(0)$ close to the glass transition suggests that Ψ_s is sensitively dependent on shear rate, which makes a direct calculation of it difficult.

A more robust way to approximate steady-state quantities comes from the insight that the growth of η at the glass transition arises by slowing down structural relaxations [9], whose characteristic time τ in the quiescent state defines a second, "dressed" Peclet (or Weissenberg [1]) number, $\mathrm{Pe} = \dot\gamma\tau$. This characterizes the influence of shear on structural relaxation and increases without bound at the glass transition, even while $\mathrm{Pe}_0 \ll 1$. We argue that the competition of structural rearrangement and shearing that arises when $\mathrm{Pe} > 1 \gg \mathrm{Pe}_0$ dominates the nonlinear rheology of colloids near the glass transition. Therefore steady-state quantities shall be determined by considering the structural relaxation under shearing and "integrating through the transient dynamics." Because $\mathrm{Pe}_0 \ll 1$, we expect ordering or layering transitions to be absent [7]; and as hydrodynamic interactions are presumed to play a subordinate role during the structural relaxation [5] we neglect these too, focusing

solely on the Brownian contribution to the transverse (shear) stress.

With t_0 the time passed since the start of shearing, the correlation function of fluctuations in variables f and g separated by time t is given by

$$C_{fg}(t, t_0) = \langle e^{\Omega_B^{(\dot\gamma)} t_0} f^* (e^{\Omega_B^{(\dot\gamma)} t} g)\rangle^{(0)}, \tag{2}$$

where the backwards Smoluchowski operator $\Omega_B^{(\dot\gamma)}$ arises from partial integrations: $\Omega_B^{(\dot\gamma)} = \sum_i (\boldsymbol{\partial}_i + \mathbf{F}_i + \mathbf{r}_i \boldsymbol{\kappa}^T) \cdot \boldsymbol{\partial}_i$, and $t, t_0 > 0$. Steady-state expectation values, $\langle g\rangle^{(\dot\gamma)} = C_{1g}(t, t_0 \to \infty)$, and variances, $\langle f^* g\rangle^{(\dot\gamma)} = C_{fg}(t = 0, t_0 \to \infty)$, then follow.

An important property of the sheared system is translational invariance [16]. It leads to spatially independent averages or, in Fourier space at wave vector $\mathbf{q}$, to $\langle f_{\mathbf{q}}\rangle^{(\dot\gamma)} = \langle f_0\rangle^{(\dot\gamma)} \delta_{\mathbf{q},0}$. In the two-time correlation functions of Eq. (2), it leads to a coupling of fluctuations of wave vector $\mathbf{q}$ with later fluctuations of the *advected wave vector* $\mathbf{q}(t) = \mathbf{q} + \mathbf{q}\boldsymbol{\kappa} t$, suggesting the definition $C_{f_{\mathbf{k}} g_{\mathbf{q}}}(t, t_0) = N C_{fg;\mathbf{q}}(t, t_0) \delta_{\mathbf{q}(t),\mathbf{k}}$. Figure 1 sketches the advection of a fluctuation with initial wavelength λ_x to one with λ_x and $\lambda_y(t) = \lambda_x/(\dot\gamma t)$ at later time t. Brownian particle motions (assisted by the interaction forces) "smear out" the fluctuation with time and cause the decay of the corresponding correlator. Because the wave number $q(t)$ increases upon shearing, smaller and smaller motions can cause the fluctuation to decay [16].

Equation (2) is an exact consequence of Eq. (1) (for the given shear flow); but to proceed further requires additional approximations. With our assumption that applied shear interacts with slow structural rearrangements, we build on the description achieved by the MCT, and analyze the approach into the steady state by monitoring the fluctuations of density ($\varrho_{\mathbf{q}} = \sum_{i=1}^N e^{i\mathbf{q}\mathbf{r}_i}$) and of the "pair density" (the square of the density in real space), aiming to establish nonlinear closed equations for these. This entails elimination of forces $\mathbf{F}_i$ in favor of the quiescent-state structure factor S_q (taken to be known) — a near-equilibrium assumption (see below) that is formally uncontrolled but motivated, at least in part, by the smallness of Pe_0.

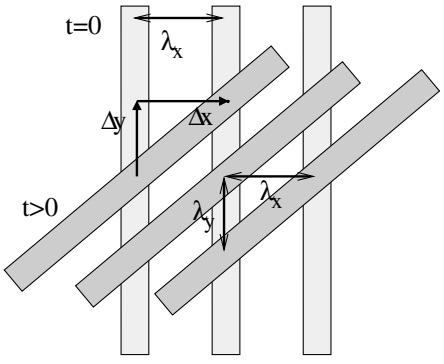

FIG. 1. Advection by steady shear of a fluctuation in the x direction with wavelength λ_x at $t = 0$. At later time t, its wavelength λ_y in the y direction obeys $\lambda_x/\lambda_y = \Delta x/\Delta y = \dot\gamma t$.

Steady state correlators are now approximated by projection onto the density modes, giving

$$\langle f^* g\rangle^{(\dot\gamma)} \approx \langle f^* g\rangle^{(0)} + \frac{\dot\gamma}{2} \int_0^\infty dt \sum_{\mathbf{k}} \frac{k_x k_y(t) S'_{k(t)}}{k(t) N S^2_{k(t)}} V^{fg}_{\mathbf{k}} \Phi^2_{\mathbf{k}}(t), \tag{3}$$

with t now the time since switch-on time, $S'_k = \partial S_k/\partial k$, and $V^{fg}_{\mathbf{k}}$ an easily found static overlap [17]. The transient density fluctuations are given by $\Phi_{\mathbf{q}}(t) = C_{\varrho\varrho;\mathbf{q}}(t, 0)/S_q$, and are normalized by S_q. They enter Eq. (3) via a factorization approximation of the density pair fluctuation functions. With the choice $f = g = \varrho_{\mathbf{q}}/\sqrt{N}$, Eq. (3) gives the steady-state structure factor under shear, whereas $f = 1$ and $g = \sigma_{xy}/V$ give the transverse stress, for which $V^{fg}_{\mathbf{k}} = (N k_x k_y/Vk) S'_k$.

The problem of calculating the steady-state averages is thus "reduced" to first finding the transient density fluctuations $\Phi_{\mathbf{q}}(t)$, given by the structural rearrangements after switching on the flow, and integrating these in Eq. (3). From Zwanzig-Mori (type) manipulations [18], one finds the exact equation of motion:

$$\dot\Phi_{\mathbf{q}}(t) + \Gamma_{\mathbf{q}}(t) \left\{ \Phi_{\mathbf{q}}(t) + \int_0^t dt' m_{\mathbf{q}}(t, t') \dot\Phi_{\mathbf{q}}(t') \right\} = 0, \tag{4}$$

where $\dot\Phi_{\mathbf{q}}(t) = \partial_t \Phi_{\mathbf{q}}(t)$, and the "initial decay rate" $\Gamma_{\mathbf{q}}(t)$ exhibits the familiar Taylor dispersion [14,19]. It is not known how to evaluate the microscopic expression for the memory function $m_{\mathbf{q}}(t, t')$ exactly. In the MCT spirit of our approach, this is approximated by projecting the fluctuating forces onto density pairs and factorizing the resulting pair-density correlation functions as

$$m_{\mathbf{q}}(t, t') \approx \frac{1}{2N} \sum_{\mathbf{k}} V^{(\dot\gamma)}_{\mathbf{q},\mathbf{k}}(t, t') \Phi_{\mathbf{k}}(t - t') \Phi_{\mathbf{q}-\mathbf{k}}(t - t'). \tag{5}$$

The vertex $V^{(\dot\gamma)}$, whose lengthy formula will be published elsewhere, is evaluated in the limit $\mathrm{Pe}_0 \ll 1$ (as argued above) but for large times so that $\dot\gamma t$ and $\dot\gamma t'$ are finite. As $\dot\gamma \to 0$, it reduces to the standard MCT vertex [9] and like the latter is uniquely determined by the equilibrium structure factor, S_q. For long times, it vanishes as $V^{(\dot\gamma)} \propto q_y q_x^{-3} \dot\gamma^{-3} t'^{-2} (t - t')^{-1}$ for $q_x \neq 0$.

Equations (3)–(5) complete our derivation of closed, self-consistent equations for the steady-state properties of dense sheared suspensions. They contain the bifurcation singularities which lie at the core of MCT. For $\dot\gamma = 0$, upon smooth changes of the input equilibrium state parameters, a fluid with $\Phi_q(t \to \infty) \to 0$ turns into an amorphous solid, $\Phi_q(t \to \infty) \to f_q > 0$. The f_q are called glass form factors and describe the arrested structure. While transport coefficients of the fluid, like the viscosity, are connected to the longest relaxation time of $\Phi_q(t)$, elastic constants of the solid, like the transverse elastic modulus G_∞, are given by the f_q [9].

In the limit of small shear rates, a stability analysis of the amorphous solid can be performed and leads to a generalization of the factorization theorem of MCT [20]. Close to the bifurcation, the dynamics are governed by $\Phi_q(t) = f_q^c + h_q\, \mathcal{G}(t)$, where the f_q^c describe the glassy structure at the instability and the critical amplitude h_q is connected to the cage-breaking particle motion; both retain their definition from the unsheared situation. Here $\mathcal{G}(t)$ contains the essential nonlinearities of the bifurcation dynamics which arise from the physical feedback mechanism (the cage effect). It depends on a few material parameters only and, for $|\mathcal{G}(t)| \ll 1$, follows from

$$\varepsilon - c^{(\dot\gamma)}(\dot\gamma t)^2 + \lambda \mathcal{G}^2(t) = \frac{d}{dt}\int_0^t dt' \mathcal{G}(t-t')\mathcal{G}(t'). \quad (6)$$

Here ε measures the distance to the transition and λ is known for some systems [9]. Monte Carlo estimations of the microscopic expression for the (new) parameter $c^{(\dot\gamma)}$ give $c^{(\dot\gamma)} \approx 3$ for hard spheres. Corrections of higher order in the small quantities (ε, $\dot\gamma$, $\mathcal{G}$) are neglected; see [21] for background on Eq. (6) for $\dot\gamma = 0$.

Equation (6) is our central result. As expected, the sign of $\dot\gamma$ does not enter, although it affects the Taylor dispersion. Because $(\dot\gamma t)^2$ dominates for long times, always $\mathcal{G}(t \to \infty) \to -t/\tau^{(\dot\gamma)}$, with $\tau^{(\dot\gamma)} = \sqrt{(\lambda - 1/2)/c^{(\dot\gamma)}}/|\dot\gamma|$. Hence, under flow, density fluctuations always decay, as this decrease of $\mathcal{G}(t)$ for long times initiates the final relaxation [where the corrections to Eq. (6) become important] of $\Phi_{\mathbf{q}}(t)$ to zero. Arbitrarily small shear rates $\dot\gamma$ melt the glass and so "interrupt" aging, as has also been found for spin glasses where shearing was mimicked by breaking detailed balance [11]. This vindicates our decision to ignore aging and to proceed via Eq. (3) in order to obtain steady-state properties.

While the non-Newtonian fluid behavior ($\varepsilon < 0$) includes two slow time scales, the familiar τ and the shear induced $\tau^{(\dot\gamma)}$, the rheology of glass ($\varepsilon \geq 0$) is determined by $\tau^{(\dot\gamma)}$ only [22]. For $\varepsilon \to 0+$ and $\dot\gamma = 0$, a fraction $f_q = f_q^c + h_q\sqrt{\varepsilon/(1-\lambda)}$ of the density fluctuations would stay arrested, while with shear these decay at a rate set by $\dot\gamma$: $\Phi_{\mathbf{q}}(t) \to f_q \Phi_{\mathbf{q}}^+(t/\tau^{(\dot\gamma)})$, where $\Phi_{\mathbf{q}}^+(x \to 0) - 1 \propto -x$, as follows from Eq. (6).

For $\varepsilon \geq 0$ and $\dot\gamma \to 0$, the time for the final decay can become arbitrarily slow compared to the time characterizing the decay onto f_q. Inserting $\Phi_{\mathbf{q}}^+(t/\tau^{(\dot\gamma)})$ into Eq. (3), the long time contributions separate out, and depend on time only via $\dot\gamma t$. Hence the glass has nontrivial shear-rate-independent limits for steady-state values: $\langle f^* g\rangle^{(\dot\gamma)} \to \langle f^* g\rangle^{(+)} \neq \langle f^* g\rangle^{(0)}$ for $\dot\gamma \to 0$. The $\langle f^* g\rangle^{(+)}$ are given by integrals over the $\Phi_{\mathbf{q}}^+(t/\tau^{(\dot\gamma)})$ and quantify those fluctuations that require the presence of shearing to avoid their arrest. For the case of the shear stress, a finite (dynamical) yield stress, $\sigma^+ = \lim_{\dot\gamma\to 0}\langle\sigma_{xy}\rangle^{(\dot\gamma)}$ for $\varepsilon \geq 0$, is thereby found. Since the glass transition is often identified by a divergence of viscosity (in terms of which we have shear thinning, $\eta(\dot\gamma \to 0) \propto \dot\gamma^{-m}$ with $m = 1$), our prediction of a finite yield stress throughout the glass phase is far from trivial. It excludes, e.g., power-law fluid behavior ($\sigma \sim \dot\gamma^{1-m}$ with $0 < m < 1$; see [10]).

These results follow from the general stability analysis of Eq. (6) and are predicted to be universal, i.e., to hold for the Brownian contribution to the shear stress and viscosity close to the glass transition in any colloidal dispersion. But because (in contrast to aging approaches [23]) we approximate nonlinear couplings under shear using equilibrium averages, we require the system to remain "close to equilibrium" in some sense. The existence of a finite yield stress σ^+ means that this is not guaranteed even as $\dot\gamma \to 0$.

Insight into an important mechanism of shear fluidization can be gained by considering transient density fluctuations with wave vector $\mathbf{q}$ perpendicular to the flow plane, $\mathbf{q} = q\hat{e}_z$. Here $V^{(\dot\gamma)}$ simplifies to the standard MCT vertex with advected (time-dependent) wave vectors [24]. While for $\dot\gamma = 0$ it exhibits the nonlinear coupling of density correlators with wavelength equal to the average particle distance, for $\dot\gamma \neq 0$, the (only) effect of shearing on this "neutral" direction consists of a shift of the advected wave vectors to higher values, where the effective potential decreases. This decreases the memory function and thus speeds up structural rearrangements. In this way the theory captures the faster decay of fluctuations caused by shear advection (cf. Fig. 1).

The presence of shear advection in the neutral z direction suggests an approximation that considers only the resulting competition of caging and advection-induced decay. In this "isotropically sheared hard sphere model" (ISHSM) [25], we neglect kinematic flow of particles so that all directions are treated as neutral. The quiescent S_q depends only on the packing fraction ϕ, and the model's glass transition lies at $\phi_c = 0.515\,91$ [21]. Figure 2 shows the stress versus strain rate curves for ϕ close to the transition. In the fluid, $\phi < \phi_c$, a Newtonian regime ($\sigma = \eta\dot\gamma$) is found for $\dot\gamma$ small enough that $\mathrm{Pe} < 1$. For $\mathrm{Pe} > 1$ there is a broad crossover to the critical yield stress value, $\sigma_c^+ = \sigma^+(\varepsilon \to 0+)$, from which σ starts to rise due to (nonuniversal) short-time effects for Pe_0 around 10^{-2} (where η_∞ will also contribute). In the glass, $\phi \geq \phi_c$, a yield stress plateau for $\dot\gamma \to 0$ is obtained, and rises strongly with increasing packing fraction. We speculate that the lack of a clear yield stress plateau at $\mathrm{Pe}_0 \geq 10^{-3}$ (and likely hydrodynamic effects) explains shear-thinning exponents $m < 1$ seen in experiments [7].

In summary, we have presented a microscopic theory of the nonlinear rheology of colloidal fluids and glasses under steady shear. It predicts a universal transition between shear-thinning fluid flow, with diverging viscosity upon increasing the interactions, and solid yielding, with a yield stress that is finite at (and beyond) the glass point. Besides its interest for dispersion flow, our work suggests a further role of colloidal systems in elucidating glasses via the study of shear-melted states for small shear rates.

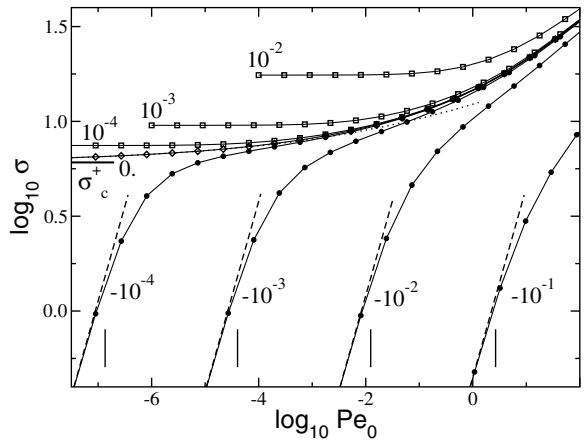

FIG. 2. Steady shear stress, $\sigma = \langle\sigma_{xy}\rangle^{(\dot\gamma)}$, in units of k_BT/d^3 versus $\mathrm{Pe}_0 = \dot\gamma d^2/D_0$, for a model of hard spheres [25] at various distances from its glass transition, $(\phi - \phi_c)$ as labeled. For the fluid cases, $\phi < \phi_c$, dashed lines indicate Newtonian fluid behavior, $\sigma = \eta\dot\gamma$, while vertical bars mark $\mathrm{Pe} = \dot\gamma\tau = 1$, with the structural relaxation time taken from $\Phi_{q=7/d}(t = \tau) = 0.1$. For the critical density, ϕ_c, the critical yield stress, $\sigma_c^+ = 6.0$, is shown by a horizontal bar, and the dotted line $\sigma = \sigma_c^+(1 + 1.0\dot\gamma^{0.17})$ matches for $\dot\gamma \to 0$.

This is of fundamental interest because, e.g., mean-field driven spin-glass theories predict nothing like a yield stress [11]. While comparison with measurements in colloids [6–8] and simulations of sheared atomic glasses [12,13] is promising, our approach represents only the first step toward rational prediction of the rheology of a glass; though physically motivated and in part inspired by the successful MCT description of the cage effect, several of our approximations remain incompletely justified. Nor is it clear whether Eqs. (4) and (5) can exhibit ''jamming'' transitions [26] at finite shear rates, or how anisotropic [27] the fluctuations can become. Extension to time-dependent shearing would be especially interesting because recent shear echo measurements [28] reveal intriguing glass-melting scenarios.

We thank J.-L. Barrat, J. Bergenholtz, L. Berthier, A. Latz, and G. Petekidis for discussions. M. F. was supported by the DFG, Grant No. Fu 309/3.

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