Suspension rheology at zero temperature

- *Shear flow of non-Brownian suspensions close to jamming*,
  B. Andreotti, J.-L. Barrat, C. Heussinger, arXiv:1112.1194.

- *A unified framework for non-Brownian suspension flows and soft amorphous solids*,
  E. Lerner, G. Düring, M. Wyart, PNAS (at press) arXiv:1112.0558.

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Suspensions of particles in a solvent are ubiquitous, with blood, paint and cement as primary examples of materials that are widely used throughout industry [1]. Perhaps the simplest model system for such materials would be an assembly of hard spherical particles immersed in a simple fluid, neglecting gravity. The rheological behaviour of this system in the dilute limit was considered by Einstein in 1906. He derived the following expression for the zero-shear viscosity: $\eta = \eta_s(1 + 2.5\varphi)$, where η_s is the viscosity of the solvent, and φ the fraction of the volume occupied by the particles. By contrast, the behaviour of the viscosity in the dense regime is not well understood. Experiments performed both for colloidal particles (in the sub-micron size range), and for non-Brownian particles (in the millimeter range) show that the shear viscosity grows very rapidly with increasing volume fraction, a behaviour which results from steric hindrance: It is hard to shear a dense suspension because particles have too little space to move past each other. This simple reasoning suggests that many-body correlations should appear at large φ and govern the microstructure and the dynamics in dense particle suspensions.

While statistical mechanics has largely treated these questions in the case of colloidal suspensions in the presence of thermal fluctuations, no such systematic approach is available to address the rheology of non-Brownian suspensions, for which the temperature is effectively zero. In the latter case, statistical mechanics tools are not obviously pertinent and most rheological models are in fact highly phenomenological [1]. Two recent numerical studies suggest that the problem is perhaps simpler than expected and provide *remarkable relations between the macroscopic rheology and the microstructure* of the suspension. These results thus pave the way for developing microscopic theoretical approaches to understand the rheology of dense suspensions at zero temperature.

Andreotti *et al.* use molecular dynamics to study the steady state rheology of dense assemblies of soft repulsive particles (such as Hertzian or elastic spheres) in the overdamped limit (no thermal noise). They use a simple flow geometry, where the imposed flow is along one direction, say $\mathbf{v}_{\text{flow}} = \dot{\gamma}y\mathbf{e_x}$, which defines the time independent shear rate $\dot{\gamma}$. Crucially, hydrodynamic flows and solvent-mediated interactions are not taken into account (and so the dilute limit cannot be described by Einstein's formula and is indeed not considered). They study the rheology of such assemblies, most notably the shear viscosity, as a function of the volume fraction, φ, and the shear rate, $\dot{\gamma}$.

As established by previous work for similar models [2], they find that when $\dot{\gamma} \to 0$ the dynamics converges to a well-defined limit that does not depend on the specific form of the dissipative force (and presumably not on the chosen interaction potential [3]). They also find that the shear viscosity is well-fitted to an algebraic divergence, $\eta/\eta_s \sim 1/(\varphi_j - \varphi)^\gamma$, where φ_j locates the jamming transition above which solid behaviour emerges [2], and $\gamma \approx 2.2$. This behaviour is similar to the one found experimentally for suspensions of rigid particles [4]. These results are physically reasonable because in the $\dot{\gamma} \to 0$ limit, pairs of particles that have been forced to overlap by the external flow have enough

time to move and relax towards a non-overlapping state. Therefore, the particle configurations that are visited typically contain no overlapping particles, and are thus similar to the states obtained in the hard sphere limit. In the latter case, simple dimensional analysis suggests indeed that the shear viscosity is a unique function of the packing fraction [2, 4]. An equivalent formulation of these results is that, in the hard sphere limit, dynamics is parametrized by the imposed deformation, $\gamma \equiv \dot{\gamma}t$, i.e. t and $\dot{\gamma}$ are not independent control parameters. As a consequence, the rheology of athermal suspensions can equivalently be studied using a variety of numerical methods where time does not play any role, such as quasi-static simulations for soft potentials [2] (employed by Andreotti *et al.*), or using event-driven simulations for hard particles (employed by Lerner *et al.*).

These macroscopic rheological results were already known but Andreotti *et al.* go further and directly relate the measured viscosity to a microscopic quantity. Using the fact that at zero temperature, viscous forces must balance contact forces, they use an energy balance argument and relate the shear viscosity to the distribution of single particle velocities: $\eta \propto \langle \delta v^2 \rangle$. Since time and deformation can be interchanged at small $\dot{\gamma}$, this can be rewritten in an even simpler form:

$$\eta(\varphi) \propto \lim_{t \to 0} \frac{R_2(t)}{\gamma^2} \equiv \ell_0^2, \tag{1}$$

where $R_2(t)$ is the non-affine mean-squared displacement, $R_2(t) = \langle N^{-1} \sum_i [\mathbf{r}_i(t) - \mathbf{r}_i(0)]^2 \rangle$ (the average flow being substracted), and ℓ_0 has the dimension of a length scale. This is a remarkable result because *it relates a macroscopic dynamical quantity to a microscopic, instantaneous observable*. This result is specific to the zero-temperature rheology and is not valid for (thermal) colloidal suspensions.

In a parallel effort, Lerner *et al.* use computer simulations to study the hard sphere limit. At the macroscopic level they obtain rheological results fully compatible with the ones of Andreotti *et al.*, and with earlier work [2]. However, they provide additional insight into the connection between microstructure and rheology, and thus they suggest an alternative microscopic understanding to Eq. (1). To this end, they treat the dense (fluid) suspension for $\varphi < \varphi_j$ on the same footing as the dense (solid) packing at $\varphi = \varphi_j$, which is mechanically stable. In the latter case, stability arises because particles have formed enough contacts with their neighbors to satisfy the condition of mechanical equilibrium [5]. This analogy suggests to express the force $\mathbf{F}_i$ experienced by particle i in terms of the contacts between neighboring particles only, namely, $\mathbf{F}_i = \sum_{ij} f_{ij} \mathbf{n}_{ij}$, where $\mathbf{n}_{ij}$ is the unit vector between particles i and j. Gathering forces $\mathbf{F}_i$ into a $3N$ dimensional vector $\mathbf{F}$, Lerner *et al.* rewrite the previous relation in a compact form, $\mathbf{F} = \mathcal{T}\mathbf{f}$, introducing the linear operator $\mathcal{T}$ acting on the vector $\mathbf{f}$ of contact forces [5]. Defining $\mathbf{\Gamma}$ as the vector of contact deformation per unit shear strain, and using as Andreotti *et al.* the force balance between viscous and contact forces, they obtain a compact expression for the shear viscosity:

$$\eta \propto \mathbf{\Gamma}^t (\mathcal{T}^t \mathcal{T})^{-1} \mathbf{\Gamma}. \tag{2}$$

Since it describes the same quantity (the shear viscosity), this equation is equivalent to Eq. (1) but *it provides an explicit microscopic expression for the particle displacements* in terms of the geometry of the contact network through the operator $\mathcal{N} \equiv \mathcal{T}^t \mathcal{T}$. By studying numerically the spectrum of $\mathcal{N}$, Lerner *et al.* discovered that it contains a narrow low-frequency band near a frequency ω_0 that seems to scale as a power law, $\omega_0 \sim (\varphi_j - \varphi)^\alpha$, and thus through Eq. (2) controls the divergence of the viscosity, $\eta \sim \omega_0^{-2} \sim 1/(\varphi_j - \varphi)^{2\alpha}$. Remark that the modes that are responsible for the divergence of η on the fluid side $\varphi < \varphi_j$ at $T = 0$ appear to be physically distinct from the soft modes discussed in the context of the un-jamming transition of soft particle systems observed for $\varphi > \varphi_j$ [6].

These results show that the dynamics of dense particle systems at strictly zero-temperature may appear simpler to address than dynamics in the presence of thermal fluctuations for which no connection such as Eqs. (1, 2) exists between instantaneous configurations and macroscopic rheology, which

adds to the ever-growing list of physical distinctions between thermal and athermal suspensions [7]. On the other hand, while statistical mechanics treatments of the microstructure of thermal suspensions are available, little is currently known from first principles about corresponding athermal systems which are mainly understood using computer studies, such as the ones described in this note. This represents a major theoretical challenge for future studies. Finally, the 'simplification' suggested by Eqs. (1, 2) is largely due to the hard sphere limit, and only holds when $\dot{\gamma} \to 0$ for soft particles [3]. Elucidating the finite shear rate behaviour for soft potentials at zero temperature is another important theoretical goal, since non-linear effects (such as shear-thinning or shear-thickening effects) should set in—and these rheological effects are experimentally extremely relevant for a broad variety of soft materials.

References

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