Discontinuous Shear Thickening in Dense non-Brownian Suspensions

1. "Microscopic Mechanism for Shear Thickening of Non-Brownian Suspensions", N Fernandez, R Mani, D Rinaldi, D Kadau, M Mosquet, H Lombois-Burger, J Cayer-Barrioz, H J. Herrmann, ND Spencer, and L Isa, arXiv:1308.1002; *Physical Review Letters* **111** (2013) 108301.

2. "Discontinuous Shear Thickening of Frictional Hard-Sphere Suspensions", R Seto, R Mari, JF Morriss, and MM Denn, arXiv:1306.5985; *Physical Review Letters* **111** (2013) 218301.

3. "Discontinuous shear thickening without inertia in dense non-Brownian suspensions", M Wyart and ME Cates, arXiv:1311.4099; *Physical Review Letters* **112** (2014) 098302.

4. "Shear thickening, frictionless and frictional rheologies", R Mari, R Seto, JF Morris, and MM Denn, arxiv:1403.6793v2 (2014).

Recommended, with a commentary by Peter Olmsted, Department of Physics, Georgetown University; April 2014.

Dense colloidal or particulate suspensions are ubiquitous; they can be found in foods (e.g. emulsions), paints, personal care products such as toothpastes, muds, slurries, cements, and many other materials. Cornstarch and water is a favorite example [1]. They often display dramatic flow behaviour such as shear thinning, fracture, shear thickening, development of normal stresses, and spatial structuring. One of the most common features is shear thickening at high volume fractions. In many non-Brownian and Brownian suspensions this shear thickening can occur as almost a step function of apparent viscosity as a function of shear rate, and has been termed Discontinuous Shear Thickening (DST), as opposed to Continuous Shear Thickening.

The simplest example is solvated particles interacting by short range (and approximately hard sphere) repulsive interaction. At low volume fractions the interactions are predominantly hydrodynamic, and well described by the Stokes formulation, valid in zero Reynolds number, creeping flow, limit, where $Re = \rho V R/\eta$ is the Reynolds number, V is the characteristic velocity, R the particle radius, and η the viscosity. In concentrated suspensions the Stokes interaction is simple, albeit computationally time consuming to calculate. At close contact the Stokes interaction gives rise to singular hydrodynamic lubrication interactions $F_n \simeq \eta v R^2/h$ (normal force) and $F_{\parallel} \simeq \eta v \ln(R/h)$ (tangential force), which in principle ensures that particles never touch [2]. This leads to so-called 'hydroclusters', in which particles remain in close proximity due to the enhanced local drag due to lubrication, but the singular force ensures that particles never touch and has not yet been capable of describing DST [3–5]

Hence one needs to go beyond simple Stokesian dynamics. There has been a flurry of recent work that incorporates ideas from other related fields, namely tribology (where the interplay between solid and fluid friction is important) and granular media (non-solvated particulate flows). The key important ingredient that has emerged is velocity independent solid-like friction between particles that come into contact. This is of course of paramount importance in granular media, but has often been considered 'heretical' in the face of the singular force presented by Stokes lubrication. However, a large laundry list of reasons suggest that contact, and thus friction, should play a role: (1) at high enough packings particles will unavoidably make contact just to fill space; (2) the Stokes approximation will break down at short times (and distances) set by inertial forces (the physics that gives rise to boundary layers); (3) thermal fluctuations (and associated inertia) similarly blur the singular response; (4) asperities and particle roughness can lead to local non-laminar flows and separations of streamlines during strong confinement; (5) asperities and/or chemical surface heterogeneity can lead to wall slip, which will soften the singularity and allow contact; (6) for sufficiently high interparticle pressures and appropriate wetting properties the lubricating layer can rupture by a thermal fluctuation, leading to contact; (7) rupture or deformation of stabilizing grafted polymer layers could lead to effective slip and aid the transition to contact.

This set of four papers brought together ideas from various fields to shed light on the problem, and a paradigm is emerging: at high shear rates the higher stresses are strong enough to break down the hydrodynamic lubrication 'barrier' against contact. This implies two different jamming volume fractions ϕ_J (Fig. 1): a jamming ϕ_{J_0} due to separated lubricated hard spheres, and a smaller jamming fraction $\phi_{J,f}$ due to the appearance of friction contacts, which allow for a percolating network at lower volume fraction. Depending on whether or not friction is present, one expects a divergent viscosity $\eta(\phi) \sim (\phi_J - \phi)^{\alpha}$, where $\alpha \simeq 2$. DST emerges as a crossover from the lubricated viscosity branch to the friction-dominated branch, since at a give volume fraction the friction-dominated branch has a higher stress, being closer to its effective jamming point. The key point in all of these works is that the application of flow, and thus stress, breaks down the lubricating layer and leads to DST.

Fernandez et al. (2013) showed this by appealing results form tribology, in which a crossover from hydrodynamic friction to 'boundary lubrication', in which solid-like lubrication applies. This is apparent in the 'Stribeck' curve of the friction coefficient between two lubricated plates (Fig. 3) as function of sliding rate, which reveals a crossover between

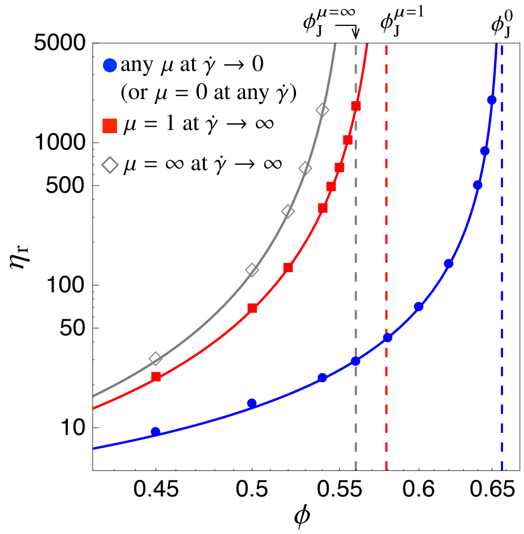

FIG. 1. Divergent viscosities $\eta_r \equiv \eta\phi$ for frictionless (ϕ_J^0) and frictional (ϕ_J^μ) suspensions. DST is posited to occur when flow generates enough stress to overcome the lubrication barrier to friction, and suddenly induce the higher viscosity behavior. The different papers identify different ways of interpolating between these rheologies to induce DST.

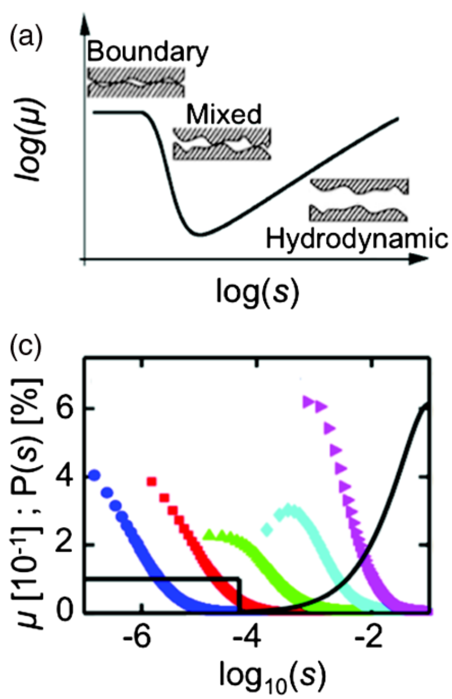

FIG. 2. Schematic and simulated Stribeck curves of the friction coefficient μ as a function of the Sommerfeld number $s \equiv \eta RV/F_n$, showing the crossover from solid-like to fluid-like lubrication. (From Fernandez et al. 2013). Fernandez et al. show that the physics embodied in this result from lubrication science can manifest itself in DST in dense suspensions.

these regimes. They performed experiments on suspensions, prepared surfaces to measure and control frictional properties, and backed this up with simulations in which the sliding lubrication law is replaced by load-independent Amontons-Coulomb friction. Their work suggested that DST occurs for $\phi_{J,f} < \phi < \phi_{J,0}$, while CST should occur for $\phi < \phi_{J,f}$.

At roughly the same time, Seto et al. (2013) performed simulations in which they also regularized the lubrication singularity by introducing a small scale cutoff length δ [6], leading to $F_n \simeq \eta v R^2/(h+\delta)$ and $F_\parallel \simeq \eta v \ln(R/(h+\delta)$. They allowed force-dependent sliding friction, $F_\parallel \leq \mu(F_n - F_{slide})$, where F_{slide} is a characteristic force; and slightly softened the hard core repulsion to allow for a shear-rate dependence. The simulations revealed an intriguing network of particle contacts in the DST-induced thickened state, which is induced at a stress characteristic of that force necessary to initiate friction, and they find a 'phase diagram' for DST as a function of shear rate and particle volume fraction. This was very recently followed up with a comprehensive manuscript (Mari et al. 2014) which included more spatial information about the DST/CST crossover, including structure factors and the morphology of the DST-induced contact network. This paper made a link to the calculations of Wyart and Cates, by extracting the fraction of frictional contacts a a function of stress, which shows a remarkable scaling for different volume fractions.

Motivated by this growing evidence, Wyart and Cates (2014) devised a phenomenological model for DST. They

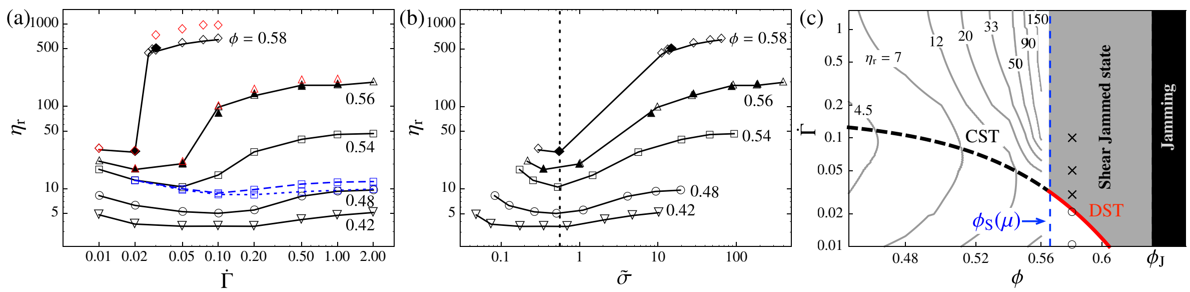

FIG. 3. (a) Apparent viscosity η_r as a function of shear rate $\dot{\Gamma}$, showing DST at volume fraction $\phi = 0.58$. (b) Apparent viscosity data showing that the stress σ at which thickening occurs is governed by the stress $\sigma_0 = F_{slide}/R^2$ that controls sliding friction (here, $\tilde{\sigma}\sigma/\sigma_0$). (c) DST-CST state diagram as a function of shear rate $\dot{\Gamma}$ and volume fraction ϕ. (From Seto et al. 2013).

assume that frictional contacts are active above a jamming volume fraction ϕ_m, which is determined by the stress needed to overcome the lubrication force and initiate the contact network. They used the insights of Boyer et al. [7], who showed that the shear stress σ and pressure P in a suspension obey the scaling relation $\sigma = \mu(I_v)P$, where $I_v = \eta\dot{\gamma}/P$ is the ratio of the viscous rearrangment time to the shear time. With this in hand, they argued that the *effective* jamming volume fraction, at a given fixed volume fraction, changes with shear rate as the stress induces lubrication layers to break down and initiate frictional contacts. By making suitable simple smooth assumptions for the dependence of ϕ_J on the fraction of frictional contacts, and for the dependence of frictional contacts on pressure, they obtained non-monotonic S-shaped constitutive relation between stress and shear rate is possible even for $\phi_c < \phi < \phi_{J,f}$ (Fig. 4). For example, if ALL frictional contacts immediately broke at a given shear rate, when the stress was a given critial value, then the the material would jump to the higher viscosity branch (Fig. 1). At a given stress this would lead to a lower shear rate, but if the shear rate was instead controlled, then the stress would have to increase discontinuously until the new flow branch was reached. Wyart and Cates propose models to soften this abrupt relation, which leads to the behaviours shown in Fig. 4.

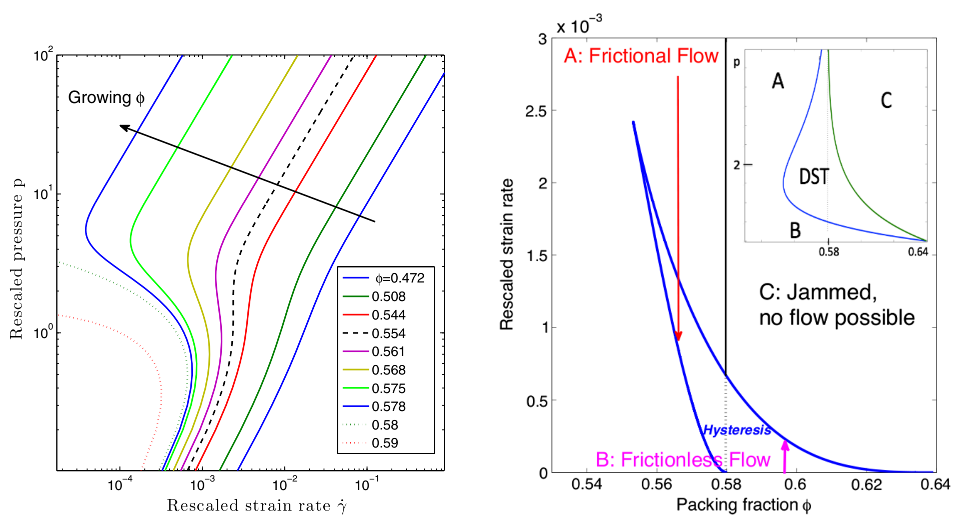

FIG. 4. (left) Non-monotonic constitutive curves derived by Wyart and Cates; the appearance of an S-curve heralds the possibility of DST. (right) State diagram showing the critical point $\phi_c \simeq 0.554$, below which only CST is predicted. DST is predicted for $\phi_c < \phi < \phi_{J,f} = 0.58$.

Of course, there remains much to do to understand DST. A conservative wish list includes understanding the spatial structure of contact networks and resulting stress, developing a more faithful understanding of the different kinds of microscopic phenomena that can lead to S-shaped constitutive curves, disentangling the correct physical descriptions of the frictional force laws, incorporating slightly attractive or non-spherical particles, and applying this knowledge to industrial contexts and to inhomogenous flows and shear banding.

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