Kinetic Theory of Plastic Flow in Soft Glassy Materials

Lydéric Bocquet,[1,*] Annie Colin,[2] and Armand Ajdari[3]

[1]*Laboratoire PMCN, Université Lyon 1, Université de Lyon, UMR CNRS 5586, 69622 Villeurbanne, France*
[2]*LOF, Université Bordeaux 1, UMR CNRS-Rhodia-Bordeaux 1 5258, 33608 Pessac cedex, France*
[3]*UMR Gulliver CNRS-ESPCI 7083, 10 rue Vauquelin, 75231 Paris Cedex 05, France*

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A kinetic model for the elastoplastic dynamics of a jammed material is proposed, which takes the form of a nonlocal—Boltzmann-like—kinetic equation for the stress distribution function. Coarse graining this equation yields a nonlocal constitutive law for the flow, exhibiting as a key dynamic quantity the local rate of plastic events. This quantity, interpreted as a local fluidity, is spatially correlated with a correlation length diverging in the quasistatic limit, i.e., close to yielding. In line with recent experimental and numerical observations, we predict finite size effects in the flow behavior, as well as the absence of an intrinsic local flow curve.

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Soft amorphous materials such as foams, emulsions, granular systems, or colloidal suspensions display at high enough concentrations complex flow properties, intermediate between that of a solid and a liquid: at rest they behave like an elastic solid, but flow ''like a liquid'' under sufficient applied stress [1–4]. This mixed fluid-solid behavior occurs above a threshold volume fraction associated with the appearance of a yield stress σ_d. The yielding behavior makes such systems particularly interesting for applications—from tooth paste to coatings to cosmetic and food emulsions—but fundamentally difficult to describe [5–7]. Furthermore, it has been recognized in recent years that this yielding behavior is, in most cases, associated with peculiar *spatial* features. These can take the form of inhomogeneous flow patterns, such as shear bands [1–3,8], or cooperativity in the flow or deformation response [9–16], potentially associated with nonlocality in the constitutive rheological law [11] and dependence of the flow on the nature of the boundaries [4,17]. While such features appear to be generic for this class of materials, suggesting an underlying common flow scenario, a consistent framework linking the global rheology to the local microscopic dynamics is still lacking.

In this Letter, we present a kinetic elastoplastic (KEP) model, which aims at constructing such a link between the microscopic and the macroscopic scales. Starting from a kinetic elastoplastic description of the dynamics, we derive systematically a (nonlocal) generic constitutive law for the flow, obtained by coarse graining the microscopic spatiotemporal dynamics. The predictions of our KEP model will be shown to capture many features of the rheology of yield stress fluids, and, in particular, the recent experimental demonstration of cooperativity in the flow behavior of jammed emulsions [11].

The KEP model, which is detailed below, is based on a generic picture which has emerged recently for the dynamics of soft glassy materials [14,18,19]. In these materials, flow occurs through a succession of global elastic deformations and *localized* plastic rearrangements associated with a microscopic yield stress; see Fig. 1. These localized events induce long-range elastic modifications of the stress over the system, thereby creating long-lived fragile zones where flow occurs. Flow in these systems is thus highly cooperative and spatially heterogeneous: a dynamically active region will induce stress fluctuations of its neighbors and thus a locally higher rate of plastic rearrangements. Correlations between plastic events are accordingly expected to exhibit a complex spatiotemporal pattern [14].

The Letter is organized as follows. (i) We first formulate the ''microscopic'' equations constituting our KEP model on the basis of the above generic scenario, Eqs. (1)–(3), (ii) then we derive the continuum *hydrodynamic* limit of these microscopic equations, and (iii) we finally deduce the constitutive nonlocal flow rules for plastic flow, as summarized in Eqs. (8). The ''micro-macro'' derivation of these constitutive rheological equations is the central result of this work. As a key point—and beyond simple symmetry expectations—nonlocality applies on a dynamical order parameter, the fluidity, defined here as the rate of plastic events.

The KEP model.—Describing the complex dynamical elastoplastic processes sketched in Fig. 1 is a formidable task. Therefore, to get further insight, we propose a sche-

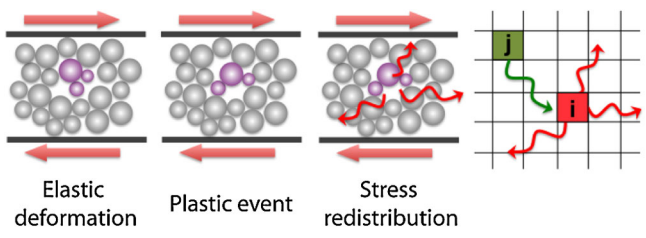

FIG. 1 (color online). Sketch of plastic deformation in amorphous media. Deformation occurs via elastic deformation, localized plastic events, and nonlocal redistribution of the elastic stress, potentially triggering other plastic events.

matic model, relying on a few simplifying assumptions. Our KEP model extends on an approach first proposed by Hébraud and Lequeux (HL) [6], by describing explicitly spatial interactions between plastic events: the sample is divided into elementary blocks $\{i\}$ of size a (typically the size of individual particles), each carrying a scalar shear stress σ_i [20], and the system is stochastically described in terms of the block stress distribution $P_i(\sigma, t)$. In this first approach, relative motion of the blocks (convective shear flow) is neglected. The distribution $P_i(\sigma, t)$ evolves via three mechanisms: an *elastic response*, under an externally imposed shear rate $\dot{\gamma}_i^o$, a stress relaxation due to *local plastic events*, and the modification of stress due to the plastic events *occurring in other blocks*, via the generated long-ranged elastic field. The corresponding mechanisms are sketched in Fig. 1. The local plastic events will be assumed to occur above a local threshold value of the stress σ_c, and lead to a complete relaxation of the local stress σ_i ($\sigma_i \rightarrow \sigma_i = 0$) on a time scale τ. These processes translate into the following master equation for the local stress distribution function P_i:

$$\partial_t P_i(\sigma, t) = -G_o \dot{\gamma}_i^o \partial_\sigma P_i(\sigma, t) - \frac{\Theta(|\sigma| - \sigma_c)}{\tau} P_i(\sigma, t) + \Gamma_i(t)\delta(\sigma) + \mathcal{L}(P, P), \quad (1)$$

with $\dot{\gamma}_i^o$ the imposed shear rate, G_o is the elastic modulus, Θ the Heaviside function; the rate of plastic events, $\Gamma_i(t)$, is fixed by

$$\Gamma_i(t) = \int \frac{\Theta(|\sigma'| - \sigma_c)}{\tau} P_i(\sigma', t) d\sigma'. \quad (2)$$

In the above equation, Eq. (1), the operator formally written as $\mathcal{L}(P, P)$ accounts for the stress modification induced by events occurring in other blocks j ($\neq i$). Indeed the occurrence of a localized plastic event in a block j induces a *nonlocal* stress relaxation at any block $i \neq j$ due to the elastic field generated. This can be written in the form $\delta\sigma_i = \Pi_{i,j}\delta\sigma_j$ where $\delta\sigma_j$ is the relaxed stress at block j and $\Pi_{i,j}$ is the elastic stress propagator [14]. Explicit forms for this (algebraically decaying) propagator were discussed in Ref. [14].

To express $\mathcal{L}(P, P)$, one should thus describe the gain and loss contributions for the probability $P_i(\sigma, t)$ due to events occurring in other blocks. Here we propose a formal analogy with the Boltzmann equation to construct the ''collision'' operator $\mathcal{L}(P, P)$. This gain and loss balance thus takes the form

$$\mathcal{L}(P, P) = \sum_{j \neq i} \int d\sigma' \frac{\Theta(|\sigma'| - \sigma_c)}{\tau} \times [P_j(\sigma', t) P_i(\sigma + \delta\sigma_i, t) - P_j(\sigma') P_i(\sigma)], \quad (3)$$

with $\delta\sigma_i = \Pi_{i,j}\delta\sigma_j = -\Pi_{i,j}\sigma'$ (using the full stress relaxation rule $\delta\sigma_j = -\sigma'$). Note that, in the same spirit as the Boltzmann Stosszahlansatz, this expression assumes a decoupling of the plastic-event dynamics.

Finally, inserting this expression for $\mathcal{L}(P, P)$ in Eq. (1) provides a closed kinetic equation for the nonlocal elasto-plastic dynamics. This KEP equation is the first key result of this work.

Towards continuum equations.—In its above form, the KEP master equation remains difficult to solve analytically. To proceed further, we first approximate the above master equation, i.e., the Boltzmann operator $\mathcal{L}(P, P)$, using a Kramers-Moyal expansion [21]. This is done by formally expanding the Boltzmann operator for small stress variations $\delta\sigma_i$ and retaining only the first terms of the expansion. The further simplification $\delta\sigma_j \approx -\sigma_c$ is also made in the integrand of the operator [for small $\dot{\gamma}$ the distribution $\Theta(|\sigma| - \sigma_c)P_i(\sigma, t)$ is peaked around σ_c]. Altogether, this simplifies Eq. (1) to a Fokker-Planck equation:

$$\partial_t P_i(\sigma, t) = -G_o \dot{\gamma}_i \partial_\sigma P_i(\sigma, t) - \frac{\Theta(|\sigma| - \sigma_c)}{\tau} P_i(\sigma, t) + \Gamma_i(t)\delta(\sigma) + D_i \partial^2_{\sigma^2} P_i(\sigma, t). \quad (4)$$

In this equation, $\dot{\gamma}_i$ is the local shear rate ($\dot{\gamma}_i = \dot{\gamma}_i^o + \frac{1}{2}\sum_{j \neq i} \Pi_{ij}\sigma_c\Gamma_j$) and the coefficient D_i quantifies what appears as a *stress diffusion* induced by the occurrence of plastic events. Its expression derives directly from the Kramers-Moyal expansion as

$$D_i(t) = \frac{1}{2}\sum_{j \neq i} \Pi_{ij}^2 \sigma_c^2 \Gamma_j(t). \quad (5)$$

This key result shows that stress diffusion D_i is related to the rate of plastic events over the whole system, Γ_j ($j \neq i$).

Switching to continuous spatial variables, a closed system of equations is obtained for the local stress diffusion $D(\mathbf{r}, t)$, rate of plastic events $\Gamma(\mathbf{r}, t)$, and stress distribution $P(\sigma, \mathbf{r}, t)$. Equation (4) keeps the same form (with $i \rightarrow \mathbf{r}$). A small slope approximation of the self-consistency equation for D (valid in the *hydrodynamic* limit where Γ varies on a length scale much larger than a) provides a nonlocal relationship between stress diffusion and rate of plastic events:

$$D(\mathbf{r}, t) = m\Delta\Gamma(\mathbf{r}, t) + \alpha\Gamma(\mathbf{r}, t), \quad (6)$$

with Δ the spatial Laplacian. In this equation, two key parameters have been introduced: a *coupling parameter* α, here defined as $\alpha = \sigma_c^2 \sum_{i \neq j} \Pi_{i,j}^2$, and an inhomogeneity parameter $m = a^2\sigma_c^2\Pi_{nn}^2$, with Π_{nn} the nearest neighbor (block-to-block) propagator and a the block elementary size. In the following we will make use of dimensionless variables, $\tilde{t} = t/\tau$, $\tilde{r} = r/a$, $\tilde{\sigma} = \sigma/\sigma_c$, $\tilde{\dot{\gamma}} = \dot{\gamma}G_o\tau/\sigma_c$, $\tilde{m} = m/a^2\sigma_c^2$, $\tilde{\alpha} = \alpha/\sigma_c^2$, $\tilde{\Gamma} = \Gamma\tau$, and $\tilde{D} = D\tau/\sigma_c^2$, but will drop the tilde to simplify notations.

Jamming and yield stress.—Let us first discuss briefly the limiting case $m = 0$ [in Eq. (6)] where heterogeneities

are omitted. Under this assumption, the above set of equations reduce *exactly* to the HL description in Ref. [6]. A key result which emerges from the HL description is that it predicts a *jamming transition* below a threshold (dimensionless) coupling parameter $\alpha < \alpha_c = \frac{1}{2}$, associated with the building up of a macroscopic *dynamic yield stress*, $\sigma(\dot{\gamma} \to 0) = \sigma_d$. As shown in Ref. [6], the dependency of σ_d on the coupling parameter α takes the form $\sigma_d \propto (\alpha_c - \alpha)^\beta$, with $\beta = 1/2$. This dynamic yield stress σ_d, which is smaller than σ_c, thus quantifies the distance to the jamming transition [22]. In the following we shall focus on the jammed state, as defined by a nonvanishing σ_d.

Nonlocal constitutive flow rules.—We now come back to the inhomogeneous case [$m \neq 0$ in Eq. (6)] and discuss the solution of the model in the stationary state ($\partial_t = 0$). In this case, the stationary Fokker-Planck equation, Eq. (4), can be solved analytically [6]. The solution is straightforward and gives an explicit expression for $P(\sigma, \mathbf{r})$ as a sum of exponentials in σ. One then deduces the local averaged stress $\bar{\sigma}(\mathbf{r}) = \int d\sigma' \sigma' P(\sigma', \mathbf{r})$ and the local rate of plastic events $\Gamma(\mathbf{r})$ as defined in Eq. (2). This provides explicit expressions for these quantities in terms of the diffusion coefficient $D(\mathbf{r})$ and local shear rate $\dot{\gamma}(\mathbf{r})$. Their general expression only involves elementary functions but is rather cumbersome, and we do not report it here. However, they simplify considerably in the limit of slow flow ($\dot{\gamma} \to 0$) and close to the jamming point, i.e., small σ_d. Choosing the plastic rate Γ as the key variable, a systematic expansion of the expressions of $\bar{\sigma}$ and D in this regime yields the following expressions:

$$\begin{aligned} \bar{\sigma} &= (6\Gamma)^{-1}\dot{\gamma}, \\ D - \alpha\Gamma &= a_1\sigma_d(\sigma_d - \bar{\sigma})\Gamma + a_2\Gamma^{3/2} + O(\Gamma^2), \end{aligned} \tag{7}$$

with σ_d the dynamic yield stress introduced above, and a_1, a_2 two numerical constants [22]. In the following we shall define $f = 6\Gamma$ as the *fluidity*: the latter naturally emerges as intimately linked to the rate of plastic events.

Together with the self-consistency relationship Eq. (6), relating D to Γ, these expressions provide a closed set of equations. We rewrite here this set in the physically meaningful form:

$$\bar{\sigma} = \frac{1}{f}\dot{\gamma}, \qquad \Delta f - \frac{1}{\xi^2}(f - f_b) = 0. \tag{8}$$

In this equation we have introduced a *bulk fluidity* $f_b(\bar{\sigma})$: $f_b(\bar{\sigma}) = 6(\frac{a_1\sigma_d}{a_2})^2(\bar{\sigma} - \sigma_d)^2$ for $\bar{\sigma} > \sigma_d$ and 0 otherwise. A fluidity *correlation length* $\xi(\bar{\sigma})$ also naturally emerges from the derivation and takes the expression

$$\xi = \sqrt{\frac{m}{a_1|\bar{\sigma} - \sigma_d|}} \tag{9}$$

for $\sigma > \sigma_d$, and with a prefactor $2^{1/2}$ for $\sigma < \sigma_d$. These coupled equations constitute the *nonlocal constitutive flow rules* which emerge from the KEP model, and are the central result of this work.

The bulk fluidity $f_b(\bar{\sigma})$ is the value of the fluidity obtained in absence of nonlocal terms, as obtained in the HL model: as can be easily verified, the KEP model predicts a Herschel-Bulkley expression for the flow rule for low shear rates, with σ_d as the dynamic yield stress: $\bar{\sigma}(\dot{\gamma}) = \sigma_d + A\dot{\gamma}^n$, with $n = 1/2$ and A a constant depending on α.

Flow cooperativity.—A key result of the derived flow behavior, in Eq. (8), is the *nonlocal nature* of the flow curve, which introduces a *flow cooperativity length* ξ. We emphasize that this nonlocal flow rule is formally identical to the phenomenological cooperative rheology introduced recently to account for the flow of confined jammed emulsions in microchannels [11]. In these experiments, the flow profiles were found to *strongly depart* from the bulk prediction for confinements typically smaller than a few tens of droplet diameters. A constitutive law similar to Eq. (8) was able to rationalize all experimental results, with a cooperativity length scale of the order of several droplets diameters. The present derivation starting from a microscopic point of view gives strong support to this phenomenological framework.

Physically, ξ quantifies the spatial spreading of the plastic activity due to the nonlocal elastic relaxation over the system. Interestingly, the correlation length diverges at the dynamical yield stress according to $\xi \propto |\sigma - \sigma_d|^{-1/2} \propto \dot{\gamma}^{-1/4}$. Such a power-law divergence is in agreement with recent numerical simulations [12,14,23] and prediction for slip avalanches in the deformation of solids [24]. Experimentally no dependence of the cooperativity length on shear rate was reported in Ref. [11]. However, the flow behavior in the $\dot{\gamma} \to 0$ limit is difficult to access experimentally and would certainly require further specific investigation. Finally, let us quote that similar cooperativity effects are reported in granular flows [12,15,16], as well as in numerical simulations of deformation of amorphous materials [13,18].

As discussed in Ref. [11], nonlocal effects in the flow curve induce strong departures from the "bulk" prediction (i.e., without nonlocal effects), as soon as the characteristic length reaches a fraction of the confinement. Such confinement effects are due to fluidity gradients that may find their origin either in the existence of stress gradients (such as in Poiseuille flow) or in boundary effects. Boundary effects have accordingly a strong influence on the flow, since Eq. (8) requires the prescription of the fluidity at the confining walls. The latter is expected to depend on surface properties, e.g., roughness: a smooth wall is indeed expected to induce a smaller wall fluidity as compared to a rough wall, which in turn will modify the shape of the flow profile in the material. The influence of boundary roughness on the flow is indeed observed experimentally in various systems [4,11,17] and would definitely deserve a

more systematic investigation. Finally, another consequence of the nonlocal terms in the flow curve, Eq. (8), is that the spatial spreading of the fluidity ''soften'' the flow velocity profiles: a finite fluidity, and a nonvanishing shear rate $\dot{\gamma}$, extend to regions where the stress is *below* the yield stress σ_d. In other words, cooperativity may be seen as suppressing the apparent yield stress of the material and confined flow can accordingly occur below the yield stress, in agreement with experiments [11].

A dynamical phase transition.—At a more formal level, it is interesting to note that the solution of Eq. (6) is the minimum of the square gradient ''free energy'':

$$\Omega(\Gamma) = \int d\mathbf{r} \frac{m}{2} (\nabla\Gamma)^2 + \omega(\Gamma, \bar{\sigma}), \tag{10}$$

with

$$\omega(\Gamma, \bar{\sigma}) = \frac{1}{2} a_1 \sigma_d (\sigma_d - \bar{\sigma}) \Gamma^2 + \frac{2}{5} a_2 \Gamma^{5/2} + O(\Gamma^4) \tag{11}$$

in the limit of small Γ. This equation is analogous to a Landau expansion close to a *second order phase transition*, with the dynamic yield stress σ_d as a critical point. The rate of plastic events Γ, i.e., the fluidity, plays the role of the (dynamic) order parameter, with stationary homogeneous solution: $\Gamma_b(\sigma) = 0$ for $\sigma < \sigma_d$ and $\Gamma_b(\sigma) \propto (\sigma - \sigma_d)^2$ for $\sigma > \sigma_d$.

Beyond the formal analogy, this suggests an interesting alternative point of view for flow inhomogeneities. While the present scenario predicts flow inhomogeneities characterized by a cooperativity length scale, in line with experimental results for dense emulsions, a ''true'' shear banding would merely correspond to a *first order phase transition* scenario: i.e., the spatial coexistence between two states of different fluidity for the same shear stress. Recent experimental findings have connected shear banding to the existence of attractive interactions between particles, thereby inducing a flow-structure coupling in the material [25]. The KEP description does not account for these features, and it would be therefore interesting to include local structure variables in the description in order to capture such couplings.

Conclusions.—In conclusion, we have derived a nonlocal constitutive equation for the flow of jammed systems starting from a ''microscopic'' kinetic elastoplastic model. The resulting description suggests the cooperative nature of the flow, in full agreement with recent experimental findings [11]. Furthermore, this framework puts forward the role of the fluidity as a dynamical order parameter characterizing the flow, and here defined as the local rate of plastic events in the material. Since one expects plastic events to trigger local velocity fluctuations, characterized by a rms velocity $\langle \delta v^2 \rangle^{1/2}$, the latter quantity could provide an indirect measure of the fluidity, in line with granular hydrodynamics approaches [26]. It is interesting to note that similar observations of nonlocality have been reported in granular flows close to the jamming transition [15,16], suggesting further universal characteristics.

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*Corresponding author.
lyderic.bocquet@univ-lyon1.fr

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