Rearrangements and Dilatancy for Sheared Dense Materials

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Constitutive equations are proposed for dense materials, based on the identification of two types of free-volume activated rearrangements associated with shear and compaction. Two situations are studied: the case of an amorphous solid in a stress-strain test, and the case of a lubricant in tribology test. Varying parameters, strain softening, shear thinning, and stick-slip motion can be observed.

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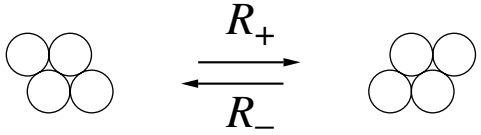
From food to beauty products, from our bone joints to the gouge of tectonic faults, the dynamical properties of dense materials determine important and ubiquitous phenomena that govern our life. The idea emerged recently that some sort of universality might be at work in structural systems, ranging from glasses to granular materials [1]. This assumption infers that a main structure of constitutive equations should hold for a wide class of dense materials and should not depend on the details of interactions between their microscopic constituents. In this Letter, I hypothesize that universality results from the crucial role played by structural rearrangements. I show how constitutive equations derive from the identification of mesoscopic observables and excluded-volume effects.

The approach I propose relies on the so-called shear transformation zone (STZ) theory, introduced to study elastoplastic transitions in amorphous solids [2]. STZ theory provides a general scheme for rearrangement kinetics in a dense material: macroscopic deformation results from local free-volume activated rearrangements at a mesoscopic scale; local states are introduced, related to orientations of the contact network; mean-field equations of motion for those local states lead to macroscopic constitutive equations. STZ theory was originally designed for solids, but it has been shown recently that it could successfully account for the rheology of granular flows [3]. It is shown here that supplementing STZ theory with free-volume kinetics permits one to account for important features of both amorphous solids (strain softening) and confined liquids (stick slip).

In previous works on STZ theory, free volume appears as a parameter. It is, however, a state variable, related to the particle density ρ of the material as $v_f = 1/\rho - 1/\rho_{\mathrm{rcp}}$, where ρ_{rcp} is the density of random closed packing. Free-volume kinetics is inspired by recent works on granular materials submitted to vertical tapping, in which local rearrangements are shown to account for logarithmic density relaxation [4]. The microstructural picture I propose for dense materials thus involves two types of rearrangements associated, respectively, with shear motion and to compaction/dilatancy. Macroscopic deformation is controlled by the interplay between those two mesoscopic processes and associated time scales. To emphasize the analogy between solids and confined liquids, two important experimental tests are considered: a stress-strain test performed on a plastic material [5] and an overdamped tribology test where a lubricant or a granular material is sheared [6–8]. In the former case, the resulting constitutive equations are shown to account for strain softening; in the latter, they account for a transition between steady sliding and stick-slip motion. The picture is completed with the study of constant stress experiments (creep tests).

Let me now present a scheme for rearrangement kinetics, along the lines of STZ theory [2]. The present approach remains at a mean-field level. The overall shear deformation is denoted ϵ and decomposed into an elastic and a plastic part: $\epsilon = \epsilon^{\mathrm{el}} + \epsilon^{\mathrm{pl}}$. The elastic deformation is proportional to the shear stress $\sigma = \mu\epsilon^{\mathrm{el}}$; the plastic deformation results from local, irreversible transformations of the contact network. A local rearrangement involves several molecules at a mesoscopic level. A shear transformation zone is defined as a locus within the material where an elementary shear is made possible by the local conformation of neighboring molecules. An essential remark that lies at the root of STZ theory is that, once a rearrangement has occurred, some contacts break, some others are formed, and the molecules involved cannot shear further in the same "direction," although they might shear backward. This leads to identifying pairs of types of arrangements that are transformed into one another by a local, elementary shear. To simplify the theory, one pair of orientations is considered, corresponding to the principal axes of the stress tensor. An elementary transformation can be sketched as follows:



although an actual rearrangement involves in general more than four grains. Populations (number densities)

of arrangements of each type are denoted $n_\pm$; the rates $R_\pm$ at which a $\pm \rightarrow \mp$ transformation occurs depend on force and free-volume fluctuations and will be defined further. The macroscopic plastic shear is evaluated from the balance between both types of elementary motion:

$$\dot{\epsilon}^{\mathrm{pl}} = \mathcal{A}_0(R_+ n_+ - R_- n_-), \tag{1}$$

where some constant $\mathcal{A}_0$ has been introduced. Equation (1) is supplemented by equations of motion for the populations $n_\pm$ [2]:

$$\dot{n}_\pm = R_\mp n_\mp - R_\pm n_\pm + |\sigma\dot{\epsilon}^{\mathrm{pl}}|(\mathcal{A}_c - \mathcal{A}_a n_\pm). \tag{2}$$

$\mathcal{A}_c$ and $\mathcal{A}_a$ are constants. The first two terms on the right-hand side (rhs) account for the internal reconformations of STZ's, while the last term introduces a coupling with the mean flow. At the macroscopic scale, the flow constantly stirs the molecules, thus creating and destroying local configurations. The rate at which the macroscopic flow induces new configurations is estimated as the ratio of the overall work, $\sigma\dot{\epsilon}^{\mathrm{pl}}$, over some (constant) typical normal force (the force required to modify a contact). An absolute value is used here only for technical reasons: to exclude spurious nonphysical solutions which might result from creation and destruction terms in the case $\sigma\dot{\epsilon}^{\mathrm{pl}} < 0$. In all cases studied, the physical domain $\sigma\dot{\epsilon}^{\mathrm{pl}} > 0$ is invariant under the overall dynamics.

Transformation rates $R_\pm$ depend on volume and force fluctuations: a local reconformation may occur if some volume is available, *and* if the local force network is appropriately oriented. In this work, I assume that the probabilities associated with those two types of fluctuations factorize $R_\pm = R^v(v_f)R^\sigma_\pm(\sigma)$ (which differs from the prescription given in [2]). v_0 denotes the typical free-volume excess that permits a rearrangement. A Poisson distribution is assumed for the sizes of voids in the material. The probability that a fluctuation of free volume larger than v_0 occurs at a given point is thus $R^v \propto \exp(-v_0/v_f)$. Shear stress σ introduces a bias of the force network. The probability that some fluctuation of local forces promotes shearing in the $+ \rightarrow -$ direction is a positive and increasing function of σ. Large forces are distributed exponentially in granular systems [1,9]; it is reasonable to expect $R^\sigma_\pm \propto \exp(\pm\sigma/\bar{\mu})$, where $\bar{\mu}$ measures some typical force of the stress network. In the following, $\bar{\mu} = 1$ is imposed, which fixes the unit of forces. The rates are thus written

$$R_\pm = R_0 \exp(-v_0/v_f)\exp(\pm\sigma),$$

with R_0, the update frequency of microscopic processes. Other choices for the rates $R_\pm$ lead to similar results [10].

This picture is now complemented with free-volume kinetics. In analogy with Eq. (2), it results from a competition between local reconformations of molecules into better packings [4], and the creation of new, random arrangements by the mean flow:

$$\dot{v}_f = -E_1 \exp[-v_1/v_f] + \mathcal{A}_v|\sigma\dot{\epsilon}^{\mathrm{pl}}|. \tag{3}$$

The first term on the rhs accounts for local optimization of packings, at a rate $\exp(-v_1/v_f)$; v_1 is introduced as the typical free-volume fluctuation required for a local collapse. (Relative values of v_1 and v_0 are expected to be related to the shape of the molecules constituting the material.) The second term on the rhs accounts for the fact that the mean flow creates new, unoptimized, hence loose, configurations. Dilatancy results by definition: random loose packing is less dense than random close packing. The average dilatancy δv_f resulting from a macroscopic shear $\delta\epsilon^{\mathrm{pl}}$ is estimated by considering that a fraction of the energy $\sigma\delta\epsilon^{\mathrm{pl}}$ is transformed into $P\delta v_f$ by a levering effect within the contact network: $\delta v_f \propto \sigma\delta\epsilon^{\mathrm{pl}}/P$. In the following, pressure dependence is incorporated in constants, $P = 1$, and $\mathcal{A}_v = 1$ is imposed, which fixes the unit of v_f. Note that in the absence of shear ($\sigma = 0$) the residual equation, $\dot{v}_f = -E_1 \exp(-v_1/v_f)$, has been proposed by several authors to account for the logarithmic relaxation of v_f [4].

Before proceeding, Eqs. (1) and (2) are written in a more appealing form. Following [2], variables

$$\Delta = \frac{n_- - n_+}{n_\infty} \quad \text{and} \quad \Lambda = \frac{n_+ + n_-}{n_\infty}$$

are introduced and the parameters $n_\infty = 2\mathcal{A}_c/\mathcal{A}_a$, $\epsilon_0 = \mathcal{A}_0\mathcal{A}_c/\mathcal{A}_a$, $\gamma = \mathcal{A}_0\mathcal{A}_c$, and $E_0 = 2\epsilon_0 R_0$. It comes

$$\dot{\epsilon}^{\mathrm{pl}} = E_0 \exp[-v_0/v_f][\Lambda \sinh(\sigma) - \Delta \cosh(\sigma)], \tag{4}$$

$$\dot{\Delta} = \frac{1}{\epsilon_0}(\dot{\epsilon}^{\mathrm{pl}} - \gamma|\sigma\dot{\epsilon}^{\mathrm{pl}}|\Delta), \tag{5}$$

$$\dot{\Lambda} = \frac{1}{\epsilon_0}\gamma|\sigma\dot{\epsilon}^{\mathrm{pl}}|(1 - \Lambda). \tag{6}$$

Variables Λ and Δ represent, respectively, the total normalized density of STZ's and the bias between the populations $n_\pm$. In the absence of shear motion, the dynamics of Λ and Δ freeze, while v_f undergoes "autonomous" relaxation. Initial values for those variables account for the preparation of the system.

In the absence of shear, $\sigma = 0$, the system undergoes time-logarithmic free-volume relaxation. If the picture of a dense material was complete, a reasonable initial state should correspond to values of v_f for which the factor $\exp(-v_1/v_f)$ is vanishingly small: the system is an aged glass. However, for a finite temperature, free volume is expected to equilibrate at some nonvanishing value. Temperature dependence is not included in the current approach; the initial condition for v_f is a fraction of v_0, which is small, but not as small as would be expected from an old material.

I first describe the resulting behavior in a constant stress experiment (creep test), before discussing constant strain-rate experiment (tribology).

Stress σ is fixed. To simplify the analysis, I separate Eqs. (4)–(6) from Eq. (3). Equations (4)–(6) admit two types of solutions: for small values of the applied stress [$\sigma \in [0, \sigma_y]$, with $\sigma_y \tanh(\sigma_y) = 1/\gamma$] the only solutions are jammed states, ($\dot{\epsilon}^{\rm pl} = 0$), with $\Delta = \tanh(\sigma)\Lambda$; above the yield stress, $\sigma > \sigma_y$, jammed states become unstable, and another branch of solution appears, which is stable: a flowing regime ($\dot{\epsilon}^{\rm pl} \neq 0$), with $\Lambda = 1$, $\Delta = 1/\gamma\sigma$, and

$$\dot{\epsilon}^{\rm pl} = E_0 \exp[-v_0/v_f][\sinh(\sigma) - \cosh(\sigma)/(\gamma\sigma)]. \quad (7)$$

Free-volume dynamics (3) admits three types of behavior: either logarithmic relaxation toward 0, $\dot{\epsilon}^{\rm pl}$ remains small, the material creeps; or v_f reaches some nonzero steady value, and the system flows; or v_f diverges, in which case, the weights $\exp(-v_1/v_f)$ and $\exp(-v_0/v_f)$ saturate to 1. The behavior displayed depends on σ, compared to the threshold σ_c, solution of $E_0[\sigma \sinh(\sigma) - \cosh(\sigma)/\gamma] = E_1$. Below σ_c, free volume remains bounded, either relaxing logarithmically to 0 or converging to some asymptotic value. Above σ_c, v_f either keeps relaxing to 0 or diverges, depending on its initial conditions. The latest stages of the dynamics, when v_f gets large, are beyond the scope of the current work. Various creep curves are shown in Fig. 1 (left panel) which compare with experiments [5].

In a stress-strain test, $\dot{\epsilon}$ is fixed, while the shear stress is determined by $\sigma = \mu\epsilon^{\rm el}$, or $\dot{\sigma} = \mu(\dot{\epsilon} - \dot{\epsilon}^{\rm pl})$. Before proceeding, let me show that the same relation is involved in overdamped stick slip. A layer of lubricant is sheared between two plates; the gap between the plates is denoted a and the surface of contact S. A pulling force F is exerted on the upper plate, with a spring of stiffness k, pulled at velocity V. Neglecting the inertia of the upper plate, $F = k(Vt - x)$. The position x of the upper plate is related to the deformation of the lubricant by $\dot{x} = 2a\dot{\epsilon}^{\rm pl}$, and the shear stress is $\sigma = F/S$, whence $\dot{\sigma} = (k/S)(V - 2a\dot{\epsilon}^{\rm pl})$, which is of the form $\dot{\sigma} = \mu(\dot{\epsilon} - \dot{\epsilon}^{\rm pl})$. Identical equations govern both systems, although, remarkably, a solid is usually considered in the former case, and a liquid (at room temperature) in the latter.

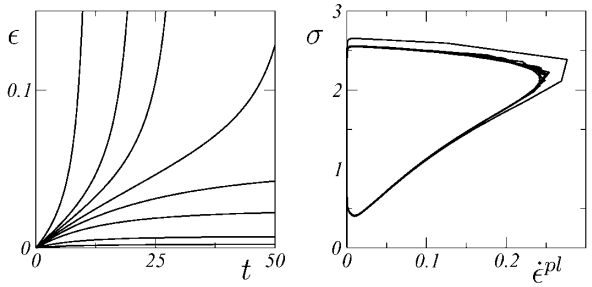


FIG. 1. Left panel: Creep test, deformation as a function of time at fixed σ. Parameters $E_0 = E_1 = 0.1$, $v_0 = 0.5$, $v_1 = 0.4$, $\gamma = 10$, $\epsilon_0 = 1$, and initial conditions $\Delta = 0$, $\Lambda = 0.01$, and $v_f = 0.25$. From bottom to top, $\sigma = 2$, 3, 3.7, 3.9, 4.1, 4.2, and 4.5. Right: Friction force ($\propto \sigma$) as a function of the velocity of the upper plate ($\propto \dot{\epsilon}^{\rm pl}$) during stick-slip motion as shown in Fig. 2 (right panel).

In the following, $\Lambda = 1$ is taken as the initial condition. This is consistent with the fact that stress-strain curves (or stiction peaks) present well-defined features after preparation of the sample by shearing, in which case the dynamics of Λ saturates, and Eq. (6) decouples.

Figure 2 presents examples of behavior resulting from the integration of Eqs. (3)–(5) with $\dot{\sigma} = \mu(\dot{\epsilon} - \dot{\epsilon}^{\rm pl})$ and for different values of $\dot{\epsilon}$. For large values of $\dot{\epsilon}$, the system follows a stress-strain curve which presents strain softening. (Note that this is true for the current choice of parameters, and that strain hardening can be seen for other values.) In the language of tribology, a stiction peak leads to steady sliding. Softening can be understood as follows: during the first stage of the dynamics, plastic deformation $\dot{\epsilon}^{\rm pl}$ remains zero, or very small, either because σ is below the yield stress, or because free volume remains small; the shear force increasing, transient creeping motion induces dilatancy, thus triggering plastic deformation, a further increase of v_f, and σ starts to decrease with the increasing activation factor $\exp(-v_0/v_f)$. For the largest values of $\dot{\epsilon}$ (here $\dot{\epsilon} = 0.1, 0.15, 0.2$), free volume diverges, leading the system to melting or breakup. For intermediate values (here $\dot{\epsilon} = 0.08$) the system converges to steady sliding with finite free volume. For small $\dot{\epsilon}$, stick-slip motion is observed, sliding peaks being associated to sudden rises of the free volume. The friction force is multivalued, and σ as a function of $\dot{\epsilon}^{\rm pl}$ is shown in Fig. 1 (left panel); the cycle

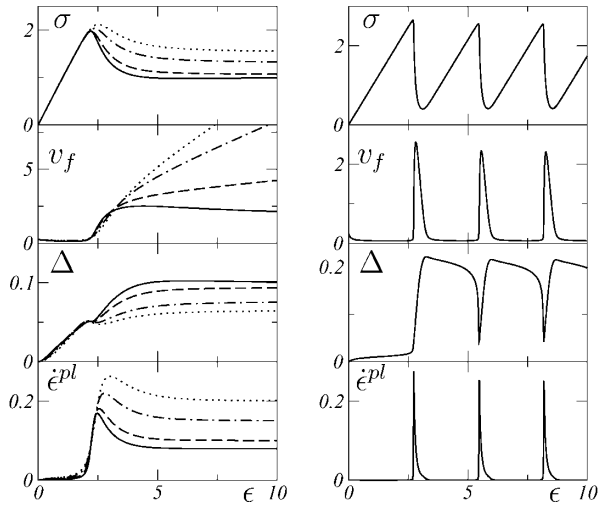


FIG. 2. Behavior of sheared materials for fixed strain rate $\dot{\epsilon}$. Parameters of the theory are $E_0 = 0.1$, $E_1 = 0.1$, $v_0 = 0.5$, $v_1 = 0.4$, $\gamma = 10$, and $\epsilon_0 = 1$. Initial conditions are $\Delta = 0$, $\Lambda = 1$, and $v_f = 0.25$. Left panel: strain softening for $\dot{\epsilon} = 0.08$ (solid line); 0.1 (dashed line); 0.15 (dot-dashed line); 0.2 (dotted line). (Those values are defined up to a time constant and do not need to compare quantitatively with actual values.) Divergence of v_f is seen for the three largest values of $\dot{\epsilon}$. Right panel: stick-slip motion obtained for $\dot{\epsilon} = 0.01$.

is followed in the clockwise direction and is remarkably similar to cycles reported in experiments [7].

It has been checked that the results presented here do not depend qualitatively on other possible choices of $R_{\pm}$, so long as free-volume activation is incorporated. In particular, the rates can be linearized around $\sigma = 0$. The factor $\exp[-v_0/v_f]$, however, is essential, and accounts for the fact that at low free volume, activated processes slow down dramatically. The expression chosen allows one to factorize $\exp[-v_0/v_f]$ in Eqs. (1)–(6); it simplifies somehow the constitutive equations, but more importantly, free volume enters the kinetics of shear motion only as far as it determines its time scale. The coupling between free-volume relaxation and shear deformation is realized through this mechanism only, which is a rather minimal and subtle effect.

It is noteworthy that an early approach to stick slip relied on the introduction of rate-and-state laws, previously used to study earthquakes dynamics [11]. In the current work, free volume is identified as a logarithmically relaxing state variable responsible for strain softening and stick-slip motion. The importance given to free volume provides a direct test of the theory since some appropriate control of the free-volume kinetics could, in principle, prevent stick-slip motion. Although difficult to realize in most experimental setups, this might turn out to have an important practical impact.

The current approach has remained at a qualitative and general level; the specificities of microscopic interactions are not considered and are expected to enter the theory via its parameters. The introduction of a few *mesoscopic* observables and the dynamical coupling via free-volume-dependent time scales leads to a simple structure of constitutive equations that strikingly accounts for important properties of dense, glassy materials. Mesoscopic observables permit one to integrate over small scales and hence introduce short-range correlations; the general structure of the resulting equations corresponds to the introduction of "symmetries" in a dynamical sense.

This theory has now to pass the test of a more stringent comparison with experimental data. This might require minor adaptations, e.g., to the case of granular materials [3,10], but the mechanism proposed is of great simplicity and should adapt to various types of structural systems. A complete picture of dynamical properties of dense materials will require, however, to incorporate the spatial extension of a sample of material and account for localization of the deformation into shear bands. Recent developments in STZ theory have shown a possible mechanism [12] and should benefit from the incorporation of free-volume dynamics.

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[10] Note that specificities of a particular material enter via the quantities $\bar{\mu}$ and R_0. In hard-sphere systems, R_0 is expected to be proportional to the collision frequency, $\sqrt{T}/v_f$ (at temperature T), and the typical normal force, $\bar{\mu}$, is expected to be proportional to the pressure P; moreover, in granular materials, temperature emerges from dynamical fluctuation, and an equation of motion for T must be introduced to complete the theory [3]. For the sake of simplicity, such questions are left for future studies; in the present work, R_0 and $\bar{\mu}$ are constants.

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