Yield stress, heterogeneities and activated processes in soft glassy materials

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Abstract. The rheological behavior of soft glassy materials basically results from the interplay between shearing forces and an intrinsic slow dynamics. This competition can be described by a microscopic theory, which can be viewed as a nonequilibrium schematic mode-coupling theory. This statistical mechanics approach to rheology results in a series of detailed theoretical predictions, some of which still awaiting for their experimental verification. We present new, preliminary, results about the description of yield stress, flow heterogeneities and activated processes within this theoretical framework.

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1. Introduction

The discussion of ‘glassy’ materials in textbooks is usually restricted to simple molecular glasses, such as silica or polymeric glasses, and hence covers the standard field of the ‘glass transition’. An impressive number of experiments performed in the last decade shows that typical glassy effects are not restricted to structural glasses, but are observed in a much wider variety of experimental systems. As a result, ‘glassy dynamics’ is being actively studied in systems as different as dirty type II superconductors, disordered ferromagnets or ferroelectrics, disordered electronic systems, soft glassy materials, granular matter. These observations not only allow to draw interesting analogies between different systems, but also to use similar theoretical paths to describe various fields. This paper is concerned with the application of a theory initially developed in the context of the statistical mechanics of glasses to describe the rheology of soft glassy materials.

The term ‘soft glassy materials’ was proposed in Ref. [1] as a generic name for a large family of complex fluids which share a similar phenomenology: colloids, emulsions, pastes, clays... It was then assumed that their physical behavior basically results from the competition between an intrinsic glassiness, in the sense of large relaxation times, and shearing forces which ‘accelerate’ their dynamics. This competition was then theoretically described using the simple trap model of Ref. [2], phenomenologically extended to account for the effect of an external flow. The model was further studied in Refs. [3]. Since then, various phenomenological approaches have been proposed, which replace the concept of ‘traps’ by ‘fluidity’ [4], ‘free volume’ [5], or ‘degree of jamming’ [6], but end up with extremely similar mathematical formulations. The interest of such models is that they are simple enough so that complicated flow and thermal histories can be easily implemented. The obvious drawback is that the
pysics is put by hand from the beginning: shear is assumed to reduce the trap depth, or to increase the fluidity, the free volume, the degree of jamming.

The approach we discuss here is less transparent but does not assume anything beyond the dynamical evolution of a Langevin type for a glassy system defined by its Hamiltonian. Interestingly, it can alternatively be viewed as a nonequilibrium extension of schematic mode-coupling theories, as we describe in section 3. A very similar approach was recently followed in Ref. 8. Static and dynamic behaviors can be studied in such detail that a very complete physical description can be proposed. The theory is also sufficiently understood that its shortcomings, domain of validity, and, in principle, the path to possible improvements are also known. At present, only the case of steady shearing was investigated in great detail, as reviewed in section 3. We hope to report on transient behaviors in a near future, the difficulties being mainly technical. The paper also contains two new steps. First, we discuss the issue of a yield stress, and present preliminary results for its behavior in section 4. Second, we show in section 5 that two dynamical solutions for a given shear stress are possible in a certain regime and discuss this feature in light of the recent observations of flow heterogeneities.

2. A microscopic approach for nonlinear rheology

What would be an ideal theory for glassy rheology? Ideally, one would like to start from a microscopic equation for the dynamic evolution of the system under study, say a supercooled liquid in a shear flow, and solve this dynamics exactly. This ‘first-principles’ approach is obviously an extraordinary challenge and approximations have to be made in order to get a system of closed dynamic equations. A well-known approximation in the field of the glass transition is the mode-coupling approximation which leads to the mode-coupling theory (MCT) of the glass transition. That the resulting equations can be obtained in a standard perturbative way is discussed in Refs. 9, 11, 12, although the original derivation makes this less transparent.

Generally speaking, starting from an evolution equation for the density fluctuations $\delta \rho(k, t)$, where $k$ is a wavevector and $t$ is time, and a given pair potential interaction $V(k)$, the mode-coupling approximation amounts to a partial resummation of diagrams in the perturbative development of the dynamical action. One obtains dynamical equations (Dyson equations) which close on two-point correlation $C(k, t, t') = \langle \delta \rho(k, t) \delta \rho(-k, t') \rangle$ and conjugated response function $\chi(k, t, t')$. At thermal equilibrium, the fluctuation-dissipation theorem $T \chi(k, t, t') = \partial C(k, t, t') / \partial t'$, implies coupled equations for the density correlators only: this is the MCT. The theory has then the pair potential (or alternatively the static structure factor $S(k) = C(k, t, t)$) as an input, and the dynamic behavior of the liquid, the correlators $C(k, t, t')$, as an output. ‘Schematic’ models focusing on a given ‘important’ wavevector, say $k_0$, were formulated. This amounts to write $S(k) \approx 1 + A \delta(k - k_0)$ and focus on $C(t, t') \equiv C(k_0, t, t')$ as a single observable. These schematic formulations lose the ‘ab-initio’ character of the full MCT, but they do essentially capture the dynamical singularities arising when the wave-vector dependence is kept.

The MCT of the glass transition has been discussed at length in the literature, although two points we would like to emphasize, though.

(i) The ‘perturbative’ derivation described above where both correlation and response are kept, opens the door to the study of nonequilibrium behaviors. The
system can be out of equilibrium because the equilibration time scale is too large for the experimental window: one focuses then on aging behaviors [15]. The system can also be driven out of equilibrium by some external force, for instance a shear force. This is the latter, rheological, situation we shall be interested in.

(ii) Kirkpatrick and Thirumalai remarked that the schematic models can be exactly derived starting from some mean-field disordered Hamiltonian, like Potts and $p$-spin models [16]. The connection was studied further in an important series of papers [17]. Beyond the basic connection made between two fields (spin and structural glasses), these works more importantly complement MCT with all the knowledge one can get from the thermodynamic studies of, say, the $p$-spin model. It is worth recalling that the $p$-spin model exactly realizes most of the thermodynamic ‘folklore’ of the glass transition [14], such as such an entropy crisis, or the existence of many metastable states [14]. Both sides, MCT and mean-field disordered models, are now indissociable and form an ensemble that could generically be called the ‘mean-field theory of the glass transition’. We shall see below how the knowledge of the free energy landscape (the ‘spin glass part’ of the theory) allows one to make qualitative predictions for the dynamics (the ‘MCT part’), that appears to be verified when the actual calculations are made.

3. Steady rheology: A brief review

The microscopic approach to glassy rheology described in general terms in the previous section was quantitatively investigated in Ref. [7]. There, a schematic model (of the $F_p$ family [14]) was extended to take into account the crucial ingredient that the dynamics is externally driven. Technically, this amounts to break detailed balance and leads to closed coupled equations for a correlator and its conjugated response function. As mentioned above, these equations can be alternatively derived from the driven Langevin dynamics of the $p$-spin model [18]. These dynamic equations were then solved in the plane (Driving force, Temperature) in the case of a constant driving force, and the results interpreted in the language of nonlinear rheology, the driving force being analogous to a shear stress $\sigma$. Recall that in the absence of the shearing force, the model has a dynamic transition at a temperature $T_c$ where the relaxation time diverges as a power law. Just above $T_c$, one has the standard two-step decay of the correlation, characterized by functional forms described in detail in Ref. [10].

In the presence of the driving force, the whole physical behavior is encoded in the time decay of correlation and response functions. Their analysis leads to predictions at several levels [7]. At the macroscopic level first, one gets flow curves relating the viscosity $\eta \equiv \int dt C(t)$ to the shear stress $\sigma$, see Fig. 1. Beyond a linear regime restricted to the high-$T$, low-$\sigma$ region, the system is strongly shear-thinning. The power law $\eta \propto \gamma^{-2/3}$ is obtained at $T = T_c$, where $\gamma = \sigma/\eta$ is the shear rate. In the glassy phase, $T < T_c$, the model describes a power law fluid, $\eta \approx \gamma^{-\alpha(T)}$, with no ‘dynamic’ yield stress ($\alpha(T) < 1 \Rightarrow \lim_{\gamma \to 0} \sigma(\gamma) = 0$) [13]. Recall that the shear-thinning behavior is derived in the present framework without any assumption. The predicted macroscopic behavior compares very well with experiments [20] and simulations [21,22].

Second, in the spirit of MCT, predictions can be made concerning the functional form of the correlators in various time regimes. They are the so-called ‘factorization property’ at intermediate times and the ‘superposition principle’ at large times. Again, these are well verified in simulation works [22]. We are not aware of any experimental
Third, a modified fluctuation-dissipation relation between correlation and response is also derived. It reads
\[ \chi(t) = -\frac{1}{T_{\text{eff}}} \frac{dC(t)}{dt}, \]  
where the effective temperature \( T_{\text{eff}} \) replaces the equilibrium temperature \( T \) \[23,24\]. While the short processes show \( T_{\text{eff}} = T \), at long time one gets \( T_{\text{eff}} > T \), showing that slow processes are ‘quasi-equilibrated’ at an effective temperature higher than the thermal bath temperature. Again, extensive numerical simulations have confirmed this prediction for various physical observables in a sheared fluid \[22,24,26,27\]. Again, this important prediction has not been checked experimentally in this context, although specific protocols have been described in Refs. \[22,26\]. We refer to Ref. \[28\] for experimental studies of aging systems.

4. Yield stress and activated processes

Having access to a microscopic (spin glass) model behind the dynamical equations allows us to understand the evolution from the geometry of the corresponding phase space. At zero external drive, this connection has been studied in much detail \[29,30,31,32\]. Above the dynamical transition temperature \( T_c \), the available phase space is dominated by one large basin in the free energy, corresponding to the ‘paramagnetic’, or ‘liquid’, state. At \( T_c \), a threshold level in free energy appears, below which the free energy surface is split into exponentially-many disconnected regions.

The aging dynamics below \( T_c \) can be understood as a gradual descent to the threshold level, starting from high energy configurations \[24\]. The slowing down is the consequence of the decreasing connectivity of the visited landscape. See Ref. \[33\] for recent similar statements at equilibrium. When the system is quenched from a
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Figure 2. Existence of a static yield stress. The system is first prepared below the free energy threshold, as in Ref. [32]. The driving force is then applied. The two curves are both taken in the subsequent nonequilibrium steady state and show solid, $\sigma < \sigma_Y$, or liquid, $\sigma > \sigma_Y$, behaviors; $T = 0.1$.

high temperature, but at the same time driven by non-conservative forces, it remains similarly drifting above the free energy threshold, constantly receiving energy from the drive.

On the other hand, if the system is prepared in one of the deep regions below the threshold, it remains trapped for all times [32]. From this picture, we expect in that case that a weak driving force will have essentially no effect beyond a trivial 'elastic' response of the system, as it is not strong enough to make the system overcome the barriers. If instead a strong drive is applied, the system should escape the low-lying valley and surfaces above the threshold, where the drive will suffice to keep it forever.

We have investigated this situation using the method of Refs. [18, 34]. A typical result is shown in Fig. 2, which shows that the above expectations deduced from the topology of the free energy landscape are well verified when the calculations are done. This proves the existence of a static yield stress $\sigma_Y(T)$ in the model [19]. Figure 2 is also clearly reminiscent of the thixotropic behaviors [35] commonly encountered in soft glassy materials.

The difference between the free energy threshold and the equilibrium energy vanishes for $T \to T_c^-$, and the very notion of a threshold disappears. One expects therefore, on physical grounds, that $\lim_{T \to T_c^-} \sigma_Y(T) = 0$. This expectation is in qualitative disagreement with Ref. [8]. We are currently studying the temperature dependence of $\sigma_Y(T)$ in our model to conclude on this issue.

We are now in a position to recall what is the main shortcoming of this mode-coupling type of approach. In any realistic system the structure of threshold and valleys may remain essentially the same, but now activated processes allow to jump barriers that are impenetrable at the perturbative level. In the spin language, barriers between states diverge at the thermodynamic limit, $N \to \infty$, where $N$ is the total number of spins.

Despite several attempts, the analytical inclusion of activation in this framework
Figure 3. Self-similar activated dynamics within the metastable states of the finite-N version of the model driven by an external force. The first curve (top left) is taken in a single realization of the process. The other curves are successive zooms of the first one, as can be seen from the time axis. A magnification factor of \( \approx 6 \) is applied 3 times until elementary jumps are resolved in the last figure (bottom right). Parameters are \( N = 20, T = 0.05 \).

remains elusive. However, a qualitative understanding may be gained by studying the driven dynamics of the finite-N version of the spin glass model corresponding to our theory. Keeping \( N \) finite directly implies that barriers are finite, allowing thermal activation to play the role it cannot play in the thermodynamic limit \([18]\). This type of study would not be possible within the MCT framework alone. We have thus investigated by means of Monte Carlo simulations the Ising \( p \)-spin model (for \( p = 3 \)), defined by the Hamiltonian

\[
H = - \sum_{i<j<k}^{N} J_{ijk} s_i s_j s_k, \tag{2}
\]

where \( J_{ijk} \) is a random Gaussian variable of mean 0 and variance \( \sqrt{3}/N \). The driving force is implemented by the use of asymmetric coupling constant, the amplitude of which being the driving force, which we call \( \sigma \), since it has the same role as the shear stress in Ref. \([7]\); see also \([18, 23]\). We numerically find clear evidences for the existence of a critical driving force \( \sigma_Y(T) \) below which the system is trapped (‘solid’), and above which it is not (‘liquid’). The novelty is that activated processes now play an important role. This can be observed in Fig. 3 where the time dependence of the energy density is represented for a driving force amplitude just below the yield value. Strong fluctuations are observed in the energy density, and the system alternates via thermal activation between trapping periods and periods of freedom \([18, 36]\).
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Figure 4. Distribution of trapping times $\varphi(\tau)$ for various temperatures in the presence of activation. The different curves are vertically shifted for clarity. The full lines are the theoretical prediction, $\varphi(\tau) \propto \tau^{-(1+T/T_c)}$, with no fitting parameter.

Interestingly, Fig. 4 also shows that this time evolution is self-similar, in the sense that zooming on a particular time window leads to a very similar picture. In Fig. 3, a zooming procedure by a factor of order 6 is repeated 3 times until unitary moves are resolved in the last figure. This strongly suggests that trapping times are power-law distributed. This is indeed what we find when the pictures of Fig. 3 are quantitatively analyzed to extract the distribution of trapping times $\varphi(\tau)$. This is shown in Fig. 4, together with excellent comparison to the theoretical prediction $\varphi(\tau) \sim \tau^{-\beta(T)}$ with an exponent $\beta(T) = 1 + T/T_c$ which is discussed below. Remark that the mean trapping time $\langle \tau \rangle = \int d\tau' \varphi(\tau')\tau'$ is infinite for $\beta(T) < 2$, which is equivalent to say that $\sigma < \sigma_Y$. Consistently with the previous discussion, we find that $\sigma_Y(T)$ decreases when $T$ is increased, and our preliminary results indicate that $\sigma_Y(T \to T_c^-) \to 0$. More quantitative studies are also in progress.

This self-similar behavior is theoretically expected since a power law distribution can be obtained invoking activation dynamics in a landscape with exponentially distributed energy barriers, as is the case of the random energy model \[37\], obtained in the $p \to \infty$ limit. This view is obviously reminiscent of the dynamics of the trap model \[2\], the connection between the two approaches being mathematically described in Ref. \[38\]. These considerations in fact led us to predict the temperature dependence of the trapping times distribution in Fig. 4 where we took $\beta(T) = 1 + T/T_c$. Note also the similarity between Fig. 4 and recent numerical investigations of the ‘potential energy landscape’ at equilibrium around $T_c$ where results were quantitatively interpreted in the framework of the trap model \[39\].

5. Flow heterogeneities

The results presented in the two previous sections have interesting implications that we now discuss in detail. We noted in section 4 that our model describes a power
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Figure 5. A singular, or ‘nonmonotonic’, flow curve for $T = 0.1$ in the model of Ref. [7]. The horizontal dashed line is the value of the yield stress at this temperature. The region where spatial flow heterogeneities (shear-bands) may appear is delimited with the vertical line at $\gamma = \gamma_c$ (see text). Vertical points symbolizes the fact that any shear stress $\sigma < \sigma_Y$ is possible when $\gamma = 0$ (this value of $\gamma$ is of course not visible in a log-scale).

law fluid at low temperatures, $\sigma \approx \gamma^{1-\alpha(T)}$, the shear-thinning exponent satisfying $\frac{\alpha}{2} = \alpha(T_c) < \alpha(T) < 1$. When the consideration on the yield stress of section 4 are included, the complete flow curve of the material admits a singular $\gamma \to 0$ limit, see Fig. 5. Hence, the flow curve becomes ‘nonmonotonic’. This singularity is well-known at the experimental level [6, 19]. Note that it physically results, in our theoretical description, from the existence of the threshold value in the free energy. Note also that a purely dynamical approach à la MCT misses this subtlety.

A direct consequence is that there is a complete interval of shear stresses, $\sigma \in [0 : \sigma_Y]$, for which two dynamical solutions are possible. This is exemplified in Fig. 6, where the correlation function is represented for the same value of the shear stress, but starting either from random initial conditions (‘shear immediately’), or from an equilibrated initial condition below the free energy threshold (‘aging then shear’). Both curves are obtained in a driven steady state for the same value of the control parameters ($T, \sigma$), and are equally stable at the mean-field level.

Looking again at Fig. 5, one sees that the flow curve defines a critical value $\gamma_c$ of the shear rate, defined by $\sigma(\gamma_c) = \sigma_Y$ and represented by a vertical line in the figure. A very interesting question now is: what happens in an experiment if a global shear rate $\gamma_{global} < \gamma_c$ is applied to the sample? An homogenous flow would indeed correspond to a stress $\sigma < \sigma_Y$ for which a second dynamical solution exists. The answer is known from experiments [10] and simulations [11]: the system will spontaneously develop flow heterogeneities, where a flowing band ($\gamma_{local} > 0$) coexist with a non-flowing band ($\gamma_{local} = 0$), both supporting the same value of the shear stress. In that case, flowing regions will display a dynamic behavior described by the dashed line in Fig. 5 whereas the second, jammed, band will be described by the full line in Fig. 5. This is also observed in a recent numerical simulation, see Fig. 2 of
Figure 6. Two possible dynamical solutions for $T = 0.1$ and $\sigma = 0.1$ in the model of Ref. [7]. They coexist in a single sample when the system displays shear-bands. Compare with Fig. 2 of Ref. [41].

Ref. [41]. This shear-banding phenomenon results then from the ‘nonmonotonicity’ of the flow curve in Fig. 6, and can again ultimately be viewed, in our model, as an experimental consequence of the notion of threshold in the free energy landscape.

An open problem, of course, is the dynamical selection of the bands in a real sheared material [42, 43]. How does the system choose the relative size of the bands? The problem is difficult, since no thermodynamic argument (such as a Maxwell construction) can be applied to this nonequilibrium situation. The same question is presently much discussed in various complex fluids, like liquid crystals [44] or wormlike micelles [45].

6. Conclusion

In these proceedings, preliminary results concerning the theoretical description of the yield stress, the flow heterogeneities and the role of activated processes in soft glassy materials via a nonequilibrium schematic mode-coupling theory were presented. An advantage of our approach is that no assumption are made, at odds with the more phenomenological models which are usually used in the field of rheology. This allows to make detailed microscopic predictions, much beyond the macroscopic rheological level where concurrent models are stuck. These predictions were briefly reviewed in section 3. We emphasize, as we did in Ref. [26], that many of them are still experimentally unverified. In that sense, the situation is very similar to the mid-1980’s, where schematic mode-coupling models were derived, but with little experimental confirmation of their main features.

We mentioned at several places of this paper that our results were only preliminary, and we took advantage of this conference to present our ‘work in progress’. Although more precise and complete studies are obviously necessary, we have shown that a qualitative understanding of the rheological behavior could be gained via this approach, beyond the steady rheological situation studied in Ref. [6].
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A more quantitative approach would be to derive a ‘full’ nonequilibrium theory for sheared fluids. This program was recently undertaken by Fuchs and Cates. Their derivation makes use of a projector formalism instead of the perturbative development described in section 2, and ends up with dynamic equations for correlators only. It is not clear at the moment if the qualitative discrepancies between the two approaches underlined in this paper are due to this alternative derivation where nonequilibrium effects (like the violation of the fluctuation-dissipation theorem) are not described. Comparisons between the two approaches would thus be very interesting.

Last, we mention also that if flow heterogeneities are suggested in our model by the existence of two dynamical solutions for a given shear stress, this does not imply that shear-banding has to actually take place, nor does it allow to gain any insight into the problem of the selection between the two solutions. This certainly requires more ambitious approaches than ours.

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