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Yield stress, heterogeneities and activated processes in soft glassy materials*

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Abstract

The rheological behaviour of soft glassy materials results essentially 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 are yet to be experimentally verified. We present new, preliminary, results about the description of yield stress, flow heterogeneities and activated processes within this theoretical framework.

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'. Many experiments performed in the last decade show 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' are being actively studied in systems as diverse as dirty type II superconductors, disordered ferromagnets or ferroelectrics, disordered electronic systems, soft glassy materials and granular matter. These observations not only allow us 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 [1] as a generic name for a large family of complex fluids which share a similar phenomenology: colloids, emulsions, pastes, clays, ... It was assumed that their physical behaviour results essentially from the competition between an intrinsic glassiness, in the sense of large relaxation times, and shearing forces which 'accelerate' their dynamics. This competition was theoretically described using the simple trap model of [2], phenomenologically extended to account for the effect of an external flow.

* This paper is dedicated to $\mathbf{2}\varepsilon$ who became $O(\mathbf{1})$ during this conference.

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The model was further studied in [3]. Various phenomenological approaches have since 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. Such models are interesting because they are simple enough that complicated flow and thermal histories can be easily implemented. The obvious drawback is that the physics is put in by hand from the beginning: shear is assumed to reduce the trap depth, or to increase the fluidity, the free volume or the degree of jamming.

The approach we discuss here is less transparent but does not assume anything beyond the dynamical evolution of Langevin type for a glassy system defined by its Hamiltonian [7]. Interestingly, it can alternatively be viewed as a nonequilibrium extension of schematic mode-coupling theories (MCTs), as we describe in section 2. A very similar approach was recently followed in [8, 9]. Static and dynamic behaviour can be studied in such detail that a 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 has been investigated in great detail, as reviewed in section 3. We hope to report on transient behaviour in the near future, when the present (mainly technical) difficulties have been overcome. The paper also contains two new steps. First, we discuss the issue of a yield stress, and present preliminary results for its behaviour 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? 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 a great 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 [10] which leads to the MCT of the glass transition [11]. That the resulting equations can be obtained in a standard perturbative way is illustrated in [10, 12, 13], 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 closed dynamical equations (Dyson equations) involving two-point correlation $C(k,t,t') = \langle\delta\rho(k,t)\delta\rho(-k,t')\rangle$ and conjugated response functions $\chi(k,t,t')$. 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 behaviour of the liquid, the correlators $C(k,t,t')$, as an output. 'Schematic' models focusing on a given 'important' wavevector, say k_0, have been formulated [14]. This amounts to writing $S(k) \approx 1 + A\delta(k-k_0)$ and focusing 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 wavevector dependence is kept [11].

The MCT of the glass transition has been discussed at length in the literature [11, 15]. There are 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 behaviour. The system can be out of equilibrium because the equilibration timescale is too large for the experimental window: one focuses then on aging behaviour [16]. The system can also be driven out of equilibrium by some external force, for instance a shear force. It is the latter, rheological, situation we shall be interested in.

(ii) Kirkpatrick and Thirumalai [17] remarked that the schematic models can be exactly derived starting from some mean-field disordered Hamiltonian, like Potts and p-spin models. The connection was studied further in an important series of papers [18]. Beyond the basic connection made between two fields (spin and structural glasses), this work, more importantly, complements MCT with 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 [15], like an entropy crisis, or the existence of many metastable states [17]. 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 appear 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 [7]. There, a schematic model (of the F_p family [11]) was extended to take into account the crucial ingredient that the dynamics is externally driven. Technically, this amounts to breaking 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 [19]. These dynamic equations were then solved in the plane (driving force, temperature) for the case of a constant driving force. The results were interpreted in the language of nonlinear rheology, with the driving force being analogous to a shear stress σ. 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 [11].

In the presence of a finite driving force, a steady state is reached at all temperatures [7, 19] The whole physical behaviour is encoded in the time decay of correlation and response functions. Their analysis leads to predictions at several levels [7].

At the macroscopic level, one gets flow curves relating the viscosity $\eta \equiv \int \mathrm{d}t\, C(t)$ to the shear stress σ; see figure 1. Beyond a linear regime restricted to the high-T, low-σ region, the system is strongly shear thinning. The power law $\eta \propto \dot{\gamma}^{-2/3}$ is obtained at $T = T_c$, where $\dot{\gamma} = \sigma/\eta$ is the shear rate. In the glassy phase, $T < T_c$, the model describes a power law fluid, $\eta \approx \dot{\gamma}^{-\alpha(T)}$, with no 'dynamic' yield stress ($\alpha(T) < 1 \Rightarrow \lim_{\dot{\gamma}\to 0} \sigma(\dot{\gamma}) = 0$) [20]. Recall that the shear-thinning behaviour is *derived* in the present framework without any assumption. The predicted macroscopic behaviour compares very well with experiments [21] and simulations [22, 23]. Note that, in practice, the situation described here is more easily realized in a soft glassy material, which is easily deformable on experimentally accessible timescales. Molecular glasses, by contrast, would exhibit shear-banding and fracture.

Second, in the spirit of MCT, predictions can be made concerning the functional form of the correlators in various time regimes, namely the so-called 'factorization property' at

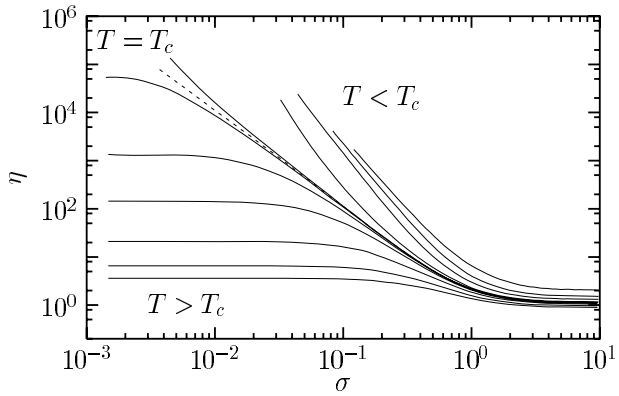

Figure 1. Stationary flow curves $\eta = \eta(\sigma)$ for various temperatures above, at and below T_c, in the model of [7]. Newtonian behaviour is obtained at low σ, high T. Power law shear-thinning is obtained everywhere else, with $\eta \propto \dot{\gamma}^{-2/3}$ at T_c. Typical viscosities at the 'glass transition' (as defined by the experimental time scale) are $\approx 10^{12}$ Pa s for simple liquids, and $\approx 10^3$ Pa s for colloidal systems.

intermediate times and the 'superposition principle' at large times[1]. Again, these are well verified in numerical works [23].

Third, a modified fluctuation–dissipation relation between correlation and response is derived. It reads

$$\chi(t) = -\frac{1}{T_{\rm eff}} \frac{{\rm d}C(t)}{{\rm d}t}, \tag{1}$$

where the effective temperature $T_{\rm eff}$ replaces the equilibrium temperature T [24, 25]. While the short processes show $T_{\rm eff} = T$, at long time one gets $T_{\rm eff} > T$, 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 [23, 26–28], though it has not been checked experimentally in this context. However, specific protocols have been described in [23, 27]. We moreover refer to [29] for experimental studies of aging systems.

4. Yield stress and activated processes

The microscopic (spin glass) model underlying 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 detail [30–33]. 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 the 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 [25]. The slowing down is a consequence of the decreasing connectivity of the visited landscape. See [34] for recent similar statements at equilibrium. When the system is quenched from a high temperature, but at the same time

[1] In [7] a time–shear stress superposition is shown to be satisfied in the steady state, in that the slow time decays of correlators superpose when the time is rescaled by a shear stress dependent relaxation time, $C(t) \sim f[t/t_{\rm rel}(\sigma)]$. For steady states, this is equivalent to a time–shear rate superposition, the latter being more conveniently tested in simulations [23, 26].

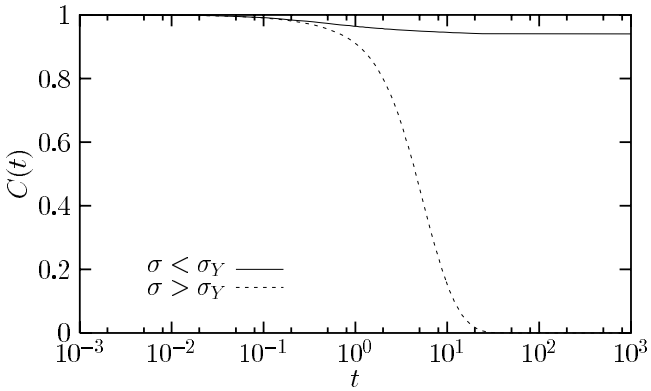

Figure 2. Existence of a static yield stress. The system is first prepared below the free energy threshold, as in [33]. The driving force is then applied. The two curves are both taken in the subsequent nonequilibrium steady state and show solid, $\sigma < \sigma_Y$, or liquid, $\sigma > \sigma_Y$, behaviour; $T = 0.1$. The same 'liquid' behaviour would be obtained starting from random initial conditions, as in figure 1.

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 [33]. 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 [19, 35]. A typical result is shown in figure 2, which shows that the above expectations deduced from the topology of the free energy landscape are verified by the calculations. This proves the existence of a static yield stress $\sigma_Y(T)$ in the model [20], as defined by the minimal force required to make it flow. Figure 2 is also clearly reminiscent of the thixotropic behaviour [36] 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 [8]. We are currently studying the temperature dependence of $\sigma_Y(T)$ in our model to resolve this issue.

We are now in a position to recall the main shortcoming of this mode-coupling-type approach. In any realistic system the structure of threshold and valleys may remain essentially the same, but now *activated processes* allow the system to jump barriers that are impenetrable at the perturbative level. In 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 description of activation in this framework 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 implies that barriers are finite, allowing thermal activation to play the role it cannot play in the thermodynamic limit [19]. This type of study would not be possible within the MCT framework alone. We have therefore investigated by means of Monte Carlo simulations the

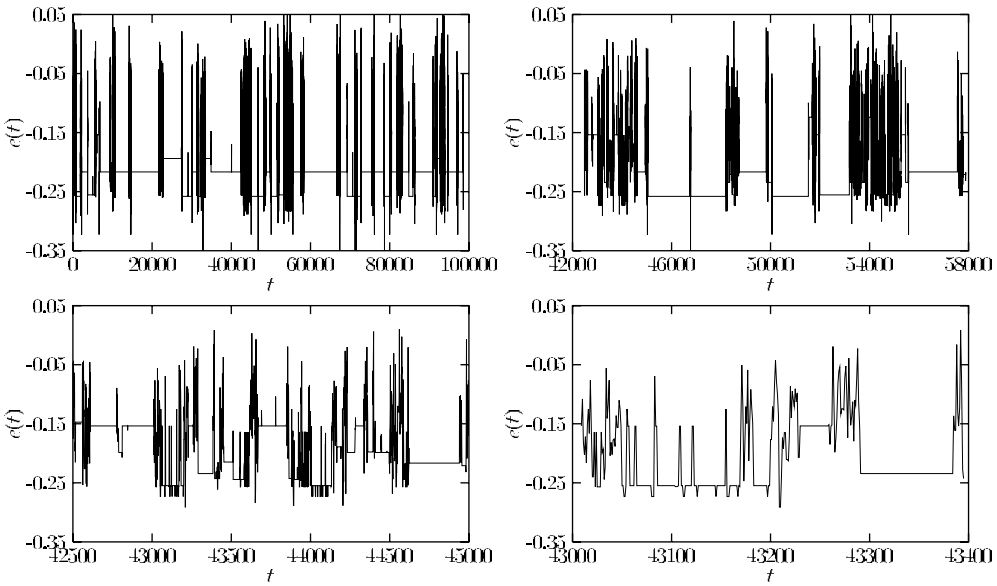

Figure 3. Self-similar activated dynamics within the metastable states of the finite-N version of the model driven by an external force. The first curve (top left) is taken in a single realization of the process. The three other curves are successive enlargement zooms of the first one, as can be seen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

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 zero and variance $\sqrt{3}/N$. The driving force is implemented by the use of an *asymmetric* coupling constant, the amplitude of which is the driving force, which we call σ, since it has the same role as the shear stress in [7]; see also [19, 24]. We find numerically clear evidence 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 figure 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 [19, 37].

Interestingly, figure 3 also shows that this time evolution is self-similar, in the sense that zooming on a particular time window leads to a very similar figure. In figure 3, a zooming procedure by a factor of approximately six is repeated three times until elementary 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 figure 3 are quantitatively analysed to extract the distribution of trapping times $\varphi(\tau)$. This is shown in figure 4, together with an excellent comparison to the theoretical prediction $\varphi(\tau) \sim \tau^{-\beta(T)}$ with an exponent $\beta(T) = 1 + T/T_c$ discussed below. We remark that the mean trapping time $\langle\tau\rangle = \int \mathrm{d}\tau' \varphi(\tau')\tau'$ is infinite for $\beta(T) < 2$, which is equivalent to saying that $\sigma < \sigma_Y$. Consistent 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.

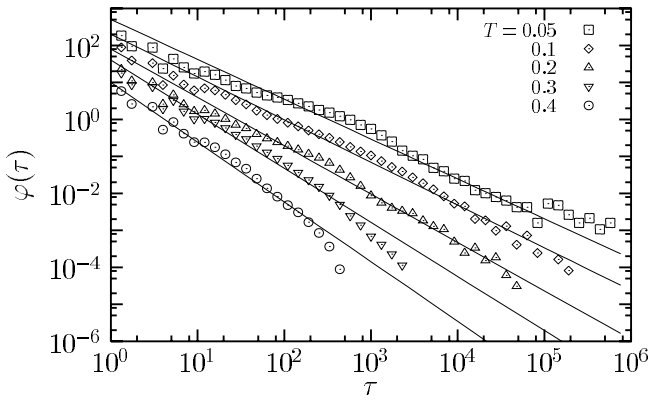

Figure 4. Distribution of trapping times $\varphi(\tau)$ for various temperatures in the presence of activation. The different curves are vertically shifted for clarity. The full lines are the theoretical prediction, $\varphi(\tau) \propto \tau^{-(1+T/T_c)}$, with no fitting parameter.

This self-similar behaviour 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 [38], 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 [39]. These considerations in fact led us to predict the temperature dependence of the trapping time distribution in figure 4 where we took $\beta(T) = 1 + T/T_c$. Note also the similarity between figure 3 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 [40].

5. Flow heterogeneities

The results presented in the two previous sections have interesting implications that we now discuss in detail. We noted in section 3 that our model describes a power law fluid at low temperatures, $\sigma \approx \dot{\gamma}^{1-\alpha(T)}$, the shear-thinning exponent satisfying $\frac{2}{3} = \alpha(T_c) < \alpha(T) < 1$. When consideration of the yield stress of section 4 is included, the complete flow curve of the material admits a singular $\dot{\gamma} \to 0$ limit; see figure 5. Hence, the flow curve becomes 'nonmonotonic'. This singularity is well known at the experimental level [6, 20]. 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 [8].

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 figure 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 ('ageing then shear'). Both curves are obtained in a driven steady state for the *same* value of the control parameters (T, σ), and are equally stable at the mean-field level.

Looking again at figure 5, one sees that the flow curve defines a critical value $\dot{\gamma}_c$ of the shear rate, defined by $\sigma(\dot{\gamma}_c) = \sigma_Y$ 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 $\dot{\gamma}_{\text{global}} < \dot{\gamma}_c$ is applied to the sample? A homogeneous flow would indeed correspond to a stress $\sigma < \sigma_Y$

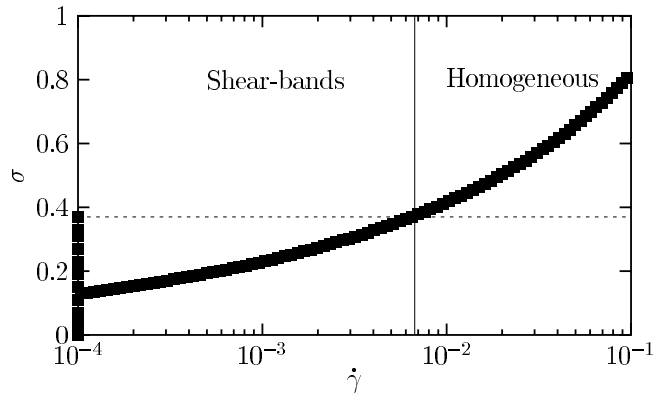

Figure 5. A singular, or 'nonmonotonic', flow curve for $T = 0.1$ in the model of [7]. The horizontal dashed line is the value of the yield stress at this temperature. The region where spatial flow heterogeneities (shear-bands) may appear is delimited with the vertical line at $\dot{\gamma} = \dot{\gamma}_c$ (see text). Vertical points symbolize the fact that any shear stress $\sigma < \sigma_Y$ is possible when $\dot{\gamma} = 0$ (this value of $\dot{\gamma}$ is of course not visible in a log scale).

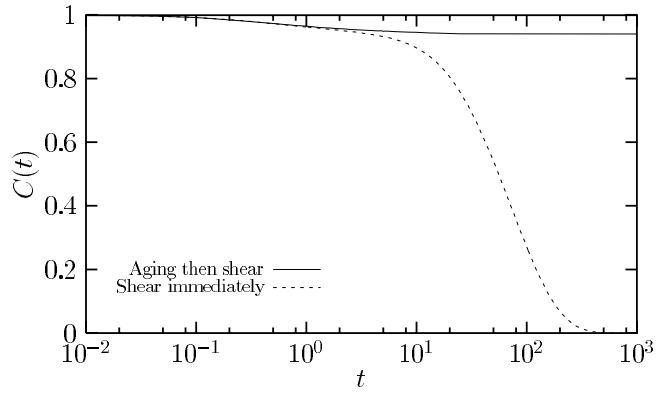

Figure 6. Two possible dynamical solutions for $T = 0.1$ and $\sigma = 0.1$ in the model of [7]. They coexist in a single sample when the system displays shear-bands. Compare with figure 2 of [42].

for which a second dynamical solution exists. The answer is known from experiments [41] and simulations [42]: the system will spontaneously develop *flow heterogeneities*, where a flowing band ($\dot{\gamma}_{\rm local} > 0$) coexists with a non-flowing band ($\dot{\gamma}_{\rm local} = 0$), with both supporting the same value of the shear stress. In that case, flowing regions will display dynamic behaviour described by the dashed curve in figure 6, whereas the second, jammed, band will be described by the full line in figure 6. This is also observed in a recent numerical simulation, see figure 2 of [42]. This shear-banding phenomenon results from the 'nonmonotonicity' of the flow curve in figure 5, and can again ultimately be viewed, in our model, as an experimental consequence of the notion of threshold in the free energy landscape.

One open problem is the dynamical selection of the bands in a real sheared material [43, 44]. 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 the context of various complex fluids, like liquid crystals [45] or wormlike micelles [46].

6. Conclusion

In these proceedings, preliminary results concerning the theoretical description of the yield stress, the flow heterogeneties and the role of activated processes in soft glassy materials via a nonequilibrium schematic MCT have been presented. An advantage of our approach is that no assumptions are made, at odds with the more phenomenological models usually used in the field of rheology. This allows us to make detailed microscopic predictions, far beyond the macroscopic rheological level where concurrent models are stuck. These predictions were briefly reviewed in section 3. We emphasize, as we did in [27], that many of them are still experimentally unverified. In that sense, the situation is very similar to the mid-1980s, where schematic mode-coupling models were derived, but with little experimental confirmation of their main features.

We have mentioned at several points in 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 necessary, we have shown that a qualitative understanding of the rheological behaviour could be gained via this approach, beyond the steady rheological situation studied in [7].

A more quantitative approach would be to derive a 'full' nonequilibrium theory for sheared fluids. This programme was recently undertaken by Fuchs and Cates [8]. 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 the fact that nonequilibrium effects (like the violation of the fluctuation–dissipation theorem) are not treated by this alternative derivation. Comparisons between the two approaches would thus be very interesting.

Lastly, 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 us to gain any insight into the problem of the selection between the two solutions. This certainly requires more ambitious approaches.

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