

# Age-dependent transient shear banding in soft glasses

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We study numerically the formation of long-lived transient shear bands during shear startup within two models of soft glasses (a simple fluidity model and an adapted ‘soft glassy rheology’ model). The degree and duration of banding depends strongly on the applied shear rate, and on sample age before shearing. In both models the ultimate steady flow state is homogeneous at all shear rates, consistent with the underlying constitutive curve being monotonic. However, particularly in the SGR case, the transient bands can be extremely long lived. The banding instability is neither ‘purely viscous’ nor ‘purely elastic’ in origin, but is closely associated with stress overshoot in startup flow.

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Many soft materials show shear banding: the separation into layers of different shear rate under imposed flow. Examples include wormlike micelle solutions [1]; granular matter [2]; star polymers [3]; and ‘soft glasses’ such as gels, pastes and emulsions [4]. Because shear banding dramatically alters the stress response at fixed shear rate or vice versa, it is central to the rheological control of such materials, whose applications range from foodstuffs and pharmaceuticals to paints and well-bore fluids. For materials that are nonergodic at rest, unexpected complexity can arise when one band is not flowing and thus subject to aging [3–6], while the other is continuously rejuvenated by flow [7]. This interplay between glassy dynamics and shear banding brings together two major current areas of nonequilibrium physics research.

A steady mean shear rate is imposed by choosing time-independent wall velocities in a rheometric device. In most reported cases, the resulting shear bands then persist indefinitely [3–5, 8–12]. In many instances, shear banding is attributable to a nonmonotonic steady-state constitutive curve  $\Sigma(\dot{\gamma})$ , where  $\Sigma$  is the shear stress and  $\dot{\gamma}$  the shear rate in the homogeneous material; regions with  $d\Sigma/d\dot{\gamma} < 0$  are mechanically unstable [1]. This is analogous to, but distinct from, a similar instability in nonlinear elastic solids, where mechanical instability likewise arises if the stress is a decreasing function of strain ( $d\Sigma/d\gamma < 0$ ); see [13]. For simplicity we refer to these as ‘viscous’ and ‘elastic’ banding scenarios respectively.

In this Letter we use the case of soft glasses to explore theoretically a distinctive, third scenario. This is where shear banding arises on startup of steady shearing, before eventually relaxing to an unbanded steady state. The resulting transient bands can be extremely long-lived and so might well be mistaken for true steady-state ones (see, e.g. [14]). We show that such bands can arise in systems *showing neither a viscous nor an elastic instability*, such as a model combining essentially linear elasticity with a near-trivial constitutive curve,  $\Sigma(\dot{\gamma}) = A + B\dot{\gamma}$ . Our results may thus prove highly relevant (alongside other

factors [15, 16]) to a number of cases where apparently steady shear banding is seen, even though the constitutive curve is predicted, by well founded theories, to remain monotonic [16–18]. They are also relevant to recent experiments where long-lived transient shear bands are directly reported [21].

We argue that transient banding behavior can arise generically in systems where the stress response  $\Sigma(t, \dot{\gamma})$  to startup of steady shear shows, as a function of time, a significant overshoot before falling to the steady-state limiting value  $\Sigma(\infty, \dot{\gamma}) \equiv \Sigma(\dot{\gamma})$ . Indeed, the simplest case is where the overshoot is created purely by nonlinear elasticity without relaxation or plastic flow. In this case,  $\gamma(t) = \dot{\gamma}t$  is an elastic strain, so that any region where  $\partial_t \Sigma(t, \dot{\gamma}) < 0$  implies the onset of the elastic instability referred to already. In reality however, soft glasses (in contrast to, e.g., elastomers [13]) have a very limited elastic deformation regime before they yield irreversibly; the stress maximum in such cases is the result of a contest between the growth of elastic stress and its decay by plastic rearrangement towards the eventual steady-state limit. Accordingly  $\gamma(t)$  is not an elastic strain within the region where  $\partial_t \Sigma < 0$ , and there can be no direct mapping onto an elastic banding scenario.

Below we show that transient shear banding does nonetheless arise, whenever the stress overshoot becomes large, both in a fluidity model of soft glasses (chosen for its tractability), and in a mesoscopic model, adapted from that of [22] which is known to capture well some subtler physics of these systems. In soft glasses the overshoot can be varied in height (*without* also varying  $\Sigma(\dot{\gamma})$ ) purely by changing the age of the system [22], making such materials ideal testing grounds for experiments and theory on transient banding. Emerging as it does from two independent models, transient shear banding should be a generic feature in the rheology of well-aged soft glasses – a fact not appreciated previously. Moreover, stress overshoots often arise in other types of soft matter, and in many of these, similar mechanisms may be at work.

*Fluidity model:* Our first model is an empirical fluidity model, along the lines of [19, 20], in which a single structural relaxation time  $\tau$  regresses continuously towards a steady-state value determined by the local shear rate. A finite diffusivity for  $\tau$  is also introduced, so as to prevent band interfaces from becoming infinitely sharp. This model shows transient banding in the overshoot region, with a strong dependence on system age, which sets the initial value of  $\tau$  and thus the overshoot height. However the lifetime of the bands apparently remains modest, regardless of the system age before shearing begins, in contrast to the adapted SGR model considered later.

To set up the fluidity model we decompose the total shear stress  $\Sigma$  into a viscoelastic stress  $\sigma$  arising from the glassy degrees of freedom and a purely viscous part:  $\Sigma = \sigma + \eta\dot{\gamma}$ . We assume translational invariance in the  $x$  (flow) and  $z$  (vorticity) directions but allow nonuniformity to develop in the flow gradient direction,  $y$ . Force balance at zero Reynolds number (neglecting inertia) then requires  $\Sigma$  to be independent of  $y$ . We suppose a Maxwell-type constitutive equation for the viscoelastic stress

$$\partial_t \sigma = G\dot{\gamma} - \sigma/\tau \quad (1)$$

where  $G$  is an elastic modulus and  $\tau$  is the structural relaxation time (inverse fluidity) with its own dynamics:

$$\partial_t \tau = 1 - \tau/\tilde{\tau}(\dot{\gamma}) + l_o^2 \partial_y^2 \tau \quad (2)$$

In the absence of flow this represents simple aging ( $\tau = t$ ) but with flow present, aging is cut off at the inverse strain rate. (This is characteristic of strain-induced plasticity [7].) Here  $l_o$  is a mesoscopic length (which could depend on  $\dot{\gamma}$  without affecting our presentation) describing the tendency for the relaxation time of a mesoscopic region to equalise with those of its neighbors.

We choose the steady-state relaxation time as  $\tilde{\tau} = \tau_0 + \lambda/|\dot{\gamma}|$ , so that in steady state  $\Sigma = \sigma + \eta\dot{\gamma}$  with  $\sigma = G\dot{\gamma}\tilde{\tau} = G\lambda + G\tau_0\dot{\gamma}$ , has a yield stress beyond which it is trivially monotonic (a Bingham fluid). We consider flow between infinite flat parallel plates at  $y = 0, L_y$ , and rescale strain, stress, time and length so that  $\lambda = G = \tau_0 = L_y = 1$ . The steady-state constitutive curve is then

$$\Sigma(\dot{\gamma}) = 1 + (1 + \eta)\dot{\gamma} \quad (3)$$

where for simplicity we now set  $\eta = 0.05$ .

We study a shear startup protocol defined as follows. First imagine preparing the sample by a deep quench at time  $t = 0$ , which we assume results in a fully rejuvenated initial state with  $\tau(y, t = 0) = 1, \sigma(y, t = 0) = 0$  across the whole sample. Next we allow the sample to age at rest (so  $\dot{\tau} = 1$ ) until a time  $t_w$ , before setting the upper plate moving along  $x$  with constant speed  $\bar{\gamma}L_y$ . This defines the average imposed shear rate  $\bar{\gamma} = \int_0^1 \dot{\gamma}(y, t) dy$ .

Transient stress curves  $\Sigma(t, \dot{\gamma})$  for shear startup in a homogeneous system are shown for several different  $t_w$

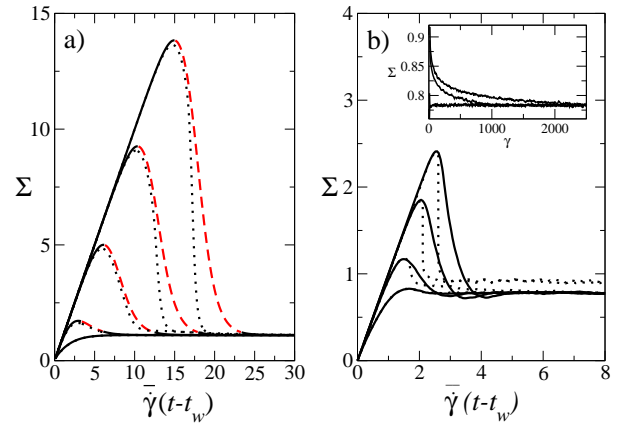


FIG. 1: (a) Homogeneous startup transient in fluidity model at applied shear rate  $\bar{\gamma} = 0.1$  for waiting times  $t_w = 10^0, 10^2, 10^4, 10^6, 10^8$  ( $t_w$  increasing with increasing size of overshoot) showing stable region (solid), unstable (dashed). Dotted curve: actual startup curve arising from inhomogeneous flow with  $l_0 = 0.01$  and  $\delta = 0.01$ . (b) Counterpart in modified SGR model for  $x = 0.3, \bar{\gamma} = 0.1, w = 0.05, n = 50, m = 300$ , and waiting times  $t_w = 10^0, 10^2, 10^4, 10^6$ . Solid lines: banding disallowed. Dotted: banding allowed. Inset: approach to steady state at later times in the banding case.

in Fig. 1a. There is an overshoot that depends strongly on the age  $t_w$  of the sample; one may show that it occurs at a strain  $\gamma_o = \dot{\gamma}(t - t_w)$  given by  $\gamma_o \exp(\gamma_o) = \dot{\gamma}t_w$ . To within a logarithmic correction this gives  $\gamma_o = \log(\dot{\gamma}t_w)$ , which we use for convenience below in preference to the full implicit expression. The response of the sample prior to (but not beyond) the stress maximum is almost elastic, so the peak stress obeys  $\Sigma_o \approx \gamma_o$ .

To gain intuition for the consequences of the overshoot, consider now a thought experiment in which one tracks this homogeneous stress transient for several identical sample replicas, each subject to a different shear rate. At any fixed time interval  $t - t_w$  this defines an ‘instantaneous constitutive curve’  $\tilde{\Sigma}(\dot{\gamma})$ , with the tilde denoting a dependence on  $t - t_w$  and  $t_w$ , suppressed for clarity of notation. As shown in Fig. 2(a), each such curve is non-monotonic over a typical shear rate window  $\dot{\gamma} < \gamma_o/(t - t_w)$ , with monotonicity being restored at higher shear rates. This can be understood as follows. At any fixed time interval  $t - t_w$ , those replicas at high shear rate  $\dot{\gamma}(t - t_w) > \gamma_o$  are expected to have reached steady state, with stresses obeying the constitutive curve, Eq.(3). In contrast those for which  $\dot{\gamma}(t - t_w) < \gamma_o$  will still be on the elastic branch of the stress transient, with a stress  $\sigma = \dot{\gamma}(t - t_w) \gg \Sigma(\dot{\gamma})$ . With this transient non-monotonicity in mind, we performed an instantaneous linear stability analysis about the evolving homogeneous flow, determining for each  $t - t_w$  an instantaneous eigenvalue whose positivity indicates instability to the onset of banding. The unstable windows are shown by dashed lines in Fig. 1a, and correspond broadly to the regions

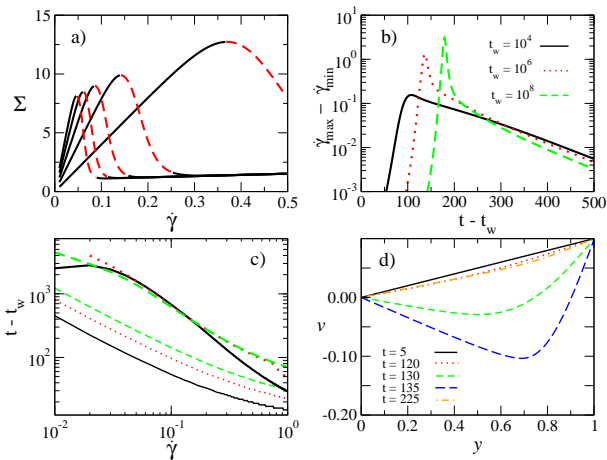


FIG. 2: (a) For chosen  $t_w = 10^6$ , plots of the instantaneous homogeneous constitutive curves during start up for  $t - t_w = 40, 80, 120, 160, 200$  with regions of instability dashed. (b) Strength of banding ( $\dot{\gamma}_{\max} - \dot{\gamma}_{\min}$ ) against  $t - t_w$  for  $\bar{\gamma} = 0.1$ , and  $t_w = 10^4, 10^6, 10^8$ . (c) For the same  $\bar{\gamma} = 0.1$ , and  $t_w = 10^4$  (solid),  $10^6$  (dotted),  $10^8$  (dashed), plots in the  $t - t_w, \dot{\gamma}$  plane showing the upper (thick lines) and lower (thin lines) boundary curves of the window in which banding is significant (threshold set by  $(\dot{\gamma}_{\max} - \dot{\gamma}_{\min})/\bar{\gamma} > 0.01$ ). (d) Snapshots of banded profiles for  $\dot{\gamma} = 0.1$ ,  $t_w = 10^6$ , and  $t - t_w = 5, 120, 130, 135, 225$  showing onset, development and eventual collapse of the bands.  $l_0 = 0.01$ ,  $\delta = 0.01$  in (b-d).

of negative slope in  $\tilde{\Sigma}(\dot{\gamma})$ . However, they do not do so exactly; this cannot be viewed solely as a ‘viscous’ instability of the instantaneous constitutive curve. Nor does the window exactly correspond to that of negative slope in  $\Sigma(t, \dot{\gamma})$ , as it would for an ‘elastic’ instability. Although clearly associated with a large stress overshoot, the banding scenario found here is thus distinct from either the viscous or the elastic one – a view reinforced by the linearity of the step strain response in (1), and the trivial monotonicity in (3), for the underlying model.

We now turn to study the full heterogeneous dynamics of the model in this shear startup protocol, allowing spatial variations in the flow gradient direction  $y$ . In each run we add a small perturbation  $\sigma(y, t = 0) = \delta \cos(\pi y)$  where  $\delta \ll 1$ , in order to trigger any banding. In each run we tracked, as a function of shearing time  $t - t_w$ , the degree of shear banding in the sample, as measured at any instant by the difference  $\dot{\gamma}_{\max} - \dot{\gamma}_{\min}$  between the maximum and minimum shear rates present in the cell, as shown in Fig. 2(b) for several different waiting times  $t_w$ . Regions of high  $\dot{\gamma}_{\max} - \dot{\gamma}_{\min}$  broadly match up with the regions of negative slope in Fig. 2(a), consistent with the idea that this instability in the instantaneous constitutive curve indeed triggers banding. Likewise in Fig. 2(c) we delineate the temporal windows, for various  $t_w$  and as a function of shear rate  $\bar{\gamma}$ , within which banding is significant, as defined by a criterion  $(\dot{\gamma}_{\max} - \dot{\gamma}_{\min})/\bar{\gamma} > 0.01$ . The shear stress that arises in these transiently banded

flows is of course different from the predictions of the homogeneous model; both are shown in Fig. 1a, as a function of  $t - t_w$ , at a fixed shear rate  $\bar{\gamma} = 0.1$  for various waiting times. Snapshots of the transient bands for one particular startup are shown in Fig. 2 (d). Note that the minimum shear rate, which arises within the low shear-rate band, can be negative during transient banding. This is because the low shear-rate band is well-aged, has been subjected only to a weak flow, and therefore remains essentially an elastic slab. However the stress  $\Sigma$  on this slab decreases post-overshoot: like any elastic solid being unloaded, it shears backwards.

*Adapted SGR model:* Our second model is a spatially resolved adaptation of the SGR model [6, 22] in which a spectrum of jump rates describes hopping over strain-modulated local rearrangement barriers at an effective noise temperature  $x = T/T_g$ ; elastic strain builds up and then is released in these plastic jump events. Following Ref. [6], where full details can be found, numerically we take  $j = 1 \dots m$  SGR elements on each of  $i = 1 \dots n$  streamlines, corresponding to  $y = 0 \dots 1$ , with periodic conditions. The stress on streamline  $i$  is  $\sigma_i = (k/m) \sum_j \ell_{ij}$ , with elastic constant  $k = 1$ . A waiting time Monte Carlo algorithm is used to choose stochastically the next element to jump. Supposing the jump occurs at element  $ij$  when its local strain is  $\ell = l$ , force balance is then imposed by updating all elements on the same streamline as  $\ell \rightarrow \ell + l/m$ , and further updating all elements throughout the system as  $\ell \rightarrow \ell - l/mn$ . In contrast to Ref. [6], here we take the noise temperature  $x$  as constant, but include instead a stochastic jump-induced straining of elements on neighboring streamlines after each jump event: further adjusting the strain of three randomly chosen elements on each adjacent streamline  $i \pm 1$  by  $lw(-1, +2, -1)$ . This creates a diffusive coupling of the dynamics on different streamlines (different  $y$ ), analogous to that in Eq.(2), with a strength set by  $w$ . It also mildly alters the constitutive curve from that of pure SGR, without losing monotonicity.

The evolution of stress with time in startup flow is shown in Fig. 1b, both for the case where homogeneous flow is imposed (solid lines), and for the full dynamics allowing banding (dotted lines). The trends are quite similar to those from the scalar model: for older samples, the dotted line departs from the solid one just after the overshoot. The initial effect of transient banding, as before, is always to decrease the stress to values below that of the homogeneous system at the same time point.

There is however a new feature in the SGR model, not seen in the fluidity model. For the oldest samples ( $t_w \sim 10^6$ , measured in units of the microscopic attempt rate for jumps) the time scale for the stress signal to decay to the limiting value (corresponding to homogeneous flow) is inordinately long: indeed, this decay can require strains  $\bar{\gamma}t$  of order thousands, as opposed to the order-unity values seen in the fluidity model. Since strain re-

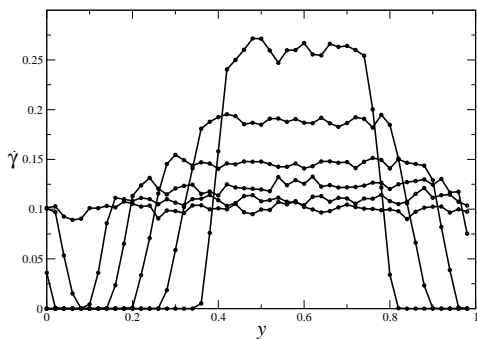


FIG. 3: Shear rate profiles corresponding to slowly decaying stress signal at  $t_w = 10^6$  in Fig. 1b averaged over time windows 250 – 500; 500 – 1000; 1000 – 1500; 1500 – 2000; 2000 – 2500; 2500 – 3000 (top to bottom at  $y = 0.5$ ).

juvenates the material, this is possible only because, for very old samples, the strain rate in the low-shear band remains extremely small compared to  $\bar{\gamma}$ . This behavior, clearly visible in the banding profiles presented in Fig. 3, is consistent with having an age-dependent static yield stress that can lie well above the homogeneous steady state stress  $\Sigma(\bar{\gamma})$ . In principle the banded state might then persist indefinitely, but in our model the low shear band seemingly is eroded slowly by the spreading of the fast band, perhaps as a result of the diffusive nonlocality in the jump dynamics.

*Discussion:* Aside from its longer-lived transients, the adapted SGR model shows broadly similar phenomenology to the fluidity model, supporting our contention that the transient shear-banding scenario reported here is generic for soft glasses. As previously stated, the soft glass case is one where the stress overshoot stems from a competition between (essentially linear) elasticity and plastic relaxation: there is no facile mapping onto either an elastic instability ( $d\Sigma/d\gamma < 0$ ) or a viscous one ( $d\Sigma/d\dot{\gamma} < 0$ ). Despite the intermediate character of the instability towards transient bands, we showed for the fluidity model that their onset is closely correlated with the occurrence a negative slope on the *instantaneous* stress versus strain rate curve ( $\partial\Sigma/\partial\dot{\gamma}|_t < 0$ ). However, linearization about the time-dependent solution for a homogeneous flow showed this to be a qualitative rather than an exact correspondence.

The scenario of transient bands we have developed for soft glasses may interact in a complicated manner with various mechanisms previously presented to explain steady-state shear bands in the same class of materials [6, 16]. However, in some cases bands of long but finite duration were already observed in startup flows [21] and the physics we have explored in this letter may be enough, on its own, to explain some of these. Even if not, the presence of a generic connection between transient banding and stress overshoots should not be overlooked in future work on these materials.

Stress overshoots in startup flows are often also seen

in other classes of viscoelastic soft matter at high enough shear rates, including non-aging systems such as entangled polymers. Indeed, some studies of polymeric materials and models have started to explore the connection between this and transient banding [17, 18]. Nonetheless, we hope that further experimental and theoretical work on soft glasses will help elucidate this connection in more general terms. Arguably these represent ideal materials with which to explore the problem, because the size of the overshoot can be varied without any change to the final steady state of the system. In all systems both the final state and the overshoot depend on the imposed flow rate and on sample composition, temperature etc.; but in glasses one can also vary the system age  $t_w$  which affects only the overshoot, and not the steady state.

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