From ductile to brittle yielding in amorphous materials

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We study theoretically the yielding of sheared amorphous materials as a function of increasing levels of initial sample annealing prior to shear, in two widely used constitutive models and three widely studied annealing protocols. In each case we demonstrate a gradual progression, with increasing annealing, from smoothly “ductile” yielding, in which the sample remains homogeneous, to abruptly “brittle” yielding, in which the sample becomes strongly shear banded. We show that this arises from an increase with annealing in the size of an overshoot in the underlying stress-strain curve for homogeneous shear. This in turn causes a shear banding instability that becomes ever more severe with increasing annealing. “Ductile” and “brittle” yielding thereby emerge as two limiting cases of a continuum of yielding transitions, from smoothly gradual to abruptly catastrophic.

Amorphous materials include soft glasses such as dense colloids, emulsions, foams and microgels [1–5], as well as hard molecular and metallic glasses [6, 7]. Under low loads or small deformations, such materials show solid-like behaviour. Under higher loads or larger deformations, they yield plastically. For soft glasses, the dynamical process whereby an initially solid-like sample yields to give a finally fluidised flow is typically rather smooth and gradual [8–16]. Hard glasses instead typically yield abruptly, with catastrophic sample failure [17]. For both “ductile” materials, which yield smoothly and gradually, and “brittle” materials, which yield abruptly and catastrophically, understanding the statistical physics of yielding is the focus of intense interest. Theories have been put forward based on a first order transition in a replica theory [18, 19]; a critical point in an elastoplastic model [20, 21]; a directed percolation transition [22, 23]; and a spinodal [24, 25]. Microscopic precursors to yielding have recently been observed in soft materials [26–28].

Recently, Ozawa et al. [29] suggested that the underlying stress-strain relation for an amorphous material undergoing homogeneous shear, $\Sigma(\gamma)$, displays a qualitative change of form from the lower to the upper solid curve in Fig. 1 (left) as the degree to which a sample is annealed prior to shear increases. Poorly annealed samples (lower curve) then yield in a smoothly “ductile” way. Well annealed samples (upper curve) instead show catastrophic “brittle” yielding, as the stress drops precipitously once the critical overhang strain is reached (dashed line). In this scenario, “ductile” and “brittle” yielding are separated by a random critical point, at which the stress-strain curve switches between two qualitatively different shapes with increasing annealing.

In this Letter we propose an alternative scenario, in which the underlying stress-strain curve for homogeneous shear has an overshoot, rather than an overhang, followed by a regime of negative slope, $\partial_\gamma \Sigma < 0$. A state of initially homogeneous shear then becomes linearly unstable to the formation of shear bands in this negatively sloping regime [30–32]. We show in particular that the severity of this banding instability, as quantified by its eigenvalue, scales as $\omega \sim -\partial_\gamma \Sigma / \dot{\gamma}$, and therefore increases without bound as the height of the stress overshoot increases with increasing levels of sample annealing prior to shear. Severe banding in strongly annealed samples then causes the measured stress-strain curve to drop precipitously from the underlying homogeneous curve, leading to abrupt “brittle” yielding: Fig. 1 (right).

In this scenario, “ductile” and “brittle” yielding emerge naturally as limiting cases in a continuum of yielding transitions, with gradually more severe shear banding, from negligible to dramatic, as the degree of initial annealing increases. Crucially, this picture does not invoke a critical point separating the two types of yielding. We demonstrate this scenario in two widely used models of amorphous materials: a continuum model [33], and a mesoscopic elastoplastic model [34].

For consistency with the vocabulary adopted in Ref. [29], we use the term “brittle” to characterise abrupt yielding, but with a caveat that we quote from [29]: “Although this phenomenon is not accompanied by the formation of regions of vacuum, as it happens in the fracture of brittle materials, the macroscopic avalanche taking place at the discontinuous yielding transition does resemble a crack induced by a brittle fracture”. Indeed, our calculations and those of Ref. [29] are performed at fixed volume, disallowing the opening of an air gap. We suggest, however, that the formation of a severe high shear

![Fig. 1. Schematic of shear stress versus strain in two possible scenarios for ductile versus brittle yielding: (left) as suggested in Ref. [29] and (right) as suggested here. In each case the upper(lower) curve is for a better(less well) annealed sample, leading to “brittle” (“ductile”) yielding. Solid line: theoretical curve in which homogeneous shear is artificially enforced. Dotted line: precipitous drop from stress overhang (left), or drop from homogenous curve due to shear banding (right).](image-url)
band in abrupt yielding will, in studies at fixed pressure, indeed lead to the rapid opening of an air gap.

We consider incompressible, inertialess deformations of a slab of material sheared between infinite flat parallel plates at $y = 0, L_y$. The shear is imposed by moving the top plate relative to the bottom one at a speed $\bar{\gamma}L_y$ in the positive $\mathbf{x}$ direction. As is standard practice, we restrict all velocities $v(y,t)$ to the main flow direction $\mathbf{x}$, and all gradients to the direction $\mathbf{y}$, defining the shear rate $\dot{\gamma}(y,t) = \partial_y v(y,t)$, which may vary across $y$ due to shear banding. The spatially average imposed shear rate $\bar{\gamma} = \int_0^{L_y} dy \dot{\gamma}(y,t)/L_y$. We track only the shear component of the stress, $\Sigma_{xy}(t)$. The condition of force balance requires $\partial_y \Sigma_{xy} = 0$. The total shear stress in any fluid element is assumed to comprise an elastoplastic contribution $\sigma_{xy}(y,t)$, and a Newtonian solvent contribution of viscosity $\eta$, such that $\Sigma_{xy}(t) = \sigma_{xy}(t) + \eta \dot{\gamma}$. Hereafter we drop the $xy$ subscript for clarity. For the dynamics of the elastoplastic stress $\sigma$, we shall consider two different constitutive models: a continuum model (Model I) [33], and a mesoscopic elastoplastic model (Model II) [34].

Model I is a highly simplified continuum model [33] that supposes a Maxwell-type constitutive equation for the viscoelastic stress:

$$\partial_t \sigma(y,t) = G\dot{\gamma} - \sigma/\tau.$$  

Here $G$ is a constant modulus and $\tau$ is a stress relaxation time, which has its own dynamics:

$$\partial_t \tau(y,t) = 1 - \frac{|\dot{\gamma}|\tau}{1 + |\dot{\gamma}|\tau_0} + \frac{\dot{\gamma}^2}{\tau_0} \partial^2 \tau.$$  

(2)

The first term on the RHS captures ageing, in which the timescale for stress relaxation increases linearly with the time $t_w$ for which a sample is aged (or “annealed”) before any deformation commences. The second term captures rejuvenation by deformation. The parameter $L_y$ in Eqn. 2 is a mesoscopic length describing the tendency for the relaxation time of a mesoscopic region to equalise with its neighbours.

Model II [34] instead considers an ensemble of elastoplastic elements, each of which corresponds to a local mesoscopic region of material. Given a shear rate $\dot{\gamma}$, each element builds up a local elastic shear strain $\dot{\gamma}$ according to $\dot{\gamma} = \dot{\gamma}_e$, giving a shear stress $G\dot{\gamma}$, where $G$ is a constant modulus. This stress is intermittently released by local plastic yielding events, each modelled as hopping of an element over a strain-modulated energy barrier $E$, governed by a noise temperature $x$, with a stochastic yielding rate $\tau_0^{-1} \min \{1, \exp[-(E - \frac{1}{2}kT)/x]\}$. Upon yielding, any element resets its local stress to zero and chooses its new energy barrier from an exponential distribution $\exp(-E/x)$. This results in a broad spectrum of yielding times, $P(\tau)$, and a glass phase for $x < x_g$, in which a sample shows ageing before deformation commences.

Non-uniform shear deformations are accounted for by taking $n = 1...N$ streamlines at discretised flow-gradient positions $y = 0...L_y$. Given an imposed average shear rate $\bar{\gamma}$ across the sample as a whole, the shear rate on each streamline $n$ is calculated by enforcing force balance: $\dot{\gamma}_n = \bar{\gamma} + (\bar{\sigma} - \sigma_n)/\eta$, where $\bar{\sigma} = (1/N) \sum_n \sigma_n$. The elastoplastic stress $\sigma_n$ on streamline $n$ obeys Eqn. 1 in Model I. In Model II, we take $M$ elastoplastic elements on each streamline, with $\sigma_n = (G/M) \sum_{m=1}^{M} \sigma_{nm}$. Adjacent streamlines are weakly coupled via the diffusion term of Eqn. 2 in Model I; and in Model II by adjusting the stress of three randomly chosen elements on each adjacent streamline by an amount $\omega(-1, +2, -1)$ following any yielding event, with $\omega$ a small parameter. To seed the bands, in Model I we add noise as $\sigma(y,t+Dt) = \sigma(y,t) + \eta \sqrt{D \delta} \cos(\pi y/L_y)$, with $r$ chosen from a top hat distribution between $-0.5$ and $+0.5$, and $\delta$ a small number. In Model II we slightly perturb the trap depths prior to shear as $E \rightarrow E[1 + \delta \sin(2\pi y/L_y)]$. We adopt closed and periodic boundary conditions in Models I and II respectively. The use of different boundary conditions and seeding in the two models verifies the scenario we report is robust to these choices.

Within both models, we consider a sample prepared at time $t = 0$ in a rejuvenated initial state with zero stress, $\Sigma(y,t=0) = 0$, then left to age undisturbed until a time $t_w$, after which it is sheared at a constant imposed shear rate $\bar{\gamma}$. As a function of the subsequently accumulating strain, $\dot{\gamma}(t) = \bar{\gamma}(t-t_w)$, we track the shear stress $\Sigma(\dot{\gamma})$. We first perform calculations imposing that the shear must remain homogeneous across the sample, $\dot{\gamma}(y,t) = \bar{\gamma}$. In separate calculations we then allow shear bands to form, $\dot{\gamma}(y,t)$, and track the degree of shear banding $B(\dot{\gamma})$, defined as the difference at any time $t$, or strain $\dot{\gamma}(t)$, between the maximum and minimum local strain rates across the sample (Model I) or the standard deviation in $\dot{\gamma}$ across $y$ (Model II), normalised by $\bar{\gamma}$.

We rescale strain, stress, time and length so that $x_g = \bar{\gamma}L_y$.
FIG. 3. Velocity profiles at the times (marked by crosses) of maximal shear banding, for the four stress-strain curves with the largest values of $t_w$ in Fig. 2.

$G = \tau_0 = L_y = 1$. The solvent viscosity $\eta \ll G\tau_0 = 1$ is unimportant to the physics we describe. We have checked for robustness with respect to variations in this value between $\eta = 0.01$ and 0.05.

We now present our results. Fig. 2 shows as solid lines the stress versus strain curves $\Sigma(\tilde{\gamma})$ calculated within Model I, imposing that the shear must remain homogeneous across the sample, for several values of the sample age $t_w$ at a single value of the imposed shear rate $\tilde{\gamma}$. Each curve shows an initially solid-like elastic regime in which the stress increases linearly with strain. At late times (large strains), in contrast, the sample flows plasticity with a constant value of the shear stress. For intermediate times (and strains), the stress shows an overshoot that increases in amplitude with increasing degree of sample annealing prior to shear (increasing $t_w$): an older sample shows a larger initial regime of solid-like plastic response before yielding into a finally flowing state.

We then perform separate calculations in which shear bands are allowed to form. The resulting stress-strain curves are shown by dashed lines in Fig. 2. For poorly annealed samples, each curve still follows that of the corresponding homogeneous calculation, to excellent approximation, indicating that the shear field remains homogeneous (or nearly so), with yielding occurring in a smoothly gradual (“ductile”) way. For well annealed samples, in contrast, the stress-strain curve of the heterogeneous calculation drops precipitously below that of the homogeneous one as the sample becomes strongly shear banded, causing abrupt (“brittle”) yielding. Velocity profiles for the best annealed samples indeed show stronger banding with increasing annealing (Fig. 3).

So far, we have explored several annealing times $t_w$ at a single imposed shear rate $\tilde{\gamma}$, as indicated by crosses in Fig. 4. We now explore the $\tilde{\gamma}, t_w$ plane more fully. For each individual deformation experiment, as defined by a coordinate pair $(\tilde{\gamma}, t_w)$, we denote by $S_{\text{max}}$ the absolute value of the maximally negative value of $\partial\gamma \Sigma$, maximised over all times during the deformation. This quantifies the abruptness of the stress drop during yielding. We further denote by $B_{\text{max}}$ the degree of shear banding maximised over all times during the deformation. This quantifies the severity of shear banding during yielding. See Fig. 4.

For any $\tilde{\gamma}$, a regime of gradual yielding (low $S_{\text{max}}$) and near homogeneous deformation (low $B_{\text{max}}$) for poorly annealed samples (low $t_w$) crosses over into a regime of precipitous yielding (high $S_{\text{max}}$) and strong banding (high $B_{\text{max}}$) for well annealed samples (high $t_w$). For $\tilde{\gamma} \ll 1$, this crossover occurs at $\tilde{\gamma} \approx 10^m/t_w$ with $m \approx 4.5$. Deviations from this scaling at higher $\tilde{\gamma}$ should be disregarded, because Model I is itself only valid for $\tilde{\gamma} \ll 1$. Increasing strain localisation with decreasing initial ‘effective temperature’ was seen in the STZ model in Ref. [32].

To understand these results, we cast Model I in a more general form, writing $\Sigma(t) = \sigma(y,t) + \eta \tilde{\gamma}(y,t)$, $\sigma(y,t) = f(\tilde{\gamma}, \sigma, \tau)$ and $\tilde{\gamma}(y,t) = g(\tilde{\gamma}, \sigma, \tau)$. To consider whether shear bands will form during any deformation experiment, we write the system’s state as the sum of a homogeneous part plus an (initially) small heterogeneous perturbation that is the precursor to any shear bands: $\gamma(y,t) = \tilde{\gamma} + \delta \gamma(t) \exp(ik y)$, $\sigma(y,t) = \tilde{\sigma} + \delta \sigma(t) \exp(ik y)$, $\tau(y,t) = \tilde{\tau} + \delta \tau(t) \exp(ik y)$. Substituting these into the model equations and expanding to first order in the amplitude of the heterogeneous perturbations, we find that for $\eta \ll 1, k l_0 \ll 1, \tilde{\gamma} \ll 1$ the shear rate heterogeneity evolves as $d \log(\delta \gamma)/dt = \omega + d \log(-f/\sigma)/d\tau$. The first term, $\omega = -g_1 f_1 f_2 + g_1 \gamma$, is an eigenvalue which, when positive, indicates an instability to the growth of heterogeneous shear bands. The second term represents a time-dependent spinning of the eigenvector $(d \tau, d \gamma)$.

For any model that is of Maxwell-like form, $f = \gamma - \sigma/\tau$, and in which the $\tau$ dynamics are such that $g_1 = -\tau, g_2 = -\gamma$, it can be shown that the eigenvalue $\omega = -\partial_{\Sigma}$. At the level of this linear calculation, the degree to which shear heterogeneity grows will scale as the time-integral of $\omega$ over the time interval when $\omega > 0$, and accordingly as the size of the stress drop from overshoot.
In this work, we have studied the shear-induced yielding of amorphous materials as a function of increasing levels of initial sample annealing prior to shear, in two widely used constitutive models and three widely studied annealing protocols. In each case, we have demonstrated a gradual progression, with increasing levels of annealing, from smoothly “ductile” yielding, in which the sample remains homogeneous, to abruptly “brittle” yielding, in which the sample becomes strongly shear banded. We have further shown that this arises from an increase with annealing in the size of an overshoot in the underlying curve of stress as a function of strain for homogeneous shear. This in turn leads to a shear banding instability that becomes ever more severe with increasing annealing. In this way, “ductile” and “brittle” yielding form two limiting cases on a continuum of yielding transitions, from smoothly “ductile” yielding, in which the sample remains homogeneous, to abruptly “brittle” yielding, in which the sample becomes strongly shear banded. We find the same progression in the stress-strain curves as in Fig. 5 for increasing values of the ageing time, $t_w$, from smooth and homogeneous yielding for poor annealing to abrupt and strongly shear banded yielding for strong annealing.

To show that the scenario we report is robust to the annealing protocol used, we finally consider within Model II two different annealing protocols. Protocol B consists of equilibrating the sample to a high initial temperature $x = x_0 = 5.0 > x_g$, then cooling to zero temperature as $x(t) = x_0 - \alpha t$, before shearing at a rate $\dot{\gamma}$. In this case, a slower cooling rate $\alpha$ corresponds to a better annealed sample. Protocol C consists of equilibrating the sample to an initial temperature $x_0 > x_g$, then at some time $t = 0$ suddenly jumping the temperature to $x = 0$, before shearing at a rate $\dot{\gamma}$. In this case, a lower initial equilibration temperature $x_0$ corresponds to a better annealed sample. Figs. 1 and 2 of the Supplementary Information (below) show results for Protocols B and C respectively. For decreasing values of the cooling rate $\alpha$ in Protocol B or of equilibration temperature $x_0$ in Protocol C, we find the same progression in the stress-strain curves as in Fig. 5 for increasing values of the ageing time, $t_w$, from smooth and homogeneous yielding for poor annealing to abrupt and strongly shear banded yielding for strong annealing.

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Supplemental Material

In the main text, we considered a protocol in which a sample was freshly prepared at time $t = 0$ at a noise temperature $x_0 < x_g = 1$, then allowed to age undisturbed for a time $t_w$ before being sheared at rate $\dot{\gamma}$. We now call this protocol A. To show that the scenario argued for in this work is robust to the annealing protocol used, we consider now two other protocols.

Protocol B consists of equilibrating the sample to a high initial temperature $x = x_0 = 5.0 > x_g$, then cooling to zero temperature at a constant cooling rate $\alpha$, such that $x(t) = x_0 - \alpha t$, before shearing at a rate $\dot{\gamma}$. In this case, a slower cooling rate $\alpha$ corresponds to a better annealed sample. Results for this protocol are shown in Fig. 6. For decreasing values of the cooling rate $\alpha$, we find the same progression in the stress-strain curves as in Protocol A for increasing values of the ageing time, $t_w$: from smooth and homogeneous yielding for poor annealing to abrupt and strongly shear banded yielding for strong annealing. The same progression also occurs with decreasing $\dot{\gamma}$ at fixed $\alpha$ (not shown).

Protocol C consists of equilibrating the sample to an initial temperature $x_0 > x_g$, then at some time $t = 0$ suddenly jumping the temperature to $x = 0$, before shearing at a rate $\dot{\gamma}$. In this case, a lower initial equilibration temperature $x_0$ corresponds to a better annealed sample. Results for this protocol are shown in Fig. 7. For decreasing values of the equilibration temperature $x_0$, we find the same progression in the stress-strain curves as in Protocols A for increasing values of the ageing time, $t_w$: from smooth and homogeneous yielding for poor annealing to abrupt and strongly shear banded yielding for strong annealing. The same progression also occurs with decreasing $\dot{\gamma}$ at fixed $x_0$ (not shown).
FIG. 6. **Left:** stress versus strain in Model II in protocol B with homogeneous flow enforced (solid lines) and shear banding allowed (dashed lines) for cooling rates $\alpha = 10^n$ with $n = -3.0, -2.5, -2.0 \ldots 0.5$ in curves right to left. **Right:** (left vertical axis) steepest negative slope in stress versus strain curve with homogeneous flow enforced (dotted line) and allowing banding (solid line); (right vertical axis) maximum degree of shear banding during any deformation simulation as a function of cooling rate. $\sqrt{2} \times \dot{\gamma} = 10^{-3}$, $x = 0.0$, $w = 0.05$, $N = 20$, $M = 80,000$.

FIG. 7. **Left:** stress versus strain in Model II in protocol B with homogeneous flow enforced (solid lines) and shear banding allowed (dashed lines) for annealing temperatures $x_0 = 1.1, 1.25, 1.5, 1.75, 2, 3, 4, 5, 6, 7, 8, 9, 10$ in curves right to left. **Right:** (left vertical axis) steepest negative slope in stress versus strain curve with homogeneous flow enforced (dotted line) and allowing banding (solid line); (right vertical axis) maximum degree of shear banding during any deformation simulation as a function of cooling rate. $\sqrt{2} \times \dot{\gamma} = 10^{-3}$, $x = 0.0$, $w = 0.05$, $N = 20$, $M = 80,000$. 