Formation and growth of shear bands in glasses: existence of an underlying directed percolation transition

Gaurav Prakash Shrivastav,1 Pinaki Chaudhuri,2 and Jürgen Horbach1

1Institut für Theoretische Physik II, Heinrich-Heine-Universität Düsseldorf,
Universitätsstr. 1, 40225 Düsseldorf, Germany
2The Institute of Mathematical Sciences,
CIT Campus, Taramani, Chennai 600 113, India

The response of glasses to mechanical loading often leads to the formation of inhomogeneous flow patterns. Among them, shear bands, associated with strain localization in form of band-like structures, are ubiquitous in a wide variety of materials, ranging from soft matter systems to metallic alloys. They can be precursors to catastrophic failure, implying that a better understanding of the underlying mechanisms of shear banding could lead to the design of smarter materials. Here, molecular dynamics simulations are used to reveal the formation of shear bands in a binary Lennard-Jones glass, subject to a constant strain rate. At a critical strain, this system exhibits for all considered strain rates a transition towards the formation of a percolating cluster of mobile regions. We give evidence that this transition belongs to the universality class of directed percolation. Only at low shear rates, the percolating cluster evolves into a transient (but long-lived) shear band with a diffusive growth of its width.

Introduction. An external shear field leads in general to a rejuvenation of the glass state, transforming the amorphous solid into a flowing fluid [1] [2]. Under a constant strain rate, the transition to plastic flow can be located via the dependence of the shear stress on the applied strain. It is marked by a maximum in the stress-strain relation which for a simple planar Couette flow geometry occurs typically at a strain of the order of 0.1 [2–5]. Beyond this maximum, the system evolves into a steady-state regime where the system displays a homogeneous flow pattern, e.g. in the case of planar Couette flow manifested as a linear velocity profile. However, the strain necessary to reach this steady-state regime depends on many factors such as the history of the initial undeformed glass state and the applied
strain rate [6, 7]. In the transient regime before the steady state is reached, the occurrence of spatially inhomogeneous flow patterns is very common. Often, such inhomogeneous response leads to the formation of shear bands, with co-existing regions of contrasting mobilities spanning large scales [8]. Even when the mechanical loading is switched off, the shear-band regions can be imprinted as frozen-in structures in the resulting solid and thus strongly affect its material properties [9, 10]. Therefore, the microscopic understanding of the formation and growth of shear bands in glassy solids is a highly-debated issue in material science.

Despite the fact that shear banding is ubiquitous in amorphous materials [8, 11–14], the microscopic processes that lead to the formation of such complex structures still remain ill understood. Unlike the case of micellar systems or granular materials, where shear bands mainly form in the regime of large applied shear rates, driven by a coupling between structures and external shear [14–17], the scenario in glasses seems to be different. The inhomogeneous response in the case of amorphous solids tends to be observed at small shear rates, with the spatio-temporal extent becoming more prominent as one approaches the yielding threshold [1, 2, 18, 19]. And, so far, there is no indication of any underlying macroscopic mechanical instability driving the formation of shear bands. While there have been several observations in experiments [20, 21], numerical simulations [22–26] and phenomenological models [27, 28], a quantitative analysis that allows to predict the conditions under which shear bands form in glasses is lacking.

The question about the origin of shear bands is intimately related to the question how flow is initiated in an amorphous solid under applied shear. Recent studies suggest that the response of the glass to the applied strain is governed by local heterogeneities that are either already present in the undeformed solid or form during the initial application of the shear field [1, 2, 4]. These heterogeneities are associated with “hot spots” of higher mobility that grow while the strain of the system increases [29, 30]. For athermal amorphous systems, evidence has been given that the transition towards plastic flow is provided by an avalanching of the mobile hot spots [31, 33]. Imprints of that have also been reported in thermal systems [34, 35]. In fact, such avalanching is well-known in the context of self-organized criticality [36]. A prominent example is the sandpile model [36]. Recently, it has been shown that a driven version of this model belongs to the directed percolation (DP) universality class [37]. In fact, as put forward by the DP conjecture [38, 39], DP universality is believed to be very robust, applying to a broad class of non-equilibrium phase transitions [40].
Thus – although not addressed so far – it is a natural question whether the onset of flow in a sheared glass is linked to a DP transition. In this work, we show that this indeed the case and thereby we elucidate the microscopic mechanism that leads to the formation and growth of shear bands in glassy systems.

Using a model glass forming system, we use large-scale molecular dynamics simulations to study the mechanical response of a quiescent amorphous solid when an external shear rate is imposed on it. We track the locations of regions of large mobilities and thereby we reveal the existence of the DP transition driven in the direction of applied shear. While the DP transition is seen for all shear-rates, visible shearbanding only only for small shear rates near the yielding threshold. We demonstrate that the glass eventually fluidizes by the long-time diffusive invasion of the shear band into the rest of the system, with the diffusion timescales dependent on the imposed shear rates. Thus, we provide a quantitative description of how initial local mobilities build up to the eventual macroscopic flow of the material.

Results. When the shear is applied to the quiescent glass at $t = 0$, the material deforms exhibiting the typical stress ($\sigma$) vs. strain ($\dot{\gamma}t$) response [shown in Fig. 1(a) for different imposed shear rates $\dot{\gamma}$] with the height of the overshoot depending on $\dot{\gamma}$ [6]. The measured steady-state stress as a function of the imposed shear rate is shown in the inset of Fig. 1(a); it has the typical Herschel-Bulkley form [6]. The corresponding single particle dynamics, during this onset of flow, can be quantified by measuring the non-affine mean-squared displacement (MSD), $\Delta r_z^2$, in the direction transverse to the applied shear; the data is shown in Fig. 1(b). The particles undergo ballistic motion at early times and are then caged, before the occurrence of a super-diffusive regime, prior to diffusion. The onset of super-diffusion occurs around the stress overshoot in the stress-strain curve, when the built-up stress is released via the particles breaking their local cages to subsequently diffuse [3].

In experiments, flow heterogeneities are often diagnosed via the spatial profiles of local velocities [21, 42]. Similarly, we measure the spatial profiles of the local flow velocities (averaged over strain intervals of 0.5%), $v_x(z)$, for an imposed shear rate of $10^{-4}$, at different times after the imposition of shear (marked on the corresponding stress-strain curve in Fig. 1(a)). The spatial profiles are shown in Fig. 1(c) for one of the initial states in our ensemble. In the elastic regime, at $\dot{\gamma}t = 0.02$, the velocity profile is linear, but starts deviating from this shape as the stress overshoot is approached at $\dot{\gamma}t = 0.06$. This deviation becomes stronger in the transient regime (at $\dot{\gamma}t = 0.5$, shown in blue diamonds). Interestingly, the
FIG. 1. (a) Stress-strain response of the glass at temperature $T = 0.2$ for an age of $t_w = 10^4$ and sheared with a constant strain rate. Data shown for different shear-rates: $\dot{\gamma} = 10^{-3}, 3 \times 10^{-4}$ and $10^{-4}$. The inset shows the flow curve. Red solid line shows the fitting with Herschel-Bulkley form $\sigma_{x_z}^s(\dot{\gamma}) = 0.3974 + 2.2963 \dot{\gamma}^{0.43}$. (b) Variation of $z$-component of the MSD of large particles with strain. Brown dotted line marks $\mu_{th} (= 0.02)$. (c) Velocity profiles, for $\dot{\gamma} = 10^{-4}$, at five different strain values marked in (a). Green solid line represents the expected linear profile. (d) MSD maps at $\dot{\gamma}t = 0.06$, 0.5 for $\dot{\gamma} = 10^{-4}$. (e) Maps of local strain corresponding to MSD maps shown in (d). (f) Maps of local mobility corresponding to (d). Mobile regions are marked in blue while immobile regions are marked in green.

velocity profile regained its linear shape as plastic flow sets in (at $\dot{\gamma}t = 1.0$, shown in orange stars). The observation of increased heterogeneity, after the stress-overshoot, is consistent with experimental observations [42].

However, local velocity profiles only capture the short-time heterogeneities in dynamics. In order to obtain a more cumulative picture from $t = 0$, we look at the spatially resolved maps of $\Delta r_z^2(t)$ [23 41], by dividing the system into small cells (see Methods for more details). In Fig. 1(d), we show the time-evolution of such a map, for an initial state under the imposed shear rate of $\dot{\gamma} = 10^{-4}$. At a strain of $\dot{\gamma}t = 0.06$, the local dynamics is nearly homogeneous on this scale. However, at $\dot{\gamma}t = 0.5$, spatially heterogeneous dynamics is
FIG. 2. Occurrence of percolation transition. (a) Variation of \( p_{\text{span}} \) with \( p \), in the box of dimension \( 20 \times 20 \times 80 \), for strain rates \( \dot{\gamma} = 10^{-2}, 10^{-3}, 3 \times 10^{-4}, 10^{-4}, 3 \times 10^{-5} \) and \( 10^{-5} \). (b) Percolating cluster at the critical point for the different strain rates: \( \dot{\gamma} = 10^{-2}, 10^{-3}, 10^{-4} \) (left to right). (c) Variation of \( p \) with strain for all strain rates shown in (a). Orange dotted line corresponds to the \( \dot{\gamma} t = 0.5 \). (d) 2-Dimensional slice of MSD map for trajectories shown in (b), at \( \dot{\gamma} t = 0.5 \).

observed, with the more mobile particles localised in a shear-band-like structure spanning the \( x - y \) plane. We can also construct similar maps of local strain \( (\epsilon_{\text{xx}} = \partial \Delta r_z / \partial x) \), which exhibit a localisation behaviour similar to the MSD maps; see Fig. 1(e). Thus, large local MSDs are also regions of large strains. Henceforth, we use \( \Delta r_z^2 \) to analyse local dynamical properties.

In order to quantify and characterise the spatio-temporal evolution of the mobile regions, we define a region to be mobile or not, by setting a threshold \( \mu_{\text{th}} = 0.02 \) on the local \( \Delta r_z^2 \). As marked by the dashed line in Fig. 1(c), such a choice of \( \mu_{\text{th}} \) is larger than the plateau value in the MSD and thus corresponds to motions beyond cage-breaking \[50\]. We then
define the local mobility $\psi$ as

$$\psi = \begin{cases} 
1 & \text{if } \mu \leq \mu_{th}, \\
0 & \text{otherwise}, 
\end{cases}$$

where $\mu$ is the average MSD of particles in a sub-box. Following this convention, we digitize the whole system into mobile and immobile regions. The mobility maps corresponding to Fig. 1(d) are shown in Fig. 1(f).

We now demonstrate that a percolation transition occurs with increasing strain, involving these mobile regions. As the system evolves under the applied shear rate, we monitor the fraction of mobile cells, $p$, at any given instant. Figure 1(d) suggests that such mobile regions do form clusters. Thus, we compute what fraction of these mobile regions, $p_{\text{span}}$, is part of a cluster that spans the system, recalling that such an observable is the order parameter for determining the occurrence of a percolation transition. In Fig. 2(a), we plot $p_{\text{span}}$ as a function of $p$, which shows that beyond a critical fraction $p_c$, all the mobile cells are part of such a spanning cluster. This indicates the occurrence of a percolation transition of these mobile cells. Furthermore, we observe that the variation of $p_c$ is nearly independent of the imposed shear-rate, as seen in Fig. 2(a) for a wide range of $\dot{\gamma}$. Thus, the percolation process is generic to the system’s response under shear. In Fig. 2(b), we visualise the spanning clusters for three different shear rates, at the $p_c$ corresponding to each $\dot{\gamma}$, starting from the same initial state. Here, we note that the spanning across the system-size occurs in the direction of flow, which we discuss further later.

Next, for different imposed $\dot{\gamma}$, if we monitor the numerical growth of mobile cells with increasing strain, a distinct variation is revealed; see Fig. 2(c). For example, at a strain of $\dot{\gamma}t = 0.5$ (marked by orange dotted line), we see that around 50% of the sites are mobile at low strain rates while the same is close to 75% at high strain rates. This implies that subsequent to the percolation transition (at the critical strain corresponding to $p_c$, which is around 0.0685 for $\dot{\gamma} = 10^{-4}$), the spatial heterogeneity of activity is more long-lived for smaller strain rates, as is shown in Fig. 2(c), via cuts in the $x - z$ plane of the local MSD maps corresponding to the evolving trajectories of the states shown in Fig. 2(b). While for the largest shear-rate, mobile regions proliferate in the system, for the smaller shear-rate it is more localised and takes the form of a well-structured shear band. Thus, one can infer that the local dynamics, post-percolation, changes with decreasing shear-rate.
To clarify the nature of the percolation transition, we determine the critical point for the percolation process, $p_c$, using finite size scaling. In Fig. 3(a), we show how $p_{\text{span}}$ varies with $p$ for four different system sizes - we observe that the onset of percolation shifts to larger values of $p$ with increasing system size. By fitting the obtained threshold for different system sizes, using the finite size scaling function $p_c(L) = p_c(\infty) + bL^{-1/\nu_\parallel}$ (see Fig. 3(b), red line), we obtain an estimate of $p_c(\infty) = 0.30339539$ and the exponent $\nu_\parallel = 1.106$, which corresponds to a DP transition \cite{43}. To compare, the corresponding numbers for standard percolation are $p_c(\infty) (= 0.3116)$ and $\nu (= 0.8765)$; the finite-size scaling function using these parameters does not fit our data (see Fig. 3(b), green line). Furthermore, we compute the size distribution of clusters of mobile cells in the vicinity of the percolation transition (Fig. 3(c)). As expected, the distribution has a power-law shape, with the corresponding DP exponent of 2.39 well characterising the distribution \cite{44,45}. Thus, the percolation of the active regions, in this regime of flow, is a directed process, driven in the direction of the external shear.

Thus far, we have discussed the existence of an underlying percolation process of the
mobile regions, which occurs for the entire range of $\dot{\gamma}$ that we have studied. Now, we will focus on how the dynamics proceeds once the percolating cluster has formed. In order to quantify that, we construct two-dimensional projections of the MSD maps. In Fig. 4(a), we show the time evolution of the local mobility, via such maps, for $\dot{\gamma} = 10^{-4}$. We observe that the $z$-width of the shear band increases with time and eventually the entire system becomes mobilised. By locating contiguous layers of mobility, we identify the shear band and, thereafter, by marking the interfaces of this band, we measure how the band-width, $\xi_b$, evolves with time. For different imposed shear rates, this time evolution is shown in Fig. 4(b). We see that $\xi_b$ initially grows quickly and then eventually it reaches a regime where the data can be fitted with $\xi_b \sim t^{1/2}$, implying that the propagating interface of the shear band has a diffusive motion. The diffusion constant is dependent on the imposed shear rate. The smaller the shear rate $\dot{\gamma}$, the slower is the diffusion, which leads to more long-lived heterogeneities, as discussed earlier. For the largest shear rate shown, $\dot{\gamma} = 10^{-3}$, the diffusive regime is very short-lived as the band quickly spans the entire system. For even larger shear rates, beyond percolation, mobile regions quickly appear everywhere and
fluidizes the whole system, and, as a consequence, a shear band is not clearly discernible.

Conclusions. In this work, we have explored how a model glass, subject to a constant strain rate, evolves from a quiescent state to plastic flow. We have shown that this process is initiated by a DP transition. Under shear, hot spots form in the amorphous solid, i.e. local regions in the system where particles have undergone large non-affine displacements. These are local structural changes transforming the initial quiescent glassy state which thus cannot be regained via thermal fluctuations. Unlike liquids, this is an essential feature of the glassy state, where a strong non-linear response to the external shear over-rides thermal fluctuations. Under continuing deformation, such hot spots percolate in the direction of the applied drive. Such a scenario conforms to the DP conjecture [38, 39], whereby a system, with no quenched disorders, transforms from a fluctuating state into an absorbing state. In our case, the dominance of the external shear leads to the system getting irreversibly trapped in an absorbed state of the percolating cluster of hot spots.

The subsequent growth of the active regions after the DP transition depends strongly on the applied shear rate. At larger shear rates, we observe a quick proliferation of mobile spots leading to a fluidisation. On the other hand, the process is asymptotically much slower at smaller shear-rates, with the mobile front slowly diffusing into the rest of the material. Thus, the existence of an initial fast timescale for the burst in mobility in the flow direction along the neutral plane and the subsequent slow timescale for the spread in the shear-gradient direction, leads to the sustenance of shear bands with long timescales for small shear rates.

I. METHODS

We consider a binary mixture of Lennard-Jones (LJ) particles (say A and B) with 80:20 ratio. This is a well-studied glass former. Particles interact via LJ potential which is defined as:

\[ U_{\alpha\beta}^{\text{LJ}}(r) = \phi_{\alpha\beta}(r) - \phi_{\alpha\beta}(R_c) - (r - R_c) \left. \frac{d\phi_{\alpha\beta}}{dr} \right|_{r=R_c}, \]

\[ \phi_{\alpha\beta}(r) = 4\epsilon_{\alpha\beta} \left[ (\sigma_{\alpha\beta}/r)^{12} - (\sigma_{\alpha\beta}/r)^6 \right] r < R_c, \]

where \( \alpha, \beta = \text{A, B} \). Interaction among particles is defined as \( \epsilon_{\text{AA}} = 1.0, \epsilon_{\text{AB}} = 1.5\epsilon_{\text{AA}}, \epsilon_{\text{BB}} = 0.5\epsilon_{\text{AA}} \). Range of interactions is given as \( \sigma_{\text{AA}} = 1.0, \sigma_{\text{AB}} = 0.8\sigma_{\text{AA}}, \sigma_{\text{BB}} = 0.88\sigma_{\text{AA}} \) and \( R_c = 2.5\sigma_{\text{AA}} \). Masses of both type of particles are equal, i.e., \( m_A = m_B = m \). All
quantities are expressed in LJ units in which the unit of length is $\sigma_{AA}$, energy is expressed in the units of $\epsilon_{AA}$ and the unit of time is $\sqrt{m\sigma_{AA}^2/\epsilon_{AA}}$. More details about the model and parameters can be found in Ref. [46].

We perform molecular dynamics (MD) simulation in the NVT ensemble using the package LAMMPS (“Large-scale Atomic/Molecular Massively Parallel Simulator”) [47]. Different geometries are considered, placing the particles in boxes of dimensions $10 \times 10 \times 40$, $20 \times 20 \times 80$, $40 \times 40 \times 40$ and $50 \times 50 \times 50$. Temperature is maintained via dissipative particle dynamics (DPD) thermostat [48].

Our method for the preparation of glass is as follows: First we equilibrate high temperature initial configuration at a temperature $T = 0.45$ in the super-cooled regime and then quench it to a temperature $T = 0.2$ below the mode coupling transition temperature [46]. We wait until $t_w = 10^4$ and apply shear on $x$-$z$ plane in the direction of $x$ with different constant strain rates $\dot{\gamma} = 10^{-2}, 10^{-3}, 3 \times 10^{-4}, 10^{-4}, 3 \times 10^{-5}$ and $10^{-5}$. To simulate a sheared bulk glass, we use Lees-Edwards periodic boundary conditions [49].

**Maps of local MSD.** To get the MSD maps, we divide the simulation box into small cubic sub-boxes having linear size of $\sigma_{AA}$. At any time $t$, we calculate the average MSD of the particles populating each sub-box at $t = 0$ (unsheared glassy state). As discussed earlier, we measure the $z$-component of MSD of each particle. The corresponding three-dimensional map, thus constructed, is shown in the different plots.

**Maps of local strain.** To plot the strain maps, again we divide the simulation box into small cubic sub-boxes, as in the case of MSD maps. The numerical derivative of $z$-component of displacement with respect to $x$, i.e. $\epsilon_{zx} = \partial \Delta r_z / \partial x$ is calculated. This derivative, which gives strain $\epsilon_{zx}$, is plotted for each sub-box to construct the map.

**Identifying the interface of shear-band.** To characterize shear bands, we choose a slightly higher threshold on MSD. In the present work, it is taken as $\mu_{th} = 0.1$. We divide the simulation box into $x$-$y$ layers of thickness one particle diameter and calculate $\mu$, which is the $z$-component of MSD, for each layer. We assign each layer a value $\psi = 1/0$ depending on whether $\mu$ is greater/lower than $\mu_{th}$. To get the size of shear band we count the number
of adjoining layers for which $\psi = 1$.

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III. AUTHOR CONTRIBUTIONS

All authors contributed to designing and performing the simulations, analysing the data and writing the manuscript.