SHEAR LOCALIZATION
AND PERCOLATION OF STABLE STRUCTURE
IN AMORPHOUS SOLIDS

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ABSTRACT
Molecular dynamics simulations of uniaxial tension in a two-dimensional model of a metallic glass exhibit varying degrees of shear localization depending upon the process by which each of the materials was produced. The samples that were quenched most gradually show the largest degree of localization. In addition higher strain rates lead to increased localization in the most rapidly quenched samples, while the more gradually quenched samples show the reverse strain rate dependence. This transition in localization and strain rate dependence coincides with a structural transition in the material. Gradually quenched samples have a higher percentage of atoms in quasi-crystal-like local environments. The transition in the mechanical properties coincides with the percolation of this backbone of quasi-crystal-like material. Shear localization occurs in regions where material is altered from this more stable structure to a fully amorphous structure under the effect of plastic deformation.

1 INTRODUCTION

Metallic glasses can be produced by a variety of processing routes ranging from casting at relatively low cooling rates of a few K per second in the case of bulk glass formers [1, 2] to splat quenching at elevated cooling rates [3] to ion beam assisted deposition, an extremely energetic non-equilibrium process [4]. Since glasses, unlike crystals, do not naturally conform to a small number of identifiable and well-defined ground state structures, it is generally expected that glasses can exhibit a range of mechanical properties depending on processing.

Localization plays an important role in the mechanical response of metallic glasses. Particularly under uniaxial tension the tendency to localize can result in dramatic failure via fracture along a single shear band [5]. This failure mode can entirely prevent any apparent hardening in tension, limiting the structures in which metallic glasses can be employed as structural materials.

Despite recent progress, understanding of the physical origin of the softening mechanism that leads to localization remains rudimentary. While the most widely accepted theories [6-8] do provide some important physical insight regarding the role of "free volume" and micromechanics in softening, none of these can be used to accurately predict the nucleation and propagation of shear bands, and none of the parameters in the current theories can unambiguously be attributed to specific atomic scale structures or mechanisms. In many cases computer simulation studies have provided critical insight for theoretical progress [9, 10] and it is in the context of this work that the current study has been undertaken.

We have performed a series of uniaxial tensile test simulations on a series of model metallic glasses. Because of the difficulty of modeling metallic glasses in their full complexity even on relatively small scales we have chosen to make a number of simplifying assumptions that preclude our simulations from providing quantitative predictions, but that allow us to simulate large enough systems to obtain important physical insights into the deformation that occurs during loading. These simplifications include simulating a binary alloy, using a pair-potential interaction to model the metallic bonding, restricting our simulation to two-dimensions, operating at rates significantly above those typically observed experimentally and using glass preparation

procedures at faster quench rates than are typically experienced in the laboratory. Despite these simplifications the results of the simulations exhibit most of the salient features of the experimental observations strain localization in metallic glasses. They also reveal some interesting trends regarding shear bands and metallic glass processing, and they provide a means to correlate deformation with atomic scale structural changes in the glass.

2 METHODOLOGY

We have performed molecular dynamics simulation on a binary alloy that consists of two species, which we will refer to as S and L for small and large, interacting via a Lennard-Jones potential of the form

$$\phi(r) = 4\varepsilon\left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right], \tag{1}$$

where ε represents the minimum energy of the bond and σ provides a length scale, the distance at which the interaction energy is zero (NB: this is not the equilibrium bond length). The SS and LL bond energies are half that of the SL bond energy, $\varepsilon_{SS} = \varepsilon_{LL} = \frac{1}{2}\varepsilon_{SL}$. The SS and LL length scales are related to the SL length scale by the relations

$$\sigma_{SS} = 2\sigma_{SL}\sin\left(\frac{\pi}{10}\right), \quad \sigma_{LL} = 2\sigma_{SL}\sin\left(\frac{\pi}{5}\right). \tag{2}$$

In this binary system we will take the reference length scale to be the σ value of the SL bond and the reference energy scale to be the energy of the SL bond. All the particles will have the same mass, m_0, which will be the reference mass scale. The reference time scale will therefore be $t_0 = \sigma_{SL}\sqrt{m_0/\varepsilon_{SL}}$. In order to make rough comparisons to experiments and to present times in physical units we will consider that for a typical material $t_0 \approx 1$ ps, and $\sigma_{SL} \approx 3$ Å.

This system was chosen because it exhibits both crystalline and quasi-crystalline ground states [11]. The existence of a quasi-crystalline ground state was of particular interest because it has been proposed that such an underlying quasi-crystalline state stabilizes Zr based glasses , some of the most stable bulk glass formers produced to date [12, 13]. This system, perhaps due to this underlying quasi-crystalline state, exhibits a strong tendency toward amorphization. Many other two-dimensional systems show a strong tendency to crystallize. In addition this system has been used extensively to study quasi-crystal and amorphous thermodynamic and mechanical properties. We chose our composition $N_L:N_S = (1+\sqrt{5}):4$ to be consistent with other studies of this system. T_g of this system is known to reside around 0.325 ε_{SL}/k, where k is the Boltzmann factor. For the sake of comparison temperatures will be measured in units of T_g.

The model system was simulated using a standard leapfrog integration scheme applied to the Newtonian equations of motion [14]. During quenching the coupling to the external heat bath was modeled using a Nose-Hoover thermostat. However, during the mechanical tests no such thermostating technique was applied.

3 SAMPLE PREPARATION AND TESTING

The initial conditions were created by starting from supercooled liquids equilibrated above the glass transition temperature. The relaxation time of the liquid was measured by tracking the decay of the structure factor. Liquids were equilibrated for many times this relaxation time. Subsequent to equilibration the temperature of the liquid was reduced to 9.2% of T_g. The most gradually
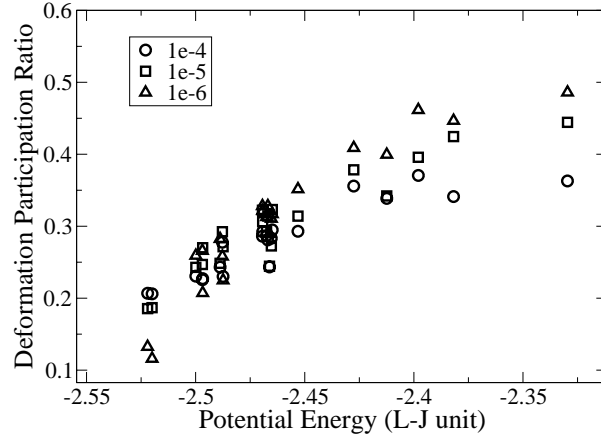
Figure 1: Deformation Participation Ratio at 5% strain in uni-axial tension tests for all 21 samples tested at strain rates of 10^{-4}, 10^{-5}, 10^{-6} ($1/t_0$).

quenched sample, was cooled at a rate of 1.97×10^{-6} T_g/t_0 corresponding to a quench over approximately 0.5 μs. Other samples were quenched at rates as much as 100 times higher. In addition a number of samples were produced by quenching instantaneously from melts of different temperatures by rescaling the particle velocities and then allowing the system to age for 100 t_0, approximately 0.1 ns. In each case the material that was produced was characterized by the potential energy per atom of the as-quenched sample. A summary of the samples produced in these ways are shown in Table 1. For the most slowly quenched samples a number of each type were produced and tested. These were used to determine the effects of sample to sample variation.

During the uniaxial tensile test the top and bottom boundaries were maintained as periodic boundaries. The left and right boundaries were free surfaces. This was necessary to avoid over-constraining the system in a way that would preclude localization. The strain was imposed by rescaling the boundaries and the positions by a small amount in the yy direction at each time step at a constant strain rates of 10^{-4}, 10^{-5} and 10^{-6} ($1/t_0$). This was performed using the molecular dynamics algorithm described in [14]. Since the strain rate imposed was significantly slower than the system size divided by the sound wave speed, inertial effects were negligible, although strain rate effects arising from the deformation mechanism were important in a number of the tests.

4 RESULTS

In order to quantify the degree of localization we will define a quantity that we will call the deformation participation ratio (DPR). The DPR is the fraction of atoms that appear to undergo a deviatoric shear strain larger than the nominal deviatoric shear strain of the entire sample. In a homogeneously deforming sample every atom is equally as likely to exceed the mean as not, and the DPR should be approximately 0.5 in this case. However, in a highly localized deformation the DPR should become negligible and approach the ratio of the shear band width to the system size in the case in which a single shear band bears all the deformation. Fig. 1 shows the DPR at 5% strain as a function of the potential energy per atom. Different symbols denote results for different quench rates. Fig. 2 shows representative images of the deviatoric strain in samples with high,

Table 1 Summary of Sample Preparation Methods Employed

$T_{starting}$ (T_g)	T_{end}(T_g)	Quenching Time (t_0)	Quenching Rate (T_g/t_0)	Potential Energy (ε_{SL})	Number of Samples
2.46	0.1	Instant	Instant	-2.330	1
1.38	0.1	Instant	Instant	-2.382	1
1.23	0.1	Instant	Instant	-2.398	1
1.15	0.1	Instant	Instant	-2.412	1
1.08	0.1	Instant	Instant	-2.428	1
2.46	0.1	5.00E+03	4.74E-04	-2.453	1
1.38	0.1	5.00E+03	2.58E-04	-2.469	1
1.23	0.1	5.00E+03	2.28E-04	-2.465	1
1.15	0.1	5.00E+03	2.12E-04	-2.469	1
1.08	0.1	5.00E+03	1.97E-04	-2.465	1
1.08	0.1	1.00E+04	9.85E-05	-2.467	3
1.08	0.1	5.00E+04	1.97E-05	-2.488	3
1.08	0.1	1.00E+05	9.85E-06	-2.498	3
1.08	0.1	5.00E+05	1.97E-06	-2.521	2

intermediate and low DPR values. This data was extracted from the atomic positions using the procedure for extracting a best fit strain introduced in [14]. Note that samples with a high DPR show regions of deformation distributed throughout the material. Samples with low DPR exhibit well defined shear bands. Samples with high potential energy per atom, corresponding to the instantaneously quenched samples, also exhibit lower DPR and increased localization at higher strain rates. Below a potential energy per atom of -2.46 ε_{SL} the dependence of the DPR on strain rate is reversed. In the gradually quenched glasses localization becomes more pronounced at lower strain rates.

Next we examined the dependence of the localization on the atomic scale structure of the samples. To accomplish this we have adapted an analysis from [15] that utilizes the fact that this system has an underlying quasi-crystaline ground state. In [15] it was noticed that the quasi-crystal is composed of nine local atomic motifs. We have examined the structure of the samples, and we have determined if each atom resides in one of these motifs. We refer to these atoms as stable atoms and other atoms as unstable atoms. A strong correlation exists between the potential energy per atom and the percent of atoms in these stable structural motifs. An analysis of the relative probability of stable and unstable atoms undergoing deformation showed only a small differentiation, but stable atoms next to other stable atoms are considerably less likely to undergo deformation. Motivated by this observation we extracted the cluster size of adjacent stable atoms in each sample. Fig. 3 shows the cluster size as a function of the potential energy per atom. At a potential energy per atom of approximately -2.46 ε_{SL} the clusters percolate. Note that the transition of the strain rate dependence in Fig. 2 also occurs at this potential energy per atom. During deformation of samples with a percolating backbone the clusters dramatically decrease in size. In non-percolating samples the clusters remain roughly the same size throughout deformation.
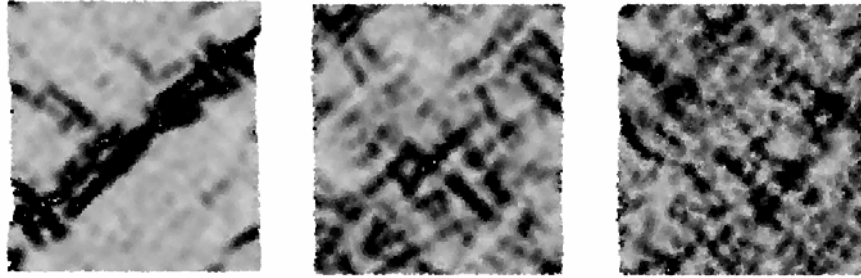
Figure 2: Deformation map for three samples corresponding to potential energies per particle of -2.521, -2.465 and -2.428 ε_{SL} loaded to 5% nominal strain at a strain rate of 10^{-6} ($1/t_0$). Color corresponds to the degree of deviatoric strain. Black is 10% shear strain, gray is 0% shear strain.

5 CONCLUSIONS

We have performed a series of uniaxial tensile test simulations on binary models of a glass forming system in two dimensions. More gradually quenched samples have higher degrees of quasi-crystalline order. These samples also show an increasing tendency to exhibit shear band formation during tensile loading. In particular, shear localization and strain rate independence appear to be robust features of the samples in which a backbone of atoms with quasi-crystal-like arrangements percolate through the material. This backbone appears to increase the mechanical strength of the material, but the softening that occurs when the material reverts to an amorphous structure leads to localization in tension.

A number of the more stable metallic glass formers precipitate quasi-crystals upon annealing. It has been proposed that small regions with quasi-crystalline order may be important for the stability of the amorphous state in these glasses [12, 13]. In addition it has been asserted that deformation takes place primarily in the amorphous region of the material [16]. In our model system we have observed that the dispersed quasi-crystalline arrangements play a critical role in controlling the qualitative nature of deformation in the material. In the material that shows the strongest tendency to form shear bands deformation converts material from quasi-crystal-like to
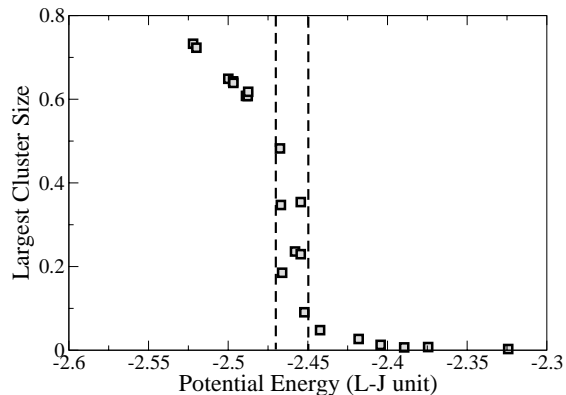

Figure 3: Largest cluster size as a fraction of system size. A percolation transition is observed around -2.46 ε_{SL} in the region denoted by the dashed lines.

fully amorphous. This implies that this conversion of material to the amorphous state may play a crucial role in the softening process that leads to the shear banding instability. However, it remains unclear whether this is true for all metallic glasses, or only those in which the stability of the amorphous phase arises from an underlying quasi-crystalline phase.

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