Yielding in multi-component metallic glasses: Universal signatures of elastic modulus heterogeneities

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Sheared multi-component bulk metallic glasses are characterized by both chemical and structural disorder that define their properties. We investigate the behavior of the local, microstructural elastic modulus across the plastic yielding transition in six Ni-based multi-component glasses, that are characterized by compositional features commonly associated with solid solution formability. We find that elastic modulus fluctuations display consistent percolation characteristics pointing towards universal behavior across chemical compositions and overall yielding sharpness characteristics. Elastic heterogeneity grows upon shearing via the percolation of elastically soft clusters within an otherwise rigid amorphous matrix, confirming prior investigations in granular media and colloidal glasses. We find clear signatures of percolation transition with spanning clusters that are universally characterized by scale-free characteristics and critical scaling exponents. The spatial correlation length and mean cluster size tend to diverge prior to yielding, with associated critical exponents that exhibit fairly weak dependence on compositional variations as well as macroscopic stress-strain curve details.

In realistic studies of bulk metallic glasses, the associated yielding properties and shear banding behavior [1] have important industrial and technological implications in terms of the ductility of the glass. The ongoing eagerness to promote the latter (in combination with hardening properties) have already shifted the focus to the development of multi-component glasses (also known as amorphous alloys and/or high-entropy metallic glasses) with the highly tunable microstructural/compositional complexity [2, 3]. The atomistic origin of this new design paradigm is based on the common observation that glasses with low atomic disparity limit (where atomic radii of constituent elements are considered to be very similar) have a high capacity to form localized deformation patterns whereas those at the opposite limit have a tendency to delocalize strain [4] and, therefore, deform in a more ductile way. In this context, the shear band structure and design-level ductility appear to be highly dependent on inherent heterogeneity as an essential elemental/compositional feature. However, microstructural origins and elemental dependence of such inhomogeneities and, more importantly, associated spatial-dynamical evolution upon shearing have yet to be explored.

Despite certain commonalities of failure properties across disordered solids [5], the sharpness of yielding transition, as a common signature of strain localization (and ductility), may show strong variations by altering thermal treatments and chemical compositions in bulk metallic glasses, ranging from uniformly distributed patterns to system-spanning crack-like features [2, 4, 6, 7]. Owing to the presence of quenched chemical/structural disorder, glassy metals may accommodate a distributed plastic flow with a significant contribution to ductility [8–10]. Certain (aged) glasses that lack this heterogeneity element [11–13] (or associated lengths don’t exceed interatomic scales) tend to localize strain within a single dominant band before shear instability results in a catastrophic brittle-type fracture. In this context, tailoring elastic heterogeneity has recently emerged as a novel design framework to build more ductile metallic glasses [8, 14]. Under special thermal treatments and variations of chemical elements, quenched metallic glasses may nucleate elastically soft clusters that become structured and span the entire system upon failure, leading to the enhanced plastic flow and, hence, ductility. This is illustrated in Fig. 1 where yielding transition, fine-tuned by the chemical compositions, is accompanied by the percolating networks of softness (shown in brown) in a driven CoNiFe and CoNiCrFe bulk metallic glass. The relevance of such a percolation transition of softness within the context of amorphous plasticity has been already established in (soft) model glasses [15–17] as well as colloidal systems [17, 18] under shear. These studies have mainly focused on certain (implicit) notions of softness based on the degree of deformation non-affinity or shear-induced mobility that exhibit percolating features near the criticality. Using a model metallic glass in [19], the glass transition was accompanied by the percolation threshold of mechanically unstable regions on approach to the transition temperature Tg. However, realistic studies of bulk metallic glasses driven out of equilibrium, with substantial industrial and technological implications, and associated composition-dependence has been more challenging.

Here, using atomistic simulations, we investigate six multi-component bulk metallic glasses including Co, Ni, Fe, Cr, and Mn at compositions that have been the focus of recent experimental investigations. The chosen compositions are commonly believed to optimize solid solution formability, characterized by low misfit coefficients δa, and not thought to promote glass forming ability typically observed for δa > 6% [20]. Nonetheless, there is a way to generate a glassy environment in the opposite

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Simulations & Protocols—Details of molecular dynamics simulations are given in [4] including relevant units, initial configurations, interatomic forces, and deformation parameters of FeNi, CoNiFe, CoNiCrFe, CoCrFeMn, CoNiCrFeMn, and Co5Cr2Fe10Mn27Ni28 model metallic glasses. In order to compute the elasticity tensor locally for simulated glasses, simple shear tests were performed.

From a broader perspective, our methodology offers a robust indicator of nonaffinity, e.g. the disorder-induced breakdown of homogeneous response, by probing elastic heterogeneity in amorphous materials. Conceptually, our approach is similar to investigations of low energy quasi-localized modes that are spatially distinguishable from long wave-length phonon modes in elastic crystalline solids (see [21] and references therein). Predictive plasticity models solely based on the notion of shear transformation zones (STZs) might fail to capture such correlations because the latter are generally believed to be (micro)structural defects but not necessarily indicative of mechanical instability. Another complication arises from a lack of robust topological signatures that can identify STZs from their parent liquid-like microstructure within a glassy matrix. There have been related efforts based upon the machine learning-rooted concept of “structural” softness but with very limited predictive capabilities in terms of glass failure and deformation [22–24]. More focused studies of metallic glasses made an attempt to associate soft spots to the presence/absence of geometrically unfavored motifs and/or short/medium range ordering [25–27] but these local motifs on their own often fail to describe the collective nature of plasticity and shear banding in a broader context of amorphous plasticity. Our study aims to augment such efforts, directed mainly towards the notion of structural heterogeneity, by adding the elastic heterogeneity picture to the scene which is more conforming with the long-established notion of structure-property correlations in (metallic) glasses.

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By probing local elasticity, in particular shear modulus \( \mu \) and its evolution toward failure, we present direct evidence that plastic yielding in driven metallic glasses takes place universally through a percolation transition of softness, characterized by regions of negative \( \mu \leq 0 \). The slope in a) indicates the softening modulus \( h_{\min} \). Upon shearing, negative-\( \mu \) clusters form percolating networks (in brown) on approach to failure that tend to correlate with the (post-failure) macroscopic properties.

The limit of \( \delta_a \to 0 \) but maintaining a larger number of elements, as in medium/high entropy metallic glasses (see [3] and references therein).

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on the $xy$ plane at a fixed strain rate $\dot{\gamma}_{xy} = 10^{-5}$ ps$^{-1}$ and temperature $T = 300$ K up to a prescribed (pre-)strain $\gamma_{xy} \leq 0.2$. In line with [28, 29], we subsequently perturbed the simulation cell through six deformation modes in Cartesian directions $xyz$ and evaluated resulting differences in atom-wise stresses to construct the local elasticity tensor for each atom (see the appendix). Relevant atom-based quantities were interpolated on a fine cubic grid to be used as input for our three-dimensional cluster processing. This methodology allows us to probe the spatial-dynamical evolution of the local shear modulus $\mu = c_{\mu xy}$ and associated percolation features near failure transition in sheared glasses. We further measure the softening modulus $h_{\min}$ defined as the maximum rate of the macroscopic stress drop for all the different compositions. The stress drop, typically defined as the difference between the overshoot stress and the subsequent flow stress, is associated with the initiation of a catastrophic shear band and has been used as an appropriate order parameter in model glass studies [30, 31] showing meaningful variations with glass compositions and processing parameters [11]. We note that in metallic glass simulations and/or experiments a robust measurement of the macroscopic drop is not always feasible due to the lack of a well-defined steady flow regime that is expected to follow the stress overshoot. In a recent work [4], we established $h_{\min}$ as a more robust experimentally-relevant indicator of shear banding and associated structural features. Here, we show that the variations in $h_{\min}$ tend to correlate with the softness properties as inferred from local elasticity.

Results — Figure 1 displays results of the shear tests performed on the quenched CoNiFe and CoNiCrFe glasses. The cluster maps based on unstable regions with $\mu \leq 0$ are visualized in Fig. 1(b) and (c). We denote the fraction of unstable sites by (probability) $p$ which appears to evolve in a non-monotonic fashion, Fig. 7(a) and (c), with a peak value $p_{\text{max}}$ that almost coincides with that of the (bulk) shear stress $\sigma_{xy}$ (at $\gamma_{xy} \approx 0.1$) as in Fig. 1(a). Apart from variations in $p_{\text{max}}$, Fig. 7(a-f) shows similar trends for the evolution of $p$ with $\gamma_{xy}$ corresponding to the other compositions. We notice $p > 0$ even in unstrained quenched glasses (at $\gamma_{xy} = 0$) owing to the distributed (but disconnected) networks of soft (Eshelby-like [32]) inclusions within the glassy matrix.

The above elasticity maps give a visual impression that the glass failure (at $\gamma_{\text{max}} \approx 0.1$) might indeed coincide with a percolation transition of softness at $p_{\text{max}}$ upon shear loading. In order to validate this picture, we adopted ideas from the classical percolation theory [33] including investigations of cluster sizes and their dynamical evolution. As a basic statistical property, $n_s$ denotes the probability distribution function associated with the number of clusters containing $s$ unstable sites. Figure 2(a) and (c) plots $n_s$ associated with the CoNiFe and CoNiCrFe glasses at three different strains. The cluster size distributions tend to develop fairly long tails as $p$ approaches $p_{\text{max}}$. Our data suggest a robust power-law decay $n_s \propto s^{-\tau}$ with $\tau = 2.0$ and 2.1, associated

FIG. 3. Mean cluster size $S$ plotted against $p$ associated with the a) CoNiFe and c) CoNiCrFe glasses. The graphs in b) and d) are the same as a) and c) but plot $S$ as a function of $1 - p/p_{\text{max}}$ with a) $p_{\text{max}} = 0.14$ and c) $p_{\text{max}} = 0.20$. The (red) curves indicate binning averaged data. The dashdotted lines denote power laws $S \propto (p_{\text{max}} - p)^{-\gamma}$ with b) $\gamma = 2.89$ and d) $\gamma = 2.20$.

FIG. 4. Correlation length $\xi$ plotted against $p$ corresponding to the a) CoNiFe and c) CoNiCrFe glasses. The graphs in b) and d) are the same as a) and c) but plot $\xi$ as a function of $1 - p/p_{\text{max}}$ with a) $p_{\text{max}} = 0.14$ and c) $p_{\text{max}} = 0.20$. The (red) curves indicate binning averaged data. The dashdotted lines denote power laws $\xi \propto (p_{\text{max}} - p)^{-\nu}$ with b) $\nu = 1.26$ and d) $\nu = 0.51$. 
TABLE I. Comparison between estimated scaling exponents associated with different chemical compositions and three dimensional (d = 3) percolation theory.

| Chemical Composition | Cluster Size Distribution | Mean Cluster Size | Correlation Length | Fractal Dimension |
|----------------------|---------------------------|------------------|-------------------|-------------------|
|                      | $n_s \propto s^{-\tau}$   | $S \propto (p - p_{max})^{-\gamma}$ | $\xi \propto (p - p_{max})^{-\nu}$ | $s \propto r_s^{d_f}$ |
| FeNi                 | 1.96                      | 2.23             | 0.87              | 2.27              |
| CoNiFe              | 1.98                      | 2.89             | 1.26              | 2.25              |
| CoNiCrFe            | 2.05                      | 2.20             | 0.51              | 2.62              |
| CoCrFeMn            | 2.02                      | 2.42             | 0.95              | 2.48              |
| CoNiCrFeMn          | 2.00                      | 2.26             | 0.62              | 2.43              |
| Co$_5$Cr$_2$Fe$_{40}$Mn$_{27}$Ni$_{26}$ | 1.93 | 2.69     | 0.94              | 2.28              |
| CuZr[17]            | –                         | –                | 0.85 ± 0.1        | 2.0               |
| Percolation (d = 3) [33] | 2.18          | 1.80             | 0.88              | 2.53              |
| Directed Percolation (d = 3) [34] | –            | 1.25             | 0.6               | 2.0               |

FIG. 5. Scatter plot of softening modulus $h_{min}$ and scaling exponents $\alpha$ a) $\gamma$ b) $\nu$ corresponding to the FeNi (●), CoNiFe ( ■), CoNiCrFe ( ●), CoCrFeMn ( ▲), CoNiCrFeMn ( ◆), and Co$_5$Cr$_2$Fe$_{40}$Mn$_{27}$Ni$_{26}$ ( ▽) bulk metallic glasses.

with CoNiFe and CoNiCrFe, over at least two decades in $s$. The estimated range of exponent $\tau$ is fairly robust showing minimal compositional dependence (as reported in Table I) and/or variations near $p_{max}$. The radius of gyration associated with a cluster of size $s$ may be defined as $r_s^2 = \sum_{i=1}^{s} |\vec{r}_i - \vec{r}_0|^2 / s$ with the center of mass $\vec{r}_0 = \sum_{i=1}^{s} \vec{r}_i / s$. Figure 2(b) and (d) illustrates that $s \propto r_s^{d_f}$ with fractal dimension $d_f = 2.3$, 2.6 corresponding to CoNiFe and CoNiCrFe, respectively. This almost agrees with Fig. 1(b) and (c) in that the soft spots tend to form fairly compact clusters in the latter glass whereas the former one is associated with more localized (but still system-spanning) features.

Figure 3(a) and (c) displays the mean cluster size $S = \sum_{s} n_s s^2 / \sum_{s} n_s s$ and its evolution with $p$. The average size reveals a certain algebraic divergence of $S$ on approach to the maximum fraction $p \to p_{max}$. This divergence is demonstrated in Fig. 3(b) and (d) with the mean size scaling as $S \propto (p_{max} - p)^{-\gamma}$ and $\gamma = 2.89$, 2.20 corresponding to the CoNiFe and CoNiCrFe compositions, respectively. The proposed scaling is valid for a little less than a decade in $1 - p / p_{max}$ down to a roll-off at small arguments potentially due to finite size effects. The (squared) correlation length $\xi^2 = 2 \sum_{s} r_s^2 s^2 n_s / \sum_{s} s^2 n_s$ is defined based on a weighted average associated with the radius of gyration $r_s^2 = \sum_{i=1}^{s} |\vec{r}_i - \vec{r}_0|^2 / s$ of a cluster of size $s$, as shown in Fig. 4(a) and (c). Here the center of mass is $\vec{r}_0 = \sum_{s} s \vec{r}_s / s$. As $p$ increase toward $p_{max}$ in Fig. 4(b) and (d), the correlation length scales like $\xi \propto (p_{max} - p)^{-\nu}$ with $\nu = 1.26$ corresponding to CoNiFe and $\nu = 0.51$ for CoNiCrFe. The center and right panels of Fig. 7? show the mean cluster size $S$ and correlation length $\xi$ as a function of applied strain $\gamma_{xy}$ associated with different compositions.

We repeated the above analysis to infer critical exponents corresponding to the other chemical compositions (see Fig. 7? and Table I) and sought for potential connections with the softening modulus $h_{min}$. Figure 5 plots the latter and estimated critical exponents $\tau$, $\gamma$, $\nu$, and $d_f$ for the FeNi, CoNiFe, CoNiCrFe, CoCrFeMn, CoNiCrFeMn, Co$_5$Cr$_2$Fe$_{40}$Mn$_{27}$Ni$_{26}$ metallic glasses. As previously discussed, the cluster size exponent $\tau$ in Fig. 5(a) shows insignificant variations with chemical compositions. The scatter plot of $h_{min}$ and mean cluster size exponent $\gamma$ in Fig. 5(b) suggests noticeable anti-correlations between the two observables. Likewise, the correlation length exponent $\nu$ seems to be statistically (anti-)correlated with $h_{min}$ as in Fig. 5(c). The increasing trend in the fractal dimension $d_f$ might
be indicative of localized (system-spanning) soft spots in good glasses that become increasingly packed in space as compositional variations lead to a smoother yielding transition. Given that there are only two independent exponents in standard percolation theory, the behavior of \( \nu \) and \( \gamma \) should be largely controlled by variabilities in \( d_f \).

Conclusions & Discussions — We have presented direct evidence that yielding transition of bulk metallic glasses is accompanied with percolation threshold of softness upon failure across different chemical compositions. The former has been quantified by the softening modulus \( h_{\text{min}} \), indicative of the sharpness of plastic yielding transition, and the latter was characterized by analyzing connected networks of mechanically soft regions that grow under application of external stress. Our analysis further indicates critical scaling features associated with the dynamics and topology of soft clusters suggesting a close relevance of the percolation transition. Relevant scaling exponents and potential correlations with the macroscopic stress-strain curve have been considered from the compositional-dependence perspective. Table I features the range of estimated exponents corresponding to different multi-component metallic glasses probed in this work and the study by Schall et al. [17] as well as those inferred from the percolation theory. In this framework, we find a fairly robust range (across different systems) associated with the cluster size exponent \( 1.9 \leq \tau \leq 2.1 \) and comparable with theoretical predictions [33]. On the other hand, \( \gamma, \nu, \) and \( d_f \), inferred from the scaling of the correlation lengths and mean cluster sizes, somewhat correlate with softening modulus \( h_{\text{min}} \) suggesting that the softness percolation might influence macroscopic yielding properties through compositional dependence. Such correlations may also suggest that the notion of universality in deformation and flow properties of multicomponent glasses could be limited owing to compositional/microstructural associations. Nevertheless, given a relatively narrow range of the softness exponents across different glasses, one might still consider the same universality class for the observed transition despite the fact that our results show substantial composition-based variabilities in yielding properties.

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SUPPLEMENTARY MATERIALS

Simple shear tests were performed on the xy plane at a fixed strain rate $\dot{\gamma} = 10^{-5}$ ps$^{-1}$ and temperature $T = 300$ K up to a prescribed strain $\gamma \leq 0.2$. We subsequently applied six deformation modes $e^{(m)}$, $m = 1...6$ in Cartesian directions $xyz$ described by the following global strain tensors

$$e^{(1)} = \epsilon_{xx} \delta_{\alpha \alpha} \delta_{\beta \beta},$$
$$e^{(2)} = \epsilon_{yy} \delta_{\alpha \alpha} \delta_{\beta \beta},$$
$$e^{(3)} = \epsilon_{zz} \delta_{\alpha \alpha} \delta_{\beta \beta},$$
$$e^{(4)} = \epsilon_{xy} (\delta_{\alpha x} \delta_{\beta y} + \delta_{\alpha y} \delta_{\beta x}),$$
$$e^{(5)} = \epsilon_{yz} (\delta_{\alpha y} \delta_{\beta z} + \delta_{\alpha z} \delta_{\beta y}),$$
$$e^{(6)} = \epsilon_{xz} (\delta_{\alpha x} \delta_{\beta z} + \delta_{\alpha z} \delta_{\beta x}).$$

(1)

Here $\delta_{\alpha \beta}$ is a Kronecker delta with $\alpha$ and $\beta$ denoting Cartesian indices. The strain magnitudes are set to $\epsilon_{xx} = \epsilon_{yy} = \epsilon_{zz} = 2\epsilon_{xy} = 2\epsilon_{yx} = 2\epsilon_{xz} = 10^{-2}$ in strain units. We made use of (deformation-induced) stress differences $\Delta \sigma^{(m)}_{i,\alpha \beta}$, $m = 1...6$ in atoms $i = 1...N$ to evaluate the corresponding elasticity tensor

$$c^{i}_{\alpha \beta \gamma} = \frac{\Delta \sigma^{(i)}_{i,\alpha \beta}}{\epsilon_{\gamma \gamma}},$$
$$c^{i}_{\alpha \beta \gamma} = \frac{\Delta \sigma^{(i)}_{i,\alpha \beta}}{\epsilon_{\gamma \gamma}},$$
$$c^{i}_{\alpha \beta \gamma} = \frac{\Delta \sigma^{(i)}_{i,\alpha \beta}}{\epsilon_{\gamma \gamma}},$$
$$c^{i}_{\alpha \beta \gamma} = \frac{\Delta \sigma^{(i)}_{i,\alpha \beta}}{2\epsilon_{\gamma \gamma}},$$
$$c^{i}_{\alpha \beta \gamma} = \frac{\Delta \sigma^{(i)}_{i,\alpha \beta}}{2\epsilon_{\gamma \gamma}},$$
$$c^{i}_{\alpha \beta \gamma} = \frac{\Delta \sigma^{(i)}_{i,\alpha \beta}}{2\epsilon_{\gamma \gamma}}.$$

(2)

Statistics of clusters with $\mu = e^{i}_{xyxy} < 0$ associated with FeNi , CoNiFe , CoNiCrFe , CoCrFeMn , CoNiCrFeMn , Co5Cr2Fe40Mn27Ni26 metallic glasses are illustrated in Fig. S1 and S2. The fraction of unstable sites $p$ appears to evolve in a non-monotonic fashion, as shown in the leftmost panels of Fig. S1(a-f), with a peak value $p_{\text{max}}$ that almost coincides with that of the (bulk) shear stress $\sigma_{xy}$ (at $\gamma_{xy} \simeq 0.1$). Overall, the evolution of $p$ with $\gamma_{xy}$ near $p_{\text{max}}$ features similar trends across all the compositions. The center and right panels of Fig. S1 show the mean cluster size $S$ and correlation length $\xi$ as a function of applied strain $\gamma_{xy}$ associated with different compositions. Both observables tend to grow on approach to failure revealing slight compositional dependence. This is further demonstrated in the first and third columns of Fig. S2(a-f) with the mean size and correlation length plotted as a function of $p$. The rescaled data in the second and fourth columns of Fig. S2(a-f) feature a power-law scaling universally described by $S \propto (p_{\text{max}} - p)^{-\gamma}$ and $\xi \propto (p_{\text{max}} - p)^{-\nu}$ near $p_{\text{max}}$. Here, we note that every panel in the second and fourth columns shows two decades in $S$ and slightly over one decade in $\xi$. The estimated critical exponents $\gamma$ and $\nu$ are reported in Table 1.
FIG. S1. Cluster statistics: probability $p$ (1st column), mean size $S$ (2nd column), and correlation length $\xi$ (3rd column) plotted against applied strain $\gamma_{xy}$ corresponding to the FeNi , CoNiFe , CoNiCrFe , CoCrFeMn , CoNiCrFeMn , and Co$_5$Cr$_2$Fe$_{40}$Mn$_{27}$Ni$_{26}$ glasses. The (red) curves indicate binning averaged data.
**FIG. S2.** Cluster statistics: mean size $S$ (1st and 2nd columns) and correlation length $\xi$ (3rd and 4th columns) plotted against probability $p$ and $1 - p/p_{\text{max}}$ corresponding to the FeNi, CoNiFe, CoNiCrFe, CoCrFeMn, CoNiCrFeMn, and Co$_5$Cr$_2$Fe$_{40}$Mn$_{27}$Ni$_{26}$ glasses. The (red) curves indicate binning averaged data. The dashdotted lines denote power laws $S \propto (p_{\text{max}} - p)^{-\gamma}$ and $\xi \propto (p_{\text{max}} - p)^{-\nu}$. The scaling exponents $\gamma$ and $\nu$ are reported in Table 1.