# Structural Signature of Plastic Deformation in Metallic Glasses

H. L. Peng,[1,2] M. Z. Li,[2,*] and W. H. Wang[1]
[1]Institute of Physics, Chinese Academy of Sciences, Beijing 100190 China
[2]Department of Physics, Renmin University of China, Beijing 100872 China

(Received 14 December 2010; published 30 March 2011)

The structure feature of a model CuZr metallic glass during deformation is investigated by molecular dynamics simulations. A spatially heterogeneous irreversible rearrangement is observed in terms of nonaffine displacement. We find that regions with smaller nonaffine displacement have more Voronoi pentagons, while in those with larger nonaffine displacement other types of faces are more populated. We use the degree of local fivefold symmetry (LFFS) as the structural indicator to predict plastic deformation of local structures and find that the plastic events prefer to be initiated in regions with a lower degree of LFFS and propagate toward regions with a higher degree of LFFS.

DOI: 10.1103/PhysRevLett.106.135503 PACS numbers: 62.20.F−, 61.43.Bn, 61.43.Dq

In amorphous solids, it is crucial to find a good indicator parameter to establish the structure-properties correlation. For instance, the dynamical indicator of the Debye-Waller factor has been quite successful for predicting the relative long-time dynamical heterogeneity and irreversible arrangement in glass-forming liquids [1]. Because of the lack of an *a priori* structural indicator such as the dislocation in crystalline counterparts, it is difficult to define a local geometric parameter as energy dissipative units for understanding the microscopic plastic deformation mechanism in metallic glasses (MGs) [2–5]. While the theory of shear transformation zone (STZ) has been proposed to explain the deformation mechanism [6–8], it did not provide any information on the specific structural feature accounting for plastic domains in disorder solids. Free volume and local potential energy proposed as structure indicators [9–12] have been demonstrated to be unable to provide a clear structure picture of the atomic symmetry or chemical ordering of local environments, either [12].

Because the icosahedral short-range order (ISRO) in MGs has a special atomic arrangement and a close relation with glass-forming ability (GFA) and mechanical properties [11–16], it is also adopted as a structural indicator for the fundamental process underlying structural relaxation [12]. However, in some MGs the ISRO is absent [17]. On the other hand, a perfect icosahedral cluster and a distorted one may exhibit different mechanical behaviors, due to the different atomic symmetry or local packing. Therefore, in addition to ISRO, atomic symmetry may be a more general concept in glass-forming alloys in which both local fivefold and translational symmetries are present [18,19]. So far, NMR experiments have shown that local cluster symmetry plays an important role in GFA of MGs [20]. Molecular dynamics (MD) studies also found the unique behavior of the Voronoi pentagons during the quenching process [21]. These imply that the degree of the local fivefold symmetry (LFFS) is crucial for the understanding of structural relaxation and mechanical properties in MGs.

In this Letter, we performed MD simulation studies with the LAMMPS code [22] and used the degree of LFFS as the structural indicator to identify and characterize plastic deformation in MGs. The fraction of pentagons in each local cluster was adopted to quantify the degree of LFFS. We find that plastic deformation starts from regions with a lower degree of LFFS, but spreads out toward regions with a higher degree of LFFS during deformation. The results show that the degree of LFFS in local structures is a key factor of controlling mechanical properties, and has implications for the atomistic scale understanding of the formation of STZ and shear bands as well as plastic deformation in MGs.

In our MD simulations, the model system of $Cu_{50}Zr_{50}$ MG was chosen with a realistic embedded-atom method potential [23]. The system contains 40 000 atoms with periodic boundary conditions. In the process of sample preparation, it was first melted and equilibrated at $T = 2000$ K, then cooled down to 300 K with a cooling rate of $1.0 \times 10^{13}$ K/s, during which the cell size was adjusted to give zero pressure in the $NPT$ ensemble. After that, compressive deformation was applied along the $Y$ axis with a strain rate of 0.25/ns at $T = 300$ K and the period boundary conditions on three dimensions. After each compression of 0.111 Å, the structure was relaxed for 10 ps (5000 MD steps). The local irreversible rearrangement of atomic configuration is evaluated by the nonaffine displacement of the central atom $i$ relative to its neighbor atoms $j$ [7]:

$$D_i^2 = \frac{1}{N}\sum_j [\vec{r}_j(t) - \vec{r}_i(t) - \vec{\varepsilon}(\vec{r}_j(t+\Delta t) - \vec{r}_i(t+\Delta t))]^2,$$

where $N$ is the number of the nearest neighbors of atom $i$ determined by Voronoi analysis, $\vec{r}_i(t)$ is the position of atom $i$ at time $t$, $\Delta t$ is the time interval for the plastic rearrangement, and $\vec{\varepsilon}$ is the maximum local elastic strain tensor [7]. Here we set $\Delta t = 10$ ps.

Figure 1 illustrates the nonaffine displacement $D^2$ each atom experiences under compressive deformation in $Cu_{50}Zr_{50}$ MG. The regions with larger $D^2$ are highly localized, and the structure response to the deformation is inhomogeneous. This implies that the local structural environments of the system and the corresponding plastic rearrangement are quite different for strain localization. As strain increases, regions with larger $D^2$ expand as shown in Fig. 1(b). This indicates that the initial glass structures determine the positions of strain localization, and strain localized regions are expanded with strain increasing.

To analyze the local structure feature, the Voronoi tessellation method was employed. It can identify the number of $i$-edged ($i = 3, 4, 5, 6, \ldots$) faces $n_i$ in a polyhedron by the Voronoi index $\langle n_3, n_4, n_5, n_6, \ldots \rangle$. An $i$-edged face reflects the local symmetry of the central atom with some nearest-neighbor atoms in a certain direction. The triangle, tetragon, and hexagon faces have the local translational symmetry feature, while pentagon faces reflect the LFFS (e.g., an icosahedral polyhedron consists of 12 pentagon faces) [18,19,21]. A drastic increase of the fraction of pentagons in contrast to other type faces in glass transition has been found [21]. It is intriguing to know how the plastic deformation correlates with these types of faces. Here we use the fraction of the number of pentagons and the number of the nearest-neighbor atoms to define the degree of LFFS in local structures as $d_5 = n_5/\sum_i n_i$.

Figure 2(a) shows the correlation between the nonaffine displacement $D^2$ and the degree of LFFS $d_5$ in Voronoi clusters. It is shown that $D^2$ decreases as $d_5$ increases in local structures. This indicates that the regions with a lower degree of LFFS can be easily deformed, and the plastic deformation is more difficult to occur in those regions with a higher degree of LFFS. In contrast to the pentagons, the nonaffine displacements become bigger as the fraction of triangle, tetragon, or hexagon faces increases in Voronoi clusters. Therefore, the results show that the nonaffine displacement often takes place in the regions with a higher degree of local translational symmetry, or a lower degree of LFFS. The higher the degree of LFFS, the lower the nonaffine displacement is in these regions. Icosahedral clusters would have the lowest probability to arise the irreversible rearrangement around such local environments. These findings are consistent with previous simulation results [21,24]. From the above analysis, we can see that the degree of LFFS as a structural indicator in MGs can simply capture the structural features responding to plastic deformation.

According to the definition of $d_5$, the Voronoi clusters will have the same $d_5$ if the indices are the same. However, their mechanical behavior may be different due to the different atomic packing of local environments, such as a perfect icosahedral cluster and a distorted one. To examine the packing effect on plastic deformation, the common-neighbor analysis was performed, which can get the information of both atomic symmetry and local packing [19]. Figure 2(b) shows the correlation between nonaffine displacement and major common-neighbor types. For simplicity, instead of four indices, here we use three indices to identify the common-neighbor types. The first index denotes two nearest-neighbor atoms determined by the above Voronoi analysis, i.e., the root pair. The second one represents the number of nearest neighbors shared by the root pair, i.e., common neighbors. The third index denotes the number of bonds formed among the common nearest-neighbor atoms. Here we typically took the first minimum of the partial pair correlation functions as the distance cutoff to determine whether a bond is formed between two common atoms. The 155 index represents a fivefold ring of common neighbors with all nearest neighbors bonded. 154 or 153 corresponds to the fivefold rings with one or two bonds broken because the distance between them is beyond the cutoff. The types of 166, 165, 144, and 142 are represented in the bulk bcc, fcc, and hcp crystals, respectively [19]. Therefore, we can obtain the information of both the local atomic symmetry and the way of atomic packing. As shown in Fig. 2(b), local structures with all bonded fivefold rings are the most difficult to be deformed compared to others. However, if the all bonded rings are distorted, some common-neighbor atoms are packed less densely and a few bonds may be broken. The local structures become unstable and easily deformed, and more severely distorted local structures can afford larger plastic deformation as shown in Fig. 2(b). This implies that the

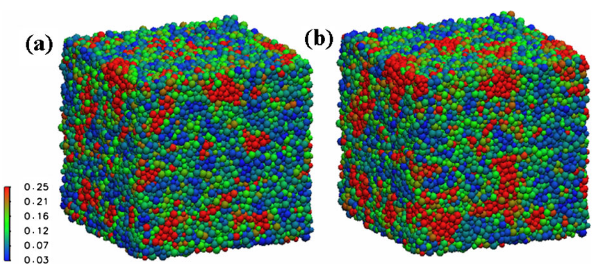

FIG. 1 (color online). Atomistic configurations of the nonaffine displacement of $D^2$ illustrated by colors at the strain of 5% when $\Delta t = 10$ ps (a) and $\Delta t = 40$ ps (b).

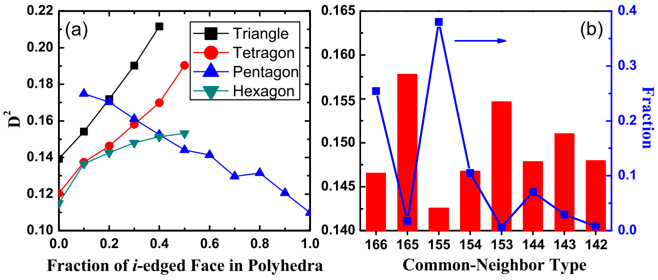

FIG. 2 (color online). (a) The correlation between the nonaffine displacements and the fraction of $i$-edged faces in polyhedra. (b) The correlation of the nonaffine displacements with the common-neighbor types and their fractions.

packing of local structures significantly influences plastic deformation. Although local structures with distorted fivefold rings can be easily deformed, plastic deformation is still much more easily triggered and propagates in regions with a lower degree of LFFS, because the total population of the 154 and 153 is much lower than that of 166, 165, 144, 143, and 142.

Next, we investigate how plastic events propagate during deformation, which is closely related to the mechanical properties of MGs [5]. We chose a critical value of $D_c^2$ (e.g., 0.2) to justify whether a local structure experiences a plastic deformation ($D^2 > D_c^2$) or not ($D^2 < D_c^2$). With this threshold, we can examine how many plastically deformed local structures are emerging in the time intervals of $(t, t + i\Delta t, i = 1, 2, 3, \ldots)$, and then examine the structural properties of the newly deformed local structures in the time intervals and analyze the propagation trends of the plastic events during deformation. As shown in Fig. 3(a), with strain increasing, the plastic events take place gradually from regions with a lower degree of LFFS toward those with a relatively higher degree of LFFS. This indicates that in the short time interval, plastic events tend to take place in regions with a lower degree of LFFS. However, in longer time intervals, they become saturated in those regions, and have to propagate to regions with a higher degree of LFFS. Figure 3(a) also shows that the different choices of $D_c^2$ do not affect the general propagation trend of plastic events and similar results are obtained. However, as $D_c^2$ increases, the average degree of LFFS $d_5$ decreases. This also implies that the larger plastic events tend to occur and propagate in regions with a lower degree of LFFS.

Figure 3(b) shows the distribution of the average degree of LFFS $d_5$ with the nonaffine displacement $D^2$. It is clear that for smaller and moderate $D^2$, the averaged $d_5$ is relatively higher, and decreases exponentially as $D^2$ increases. For larger $D^2$, however, averaged $d_5$ is almost not changed with it. This indicates that at the early stage of deformation, local structures respond to it accordingly, lowering the degree of LFFS in them. As they are deformed to some extent, local structures reach a saturated situation in which the degree of LFFS cannot be reduced anymore. Further deformation will be transformed into the less deformed regions where the degree of LFFS is relatively higher as illustrated in Fig. 3(a).

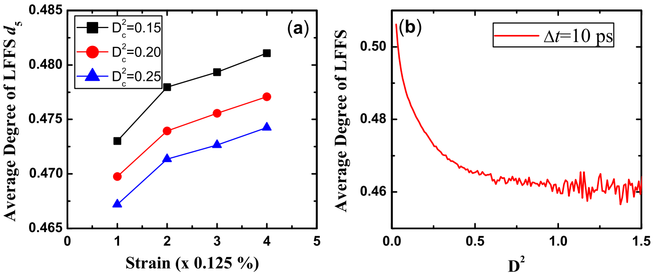

FIG. 3 (color online). (a) The variation of the average $d_5$ with strain; (b) the distribution of the average $d_5$ with the nonaffine displacement.

We also compare the structural indicator of the degree of LFFS and the spatial distribution of plastic events as shown in Fig. 4. The black points represent the atoms having a higher degree of LFFS in a structural configuration. The red (dark gray) areas are the mostly deformed regions that emerged in the time interval of 40 ps, while the blue (light gray) areas present the less deformed regions. As shown in Fig. 4, the atomic irreversible rearrangement during deformation tries to avoid local structures having a higher degree of LFFS, preferring to occur in regions with a lower degree of LFFS. We note that this is not absolute, and a few irreversible rearrangements also occur in regions with a higher degree of LFFS as shown in Fig. 4. This is consistent with the above analysis, local structures with the same degree of LFFS exhibiting different mechanical behavior, because of the different atomic packing in local environments. Statistically, however, regions with a lower degree of LFFS still play an essential role in the potential plastic deformation zones in MGs, and the regions with a higher degree of LFFS play the role of preventing the propagation of the plastic events during deformation.

To understand how local atomic symmetry correlates with the macroscopic quantities such as yield stress, we investigated the structural features and measured the yield stress of samples obtained with different cooling rates. It is found that the degree of LFFS increases as the cooling rate decreases, as shown in Fig. 5, which is consistent with previous studies [24]. The results indicate that the higher the degree of LFFS a metallic glass structure contains, the higher the yield stress of the structure. Therefore, the increase of the degree of LFFS may lead to the increase of the yield stress of MGs.

It should be noted that purely localized parameters such as potential energy and free volume cannot describe well

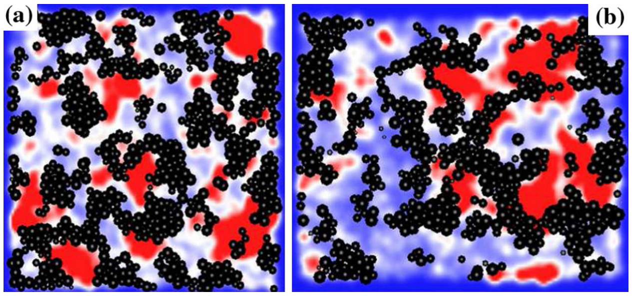

FIG. 4 (color online). Correlation between the structures having a higher degree of LFFS and the irreversible rearrangement during deformation. The slice in the middle of the samples along $Z$ direction with thickness of the first minimum of the pair correlation function was taken for the illustration. Black points denote the atoms having a higher degree of LFFS ($d_5 \geq 0.5$) at the strain of (a) 5% and (b) 10%. The colors represent values of $D^2$ with $\Delta t = 40$ ps.

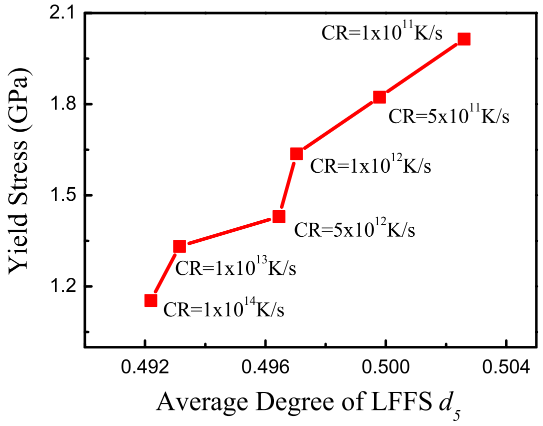

FIG. 5 (color online). Correlation between the yield stress and averaged $d_5$ measured with different cooling rates (CR).

the irreversible rearrangement events in MGs [9–12]. For the structural indicator of the degree of LFFS, however, it reflects specific atomic symmetry and local packing in local structural environments. In fact, the structural indicator of the degree of LFFS has included the information of potential energy and free volume. It is shown that the pentagons have low potential energy which contributes to local structural stability [21]. Meanwhile, the free volume decreases as the fraction of pentagons increases [21], indicating the structural configuration of pentagons is packed more densely. According to our analysis, the regions with a higher degree of LFFS and less free volumes are harder, while regions with more local translational symmetry and more free volumes are softer. The corresponding hard and soft zones have been observed in plastic MGs [25]. On the other hand, the strain localized regions shown in Fig. 1 should correspond to the STZs based on the STZ model [7]. Therefore, STZs could be initially formed in regions with a lower degree of LFFS, and propagate to regions with a relatively higher degree of LFFS. We also investigated the correlation between local atomic stress [26] and the degree of LFFS; no explicit correlation was found.

We note that the order of LFFS is inhomogeneous. Previous studies show that structures containing a higher degree of LFFS tend to form a stringlike order, which makes the structures of MGs inhomogeneous [15]. The heterogeneity of the local atomic symmetry may lead to the heterogeneity of the mechanical response as shown in Fig. 1, and guide the propagation of the plastic events. During deformation, the structures of MGs evolve from the local fivefold symmetry to the local translational symmetry, becoming more homogeneous.

In conclusion, we demonstrate that a structural indicator of the degree of LFFS can be used to characterize the mechanical properties of MGs. The structures with a lower degree of LFFS can accommodate larger plastic deformation, and the irreversible rearrangement is more difficult to be induced in regions with a higher degree of LFFS. On the other hand, the plastic events propagate in regions with a lower degree of LFFS, and expand toward regions with a higher degree of LFFS as strain is increasing. The heterogeneity of the local atomic symmetry leads to the inhomogeneous plastic deformation in MGs.

The work was supported by NSF of China (No. 51071174, No. 50731008, and No. 50921091), MOST 973 (No. 2007CB613904), and the Fundamental Research Funds for the Central Universities, and the Research Funds of Renmin University of China (No. 10XNJ002).

---

*maozhili@ruc.edu.cn

[1] A. Widmer-Cooper and P. Harrowell, Phys. Rev. Lett. **96**, 185701 (2006).
[2] J. J. Lewandowski, W. H. Wang, and A. L. Greer, Philos. Mag. Lett. **85**, 77 (2005).
[3] C. A. Schuh, T. C. Hufnagel, and U. Ramamurty, Acta Mater. **55**, 4067 (2007).
[4] A. R. Yavari, J. J. Lewandowski, and J. Eckert, MRS Bull. **32**, 635 (2007).
[5] M. W. Chen, Annu. Rev. Mater. Res. **38**, 445 (2008).
[6] A. S. Argon, Acta Metall. **27**, 47 (1979).
[7] M. L. Falk and J. S. Langer, Phys. Rev. E **57**, 7192 (1998).
[8] D. Pan *et al.*, Proc. Natl. Acad. Sci. U.S.A. **105**, 14 769 (2008).
[9] F. Spaepen, Acta Metall. **25**, 407 (1977).
[10] W. L. Johnson and K. Samwer, Phys. Rev. Lett. **95**, 195501 (2005).
[11] D. B. Miracle *et al.*, MRS Bull. **32**, 629 (2007).
[12] Y. Q. Cheng, E. Ma, and H. W. Sheng, Appl. Phys. Lett. **93**, 051910 (2008).
[13] F. C. Frank, Proc. R. Soc. A **215**, 43 (1952).
[14] H. Jonsson and H. C. Andersen, Phys. Rev. Lett. **60**, 2295 (1988).
[15] M. Z. Li *et al.*, Phys. Rev. B **80**, 184201 (2009); H. L. Peng *et al.*, Appl. Phys. Lett. **96**, 021901 (2010).
[16] A. Di Cicco *et al.*, Phys. Rev. Lett. **91**, 135505 (2003).
[17] L. Huang *et al.*, Phys. Rev. B **81**, 014108 (2010).
[18] P. J. Steinhardt, D. R. Nelson, and M. Ronchetti, Phys. Rev. B **28**, 784 (1983).
[19] J. D. Honeycutt and H. C. Andersen, J. Phys. Chem. **91**, 4950 (1987).
[20] X. K. Xi *et al.*, Phys. Rev. Lett. **99**, 095501 (2007).
[21] M. Wakeda *et al.*, Intermetallics **15**, 139 (2007).
[22] http://lammps.sandia.gov.
[23] M. I. Mendelev *et al.*, J. Appl. Phys. **102**, 043501 (2007); **102**, 093518 (2007); **104**, 123532 (2008).
[24] Y. Q. Cheng *et al.*, Acta Mater. **56**, 5263 (2008).
[25] Y. H. Liu *et al.*, Science **315**, 1385 (2007).
[26] T. Egami *et al.*, Phys. Rev. B **76**, 024203 (2007).