

Stress-versus temperature-induced structural evolution in metallic glasses

H. L. Peng, M. Z. Li, and W. H. Wang

Citation: *Appl. Phys. Lett.* **102**, 131908 (2013); doi: 10.1063/1.4800531

View online: <http://dx.doi.org/10.1063/1.4800531>

View Table of Contents: <http://apl.aip.org/resource/1/APPLAB/v102/i13>

Published by the [American Institute of Physics](#).

Additional information on *Appl. Phys. Lett.*

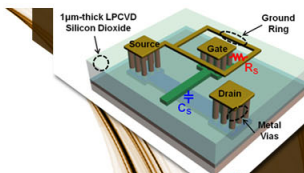
Journal Homepage: <http://apl.aip.org/>

Journal Information: http://apl.aip.org/about/about_the_journal

Top downloads: http://apl.aip.org/features/most_downloaded

Information for Authors: <http://apl.aip.org/authors>

ADVERTISEMENT

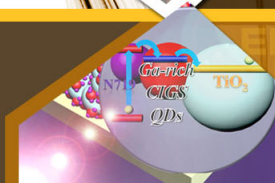


SURFACES AND INTERFACES

Focusing on physical, chemical, biological, structural, optical, magnetic and electrical properties of surfaces and interfaces, and more...

EXPLORE WHAT'S
NEW IN APL

SUBMIT YOUR PAPER NOW!



ENERGY CONVERSION AND STORAGE

Focusing on all aspects of static and dynamic energy conversion, energy storage, photovoltaics, solar fuels, batteries, capacitors, thermoelectrics, and more...

Stress-versus temperature-induced structural evolution in metallic glasses

H. L. Peng,^{1,2} M. Z. Li,^{2,a)} and W. H. Wang¹

¹*Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*

²*Department of Physics, Renmin University of China, Beijing 100872, China*

(Received 15 November 2012; accepted 25 March 2013; published online 4 April 2013)

Structure evolution induced by shear deformation was investigated via molecular dynamic simulation on CuZr metallic glass system and compared with that induced by temperature. Voronoi tessellation analysis found that the local structures evolve to a liquid-like state as shear stress increases, similar to the temperature-induced structure evolution. However, shear stress induces little change to the radial distribution functions (RDFs) compared to temperature, indicating that the global glassy state still sustains. This is in contrast to the opinion that shear deformation may be similar to temperature increase and induce a metallic glass system to experience glass-liquid transition. The analysis of anisotropic part of RDFs reveals that shear deformation can induce significant anisotropic structural evolution, while pure increase of temperature cannot. Our findings demonstrate that shear deformation may induce different structural evolution in metallic glasses compared to temperature.

© 2013 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4800531>]

It has been expected that the external stress could play a similar role in glass-liquid transition as temperature does in amorphous solids.^{1–3} De-vitrification can be induced by stress when the glass sample is yield, which is analogous to the glass-liquid transition purely caused by increasing temperature. In the view of energy landscape, shear stress can make a local energy minimum disappear, promote atoms to jump into a new local energy minimum, thus enhance the structural relaxation and self-diffusion.⁴ The stress-biased relaxation had been purely separated from temperature in athermal process in molecular dynamics (MD) simulations by Nandagopal and Utz.⁵ It is found that the dynamics caused by thermal relaxation and athermal deformation is strikingly similar, which supports the concept of a strain-induced glass-liquid transition or shear-based relaxation.⁶ Recently Guan *et al.* formulated the equivalent effect of temperature and stress on viscosity in steady-flow region in metallic glasses.⁷ However, despite of these dynamical evidences for the similar effect of temperature and stress, no explicit structural evidence has ever been explored for the “temperature effect” of stress. Many experiments have investigated the structural evolution in the limit of elastic region, but failed in the region upon yield point.^{8–10} On the other hand, so far no computer simulations have been conducted to clarify it, either.¹¹ Therefore, to quantify and compare the structure evolution induced by stress and temperature in amorphous solids is quite important for further understanding both the deformation mechanisms of metallic glasses and the nature of glass transition.

In this work, we investigated the atomic structure evolution under shear deformation with various strain rates and compared it with the structure feature at different temperatures by performing classical MD simulations on CuZr metallic glass system. It is found that for the sheared samples with relatively high shear strain rate, the local structure feature characterized by Voronoi tessellation is very similar to that of the unsheared samples at above glass transition

temperatures. However, isotropic component of radial distribution functions (RDFs) shows that the structure evolution induced by stress is not as significant as that induced by temperature. Based on the fact that the split of second peak holds all the time, stress would not induce glass-liquid transition as temperature does. Anisotropic part of RDF shows that the external biased activation is mostly contained by the anisotropic structural arrangement, while thermal activation is just opposite. All these indicate that the structural evolution induced by stress and temperature is different.

Classical MD simulations were carried out for Cu₅₀Zr₅₀ metallic alloys with realistic embedded-atom method (EAM) potential.¹² The sample contains 40 000 atoms with periodic boundary conditions. It is equilibrated at 2000 K, cooled down to 100 K with a cooling rate of 1.0×10^{13} K/s. Then the sample is heated up to 2000 K with the same heating rate. In the heating process, we get different temperature samples, which are used to compare with the sheared samples under different strain rates. At each temperature, the cell size is adjusted to zero pressure in isothermal-isobaric (NPT) ensemble. By examining the change of potential energy (PE) with temperature in the cooling and heating processes, it is found that the glass transition temperature is 690–700 K on cooling, while 790–800 K under heating. In order to get homogeneous shear deformation, velocity controlling method is used with SLLOD equation of motion and Lees-Edwards boundary condition.^{13,14} Mechanical shear deformation is conducted at 100 K with 11 different strain rates (denoted as R0, R1, ..., R10, respectively) as listed in Table I. The corresponding flow stresses were also presented. All data analyzed below were collected in plastic flow region, which indicates that the shear stress reaches steady state.

Voronoi tessellation method was employed to characterize the local structures with Voronoi index $\langle n_3, n_4, n_5, n_6, \dots \rangle$, where n_i ($i = 3, 4, 5, 6, \dots$) represents the number of i -edged face which reflects the local symmetry of the central atom with some nearest-neighbor atoms in a certain direction.¹⁵ Based on the Voronoi analysis, a structural indicator of local five-fold symmetry (LFFS) is defined as the fraction of the

^{a)}Email: maozhili@ruc.edu.cn

TABLE I. Different shear strain rate imposed on $\text{Cu}_{50}\text{Zr}_{50}$ metallic glasses at 100K. Flow stress is calculated as the steady-state shear stress in plastic flow region. Here, $\dot{\gamma}_0 = 3.125 \times 10^8/\text{s}$.

Sample	R0	R1	R2	R3	R4	R5	R6	R7	R8	R9	R10
Strain rate ($\times \dot{\gamma}_0$)	2^0	2^1	2^2	2^3	2^4	2^5	2^6	2^7	2^8	2^9	2^{10}
Flow stress (GPa)	0.89	0.91	0.95	0.98	1.04	1.12	1.24	1.41	1.66	2.04	2.71

5-edged face in a Voronoi polyhedron, $d_5 = n_5 / \sum_i n_i$, to describe the local atomic symmetry feature.¹⁶ It has been proved to play important roles in liquid-glass transition and mechanical deformation in metallic glasses.^{15–19} We also defined local configurational entropy as

$$S = - \sum_{i=1}^N P_i \log P_i,$$

to characterize the local structure distribution, in which the type of Voronoi polyhedra was used. Here, P_i represents the fraction of Voronoi type i in a sample. If P_i is 1 and the probability of all other types of polyhedra is 0, then $S = 0$, which indicates that the system is in the most ordered structure configuration. However, if all the structure types have the same probability of $1/N$, S will get its maximum value, which indicates that the system is in the most disordered configuration.

Figure 1 shows the changes of the PE, the average LFFS, and Voronoi entropy with temperature in heating process. As shown in Figs. 1(b) and 1(c), at low temperature average LFFS and Voronoi entropy both approximately exhibit a linear change with temperature increasing. And they have drastic drop or increase at about 800 K, which indicates that the glass to liquid transition occurs, consistent with the change of PE with temperature change shown in Fig. 1(a). This indicates that LFFS and Voronoi entropy can well characterize the structure change in metallic glasses.

This situation sustains when the external force is applied as shown in Figs. 1(d) and 1(e). With strain rate increasing, the average LFFS decreases, while Voronoi entropy increases. This trend is similar to that in the case of temperature change. Especially, at the highest strain rate (i.e., R10), the value of LFFS or Voronoi entropy is almost equal to that at 1050 K as shown in Figs. 1(b) and 1(c). This indicates that the glass to liquid transition can be induced not only by temperature but also by external stress. Note that the behavior of LFFS and Voronoi entropy with temperature shown in Figs. 1(b) and 1(c) is very different from that with strain rate shown in Figs. 1(d) and 1(e). This is because strain rate is not directly connected with the equivalence of temperature.⁷

The average LFFS and Voronoi entropy provide the average structure information of a system. We also compared the specific local structural change induced by both temperature and stress. Figure 2 shows the evolution of LFFS distribution with temperature and shear strain rate, respectively. It can be clearly seen that temperature and strain rate induce similar change of LFFS distribution. The left part of LFFS distribution is increasing while the right part is decreasing. As temperature or shear stress increases, the local structures

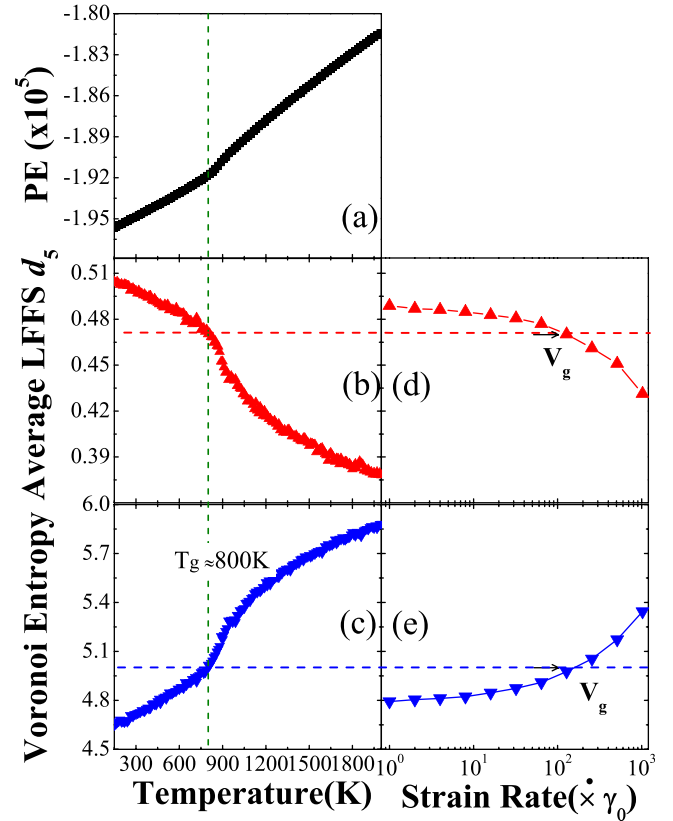


FIG. 1. The change of PE (a), average LFFS (b), and Voronoi entropy (c) with temperature in heating process and the change of average LFFS (d), and Voronoi entropy (e) with strain rate. The glass-liquid transition in heating process occurs at about 800 K. V_g in (d) and (e) represents the value of LFFS (d_5) and Voronoi entropy at glass transition temperature in (b) and (c).

with higher LFFS are destroyed, while those with lower LFFS are formed. This is consistent with previous studies that the population of icosahedral clusters becomes less with temperature increasing¹⁵ or stress increasing.^{17,18} By comparing the distribution of LFFS (d_5), it can be found that the distribution at 1100 K is very close to that at strain rate of R10 as shown in Fig. 2. This implies that the local structure in R10 is very similar to that at 1100 K. On the other hand, 1100 K is well above glass transition temperature. In this sense, a similar glass-liquid transition could be induced to the local structures, if the shear stress is large enough.

Based on the above analysis, it can be seen that shear stress can induce structural evolution from glassy to liquid state just as temperature does. However, we also investigated the RDF which shows that temperature and shear stress may have different but very important effect on structural evolution. Figures 3(a) and 3(b) show the RDFs in unsheared samples at different temperatures and in sheared samples with different strain rates, respectively. In the unsheared samples, temperature increasing makes the RDF change significantly, the amplitude of the first peak decreasing, and the split of the second peak disappearing as shown in Fig. 3(a). However, in the sheared samples, RDF does not change much with strain rate increasing as shown in Fig. 3(b). The first peak of RDF changes so little that they are almost the same as that in the unsheared sample at 100 K. This indicates that the coordination number (CN) changes little under shear deformation, but significantly as temperature increases. Moreover, the

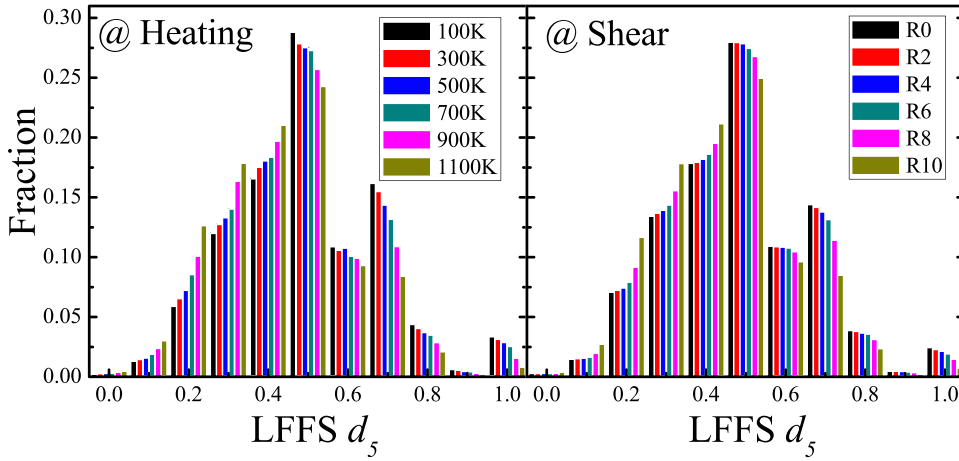


FIG. 2. Distribution of LFFS (d_5) at different temperatures (a) and different strain rates (b).

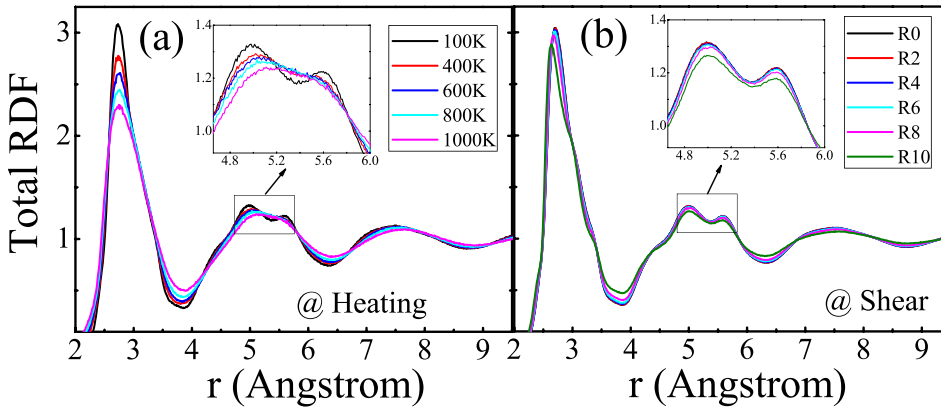


FIG. 3. Total reduced radial distribution function at different temperatures (a) and different strain rates (b).

shoulder of the second peak in RDFs changes little with strain rate increasing as shown in Fig. 3(b), and the shape of the second peak is almost the same as that in the unsheared sample. Since the shoulder of second peak in RDF is considered as one of the most hallmarks for glass state,^{20–22} the persistence of this shoulder as strain rate increases indicates that the global glassy state sustains during shear deformation. In this sense, stress might not induce the glass to liquid state transition as temperature does.

It should be noted that RDFs shown in Fig. 3 are isotropic, while a non-hydrostatic deformation can often induce anisotropy on local environment.²³ Thus, it is necessary to investigate the anisotropic part for better understanding the structural evolution. The anisotropy can be evaluated by expanding RDFs with the spherical harmonic function $Y_l^m(x)$ as: $g(\vec{r}) = \sum_{l,m} g_l^m(r) Y_l^m(\frac{\vec{r}}{r})$. For axial symmetry, m equals 0, and the anisotropic terms with $l > 2$ can be neglected. The non-zero terms are $l=0$ and $l=2$, which represent the isotropic and elliptical components, respectively (see Ref. 24 for details).

With this method, we evaluated the isotropic and anisotropic terms of RDFs. For isotropic term $g_0^0(r)$, which can recover the radial distribution function, it shows completely the same behavior as shown in Fig. 3. However, anisotropic term $g_2^0(r)$ presents a different behavior (see Figure 4). With strain rate increasing, the anisotropy becomes more and more obvious. Since the data were collected in homogeneous steady-flow region, the amplitude of any peak in $g_2^0(r)$ would be proportional to the applied force.²⁴ It indicates that the external stress is mostly maintained by anisotropic rearrangement of

structure, while the isotropic component will persist as the unsheared state. This situation is reversed as temperature increases. Anisotropic component is almost zero with temperature increasing (see the inset in Fig. 4). This shows that the temperature only changes the isotropic component but fails to the anisotropic part. In this sense, the effect of temperature and stress on the structure evolution is very different.

The result of anisotropic evolution is consistent with *in situ* neutron diffraction⁹ and x-ray diffraction⁸ results that the direction of nearest neighbor atoms pairs will

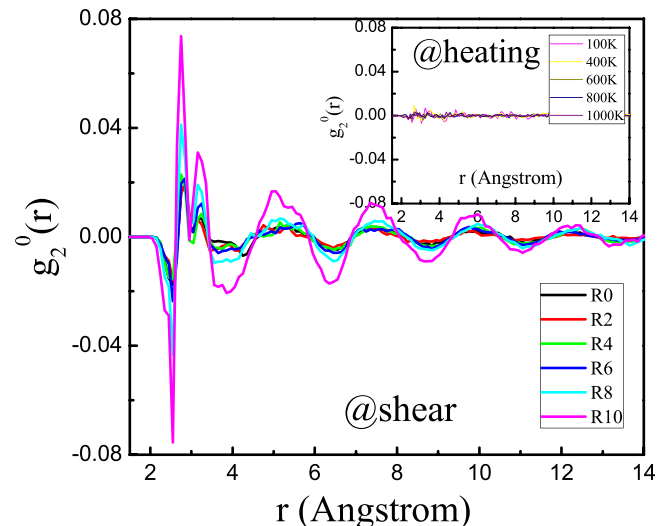


FIG. 4. Anisotropic radial distribution function at different strain rates and different temperatures (inset).

re-orientate along axis of uniaxial deformation in elastic deformation region, as well as some computer simulation results.²⁵ However, this anisotropy cannot be reflected in any structural analysis with distance cutoff, for example, Voronoi tessellation, bond angle distribution, etc., because the isotropic atom-interaction potential will try to recover this anisotropy by breaking bonds along the polarization direction and forming new bonds on the perpendicular direction.⁸ With stress increasing, the anisotropic polarization becomes more and more serious. However, in a certain radial distance cutoff, the number of nearest neighbors sustains. Meanwhile, polarization promotes the local structures to change their symmetric properties (i.e., becomes less degree of LFFS) to achieve flow states. That is why we see almost the same local structural evolution induced by stress and temperature shown in Figs. 1 and 2.

In conclusions, shear stress can induce the local geometric evolution from glass to liquid like state, especially for the local symmetric property and the degree of disorder. This effect is very similar to the structural change induced by temperature. However, isotropic component of radial distribution function indicates that the global glassy state will persist no matter how high strain rate is imposed. This suggests that no glass-liquid transition occurs on structure. Anisotropic part of RDF indicates the local structural evolution induced by stress is mainly achieved by the anisotropic polarization of atomic structure rearrangement. For thermal activation, only isotropic rearrangement happens. These findings show that despite of some evidences on dynamics, the effect of stress would not be the same as temperature on the aspect of glassy structures.

This work was supported by NSF of China (Nos. 51071174, 51271197, and 51271195), the National Basic Research Program of China (No. 2012CB932704), NCET-

11-0498, the Fundamental Research Funds for the Central Universities, and the Research Funds of Renmin University of China (No. 10XNJ002).

¹A. S. Argon, *Acta Metall.* **27**, 47 (1979).

²M. L. Falk and J. S. Langer, *Phys. Rev. E* **57**, 7192 (1998).

³C. A. Schuh, T. C. Hufnagel, and U. Ramamurty, *Acta Mater.* **55**, 4067 (2007).

⁴D. L. Malandro and D. J. Lacks, *Phys. Rev. Lett.* **81**, 5576 (1998).

⁵M. Nandagopal and M. Utz, *J. Chem. Phys.* **118**, 8373 (2003).

⁶V. Khonik, A. Kosilov, V. Mikhailov, and V. Sviridov, *Acta Mater.* **46**, 3399 (1998).

⁷P. Guan, M. W. Chen, and T. Egami, *Phys. Rev. Lett.* **104**, 205701 (2010).

⁸Y. Suzuki, J. Haimovich, and T. Egami, *Phys. Rev. B* **35**, 2162 (1987).

⁹N. Mattern, J. Bednarcik, S. Pauly, G. Wang, J. Das, and J. Eckert, *Acta Mater.* **57**, 4133 (2009).

¹⁰G. Wang, N. Mattern, S. Pauly, J. Bednarcik, and J. Eckert, *Appl. Phys. Lett.* **95**, 251906 (2009).

¹¹Y. Q. Cheng and E. Ma, *Prog. Mater. Sci.* **56**, 379 (2011).

¹²M. I. Mendeleev, D. K. Rehbein, R. T. Ott, M. J. Kramer, and D. J. Sordelet, *J. Appl. Phys.* **102**, 093518 (2007).

¹³D. J. Evans and G. P. Morris, *Statistical Mechanics of Nonequilibrium Liquids* (Academic, New York, 1990).

¹⁴A. W. Lees and S. F. Edwards, *J. Phys. C* **5**, 1921 (1972).

¹⁵M. Wakeda, Y. Shibutani, S. Ogata, and J. Park, *Intermetallics* **15**, 139 (2007).

¹⁶H. L. Peng, M. Z. Li, and W. H. Wang, *Phys. Rev. Lett.* **106**, 135503 (2011).

¹⁷Y. Q. Cheng, E. Ma, and H. W. Sheng, *Appl. Phys. Lett.* **93**, 111913 (2008).

¹⁸Y. Q. Cheng, A. J. Cao, H. W. Sheng, and E. Ma, *Acta Mater.* **56**, 5263 (2008).

¹⁹Ch. E. Lekka, A. Ibenskas, A. R. Yavari, and G. A. Evangelakis, *Appl. Phys. Lett.* **91**, 214103 (2007).

²⁰C. H. Bennett, *J. Appl. Phys.* **43**, 2727 (1972).

²¹A. Huerta, D. Henderson, and A. Trokhymchuk, *Phys. Rev. E* **74**, 061106 (2006).

²²S. P. Pan, J. Y. Qin, W. M. Wang, and T. K. Gu, *Phys. Rev. B* **84**, 092201 (2011).

²³N. Morito and T. Egami, *Acta Metall.* **32**, 603 (1984).

²⁴W. Dmowski, T. Iwashita, C.-P. Chuang, J. Almer, and T. Egami, *Phys. Rev. Lett.* **105**, 205502 (2010).

²⁵J. Ding, Y. Q. Cheng, and E. Ma, *Appl. Phys. Lett.* **101**, 121917 (2012).