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Unusual vortex-like atomic motion observed for viscoelasticity in metallic glass

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ABSTRACT

Viscoelasticity in metallic glasses (MGs) is commonly attributed to atomic slips or the activation of local shear transformation events. However, based on our extensive molecular dynamics simulations of creep in MGs, we find that a significant portion of viscoelasticity in MGs is mediated through back-and-forth anharmonic motions of atoms. Interestingly, we also show that such anharmonic oscillatory atomic motion stems mainly from collective atomic rotations, which generates local vorticities with an intensity ten folds of the vorticity induced directly by local shear. As deformation proceeds, the "vortex-like" atomic motion shows an increasing correlation with local atomic slips or the activation of shear transformation zones. Our findings indicate that the viscoelastic deformation during creep in MGs is a two-step process, which echoes very well with the recent discovery of two secondary relaxations in a variety of MGs.

1. Introduction

Understanding local plasticity initiation and viscoelasticity in MGs has been a topic of active research for decades [1,2]. Unlike crystalline solids, MGs lack well-defined structural defects that can easily "migrate" to initiate local plasticity [3]. Consequently, various theoretical models were developed to elucidate the atomic origin of local plasticity in MGs. These include the early efforts which linked plastic flow to individual atomic jumps, such as the free volume model [4], or to cooperative shear transformation of a group of atoms, such as the shear transformation zone (STZ) model [5,6] and the cooperative shear model [7]. Currently, in the MG community, there is general agreement that local plasticity initiation in MGs be related to nano-scale inhomogeneity in their amorphous structure [8–10]. STZ activation may be facilitated by geometrically unfavored motifs [11], regions poor in local fivefold symmetry [12] or "flow" units [13], which undergo large non-affine displacements upon mechanical agitation and conceptually behave in a liquid-like manner [14,15].

Aside from these theoretical efforts, extensive experiments were also carried out, aiming at revealing the dynamic origin of local plasticity in MGs through the study of their stress relaxation behaviors [16–20]. According to Refs. [17,21], steady state plastic flows could be associated with α relaxation [17], which involve large-scale inelastic atomic motions [22], and STZ activation could be associated with β relaxation because of the commonality in their activation energies ($\sim$1 eV for β relaxation and $>$ 2 eV for α relaxation) [18]. Recent experiments clearly demonstrated that there are two secondary relaxation processes in MGs and the additional secondary one (termed as the "fast β relaxation" in Refs. [19,20]) possesses an average activation energy much smaller than that of a STZ, falling into a narrow range between 0.3 and 0.6 eV [19,20] and being insensitive to the glass transition temperature T_g [20]. These findings are intriguing, which suggest that there might be a more localized process of fast motion of fewer atoms prior to STZ. However, the atomistic mechanism for such a process, if there is any, is yet to be understood.

In this paper, we intend to investigate the atomistic mechanism for viscoelasticity during creep in MGs with molecular dynamics (MD) simulations. Unlike the previous studies [23,24], we studied the initiation of plastic flow in a model $Zr_{50}Cu_{50}$ (in atomic %) MG subject to a constant stress, i.e. creep. By tracking the trajectories of the individual atoms with the field analyses, we were able to identify the subtle viscoelastic deformation process prior to overall yielding.

2. Materials and methods

The MD simulations were carried out with the embedded atom method (EAM) potential [25]. The model $Cu_{50}Zr_{50}$ system contained 50,000 atoms and had the dimension of $\sim$10 nm $\times$ 10 nm $\times$ 10 nm,

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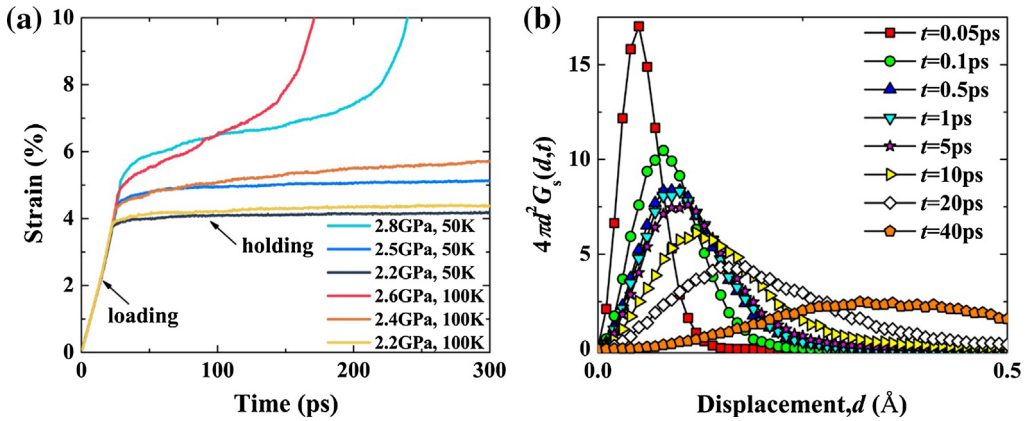

Fig. 1. (a) Strain-time curves under different holding stresses at 50 K and 100 K. (b) Time evolution of the self-part of the van Hove function for $\sigma = 2.8$ GPa at 50 K.

which was subjected to periodic boundary conditions (PBC) for all three dimensions. To obtain a glassy state, the system was initially melted and equilibrated at $T = 2000$ K for 100 ps, and subsequently quenched at a cooling rate of 10^{11} K/s to different temperatures (50 K and 100 K). The whole process was in the isothermal-isobaric (NPT) ensemble controlled via the Nose-Hover thermostat and barostat, respectively [26]. To simulate creep, a series of uniaxial compressive stresses along z direction were applied to the model MG at a nominal stress rate of 0.1 MPa/fs and held for 300 ps.

3. Results and discussion

Fig. 1(a) shows the typical strain–time curves at the holding stress $\sigma_h = 2.2$, 2.5 and 2.8 GPa obtained at the temperature $T = 50$ K, and $\sigma_h = 2.2$, 2.4 and 2.6 GPa at $T = 100$ K. During load hold, the overall strain ε first exhibits transience and develops afterwards into a steady state, which obeys the linear scaling $\varepsilon \propto t$, where t is the time lapse. At a relatively high stress, such as $\sigma_h = 2.8$ GPa at $T = 50$ K and $\sigma_h = 2.6$ GPa at $T = 100$ K, a pronounced acceleration of strain rate can be also observed following the steady state or secondary creep within the simulation time window (see Fig. 1(a)).

To understand the deformation dynamics during load hold, we first calculated the self-part of the van Hove function [27] $G_s(\mathbf{r}, t) = (1/N)\sum_{i=1}^{N}\langle\delta(\mathbf{r}-(\mathbf{r}_i(t)-\mathbf{r}_i(0)))\rangle$, where N is the number of atoms, $\delta(\cdot)$ is the Dirac delta function, $<\cdot>$ represents the ensemble average, and $\mathbf{r}_i(t)$ is the position of atom i at time t. According to Ref. [27], $G_s(\mathbf{r},t)$ is the probability density of an atom moving by a displacement $\mathbf{r}$ over the time lapse t, and $4\pi d^2 G_s(d, t) = (1/N)\sum_{i=1}^{N}\langle\delta(d-|\mathbf{r}_i(t)-\mathbf{r}_i(0)|)\rangle$ is the radial probability density, where d is $|\mathbf{r}|$. As shown in Fig. 1(b), $4\pi d^2 G_s(d,t)$ displays a Gaussian distribution within short time, which indicates independent random motion of atoms and physically corresponds to a Markovian process [28,29]. However, $4\pi d^2 G_s(d,t)$ deviates from the Gaussian form with time and develops a fat tail for large d. Such a non-Gaussian behavior usually signals a collective atomic motion and the rise of dynamic heterogeneities [30,31].

Aside from the self-part of the van Hove function, we computed the mean square displacement (MSD) of atoms during load hold, which is expressed as $\langle d^2(t)\rangle = \langle|\mathbf{r}(t)\text{-}\mathbf{r}(0)|^2\rangle$. Fig. 2 shows the plot of MSD versus t for varied σ_h at 50 K and 100 K. Evidently, MSD is proportional to t^2 for $t < 0.1$ ps, which can be attributed to ballistic atomic motion [32]. As time lapses, a plateau emerges, implicative of restricted atomic motions (or "cage" dynamics) [28,32]. For $t > 5$ ps, one can observe that MSD is proportional to t, which signals diffusive atomic motion [32] after the event of "cage" breaking. The transition from the ballistic to caging regime is stress independent, while the cage breaking shows high stress dependence. In addition, we calculated the two-time correlation function G_4 [33] or the joint probability of an atom moving by

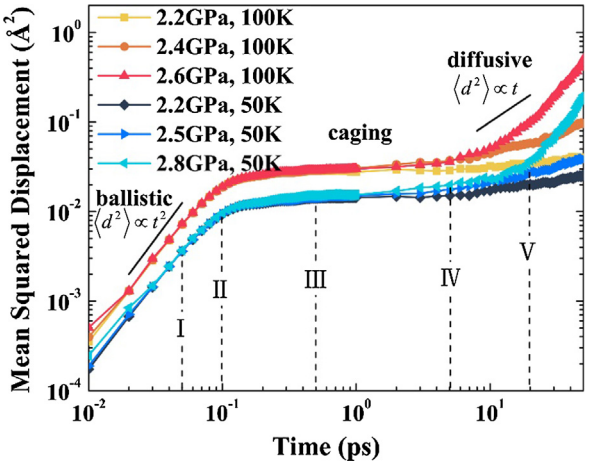

Fig. 2. Mean squared displacement at varied temperatures and stresses. Ballistic, caging and diffusive regime are observed in each case. The cage-trapping plateau is temperature dependent and this transient "cage" breaks up in a shorter time under higher applied stresses. Roman numerals denote five key moments for the curve of $\sigma_h = 2.8$ GPa at 50 K.

d_{1i} in the time window $[0, t]$ and then by d_{2i} in $[t, 2t]$ along the i direction: $G_4(d_{1z},d_{2z},t) = \langle\delta(d_{1z}\text{-}(r_z(t)\text{-}r_z(0)))\delta(d_{2z}\text{-}(r_z(2t)\text{-}r_z(t)))\rangle$, where $i = x$, y, or z, standing for the direction, and r_i is the i coordinate of an atom [33]. To understand the dynamics for the change of deformation mechanisms, we selected five critical moments for study, as marked by the Roman numerals in Fig. 2.

Being keyed to the five key moments for the curve of $\sigma_h = 2.8$ GPa at 50 K in Fig. 2, Fig. 3 display the contour plots of G_4 we obtained through the extensive analysis of the trajectories of atoms. In the ballistic regime, atoms move in a unidirectional manner, as seen in Fig. 3(a)–(c), and the data points accumulate in the first and third quadrats around the trend-line $d_{1i} = d_{2i}$. Interestingly, at the transition from the ballistic motion to cage dynamics, a significant amount of atoms exhibit a back-and-forth mode of movement, as exemplified by Fig. 3(d)–(f), which clearly shows that the G_4 data start to shift from the first and third quadrants with the trend-line $d_{1i} = d_{2i}$ to the second and forth quadrants with $d_{1i} = -d_{2i}$. Note that the magnitudes of the forward and backward atomic movements are generally correlated, which implies a "memory" effect and a non-Markovian process [33]. This trend towards back-and-forth atomic movements becomes prominent with increasing displacement in the caging regime [Fig. 3(g)–(i)], which is sensible since physical confinement can cause a moving atom to bounce back if it undergoes an excessive displacement. Furthermore, it is worth noting that the forward and backward displacement are not exactly equal, which can lead to the build-up of local elastic stresses. As