Stress-Temperature Scaling for Steady-State Flow in Metallic Glasses

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Through computer simulation of steady-state flow in a $Zr_{50}Cu_{40}Al_{10}$ metallic glass using a set of realistic potentials we find a simple scaling relationship between temperature and stress as they affect viscosity. The scaling relationship provides new insight into the microscopic mechanism of shear flow in the glassy state, in terms of the elastic energy of the applied stress modifying the local energy landscape. The results suggest that the plastic flow and mechanical failure in metallic glasses are consequences of stress-induced glass transition.

The absence of a well-defined atomic structure in liquids and glasses presents a major challenge to the effort of elucidating their properties based upon the atomic structure. Understanding mechanical properties of glasses, such as deformation and fracture, is even more difficult to achieve because the phenomena are highly dynamic and heterogeneous down to atomic dimensions. However, through computer simulation we can attain the state of steady flow which is more accessible for analysis. By modeling the flow of atoms within the shear band of metallic glasses with controlled homogeneous steady-state flow we should be able to gain more fundamental understanding of deformation and failure. In this Letter we demonstrate a direct link between stress-induced mechanical flow and glass-to-liquid transition, through a new scaling relationship observed by simulation on a model bulk metallic glass (BMG).

As a model system we chose a Zr-Cu-Al glass which has a relatively low critical cooling rate for glass formation and can readily be cast into a BMG [1]. The model system is made of 4000 atoms representing an alloy of $Zr_{50}Cu_{40}Al_{10}$, interacting with the embedded atom method (EAM) potential [2], and molecular dynamics (MD) simulation was carried out using the open code LAMMPS program [3]. The periodic boundary condition was maintained throughout the simulation, and temperature, T, was kept constant through the Nosé-Hoover thermostat [4]. A glassy state was obtained by melting and equilibrating the system at high temperature (2000 K) for 10 ns, then cooling the liquid to designated temperatures with the cooling rate of 10^{10} K/sec. The change in the thermal expansion coefficient at T_0 (= 860 K) suggests a glass transition at the time scale of the MD simulation. Mechanical shear with a constant rate ($\dot{\gamma}$) was applied through the Lees-Edwards moving boundary condition [5], keeping the temperature and volume constant.

Typical stress-strain curves are shown in Fig. 1 for various strain rates. In this geometry only one stress component, $\sigma = \sigma_{xy}$, is relevant and others are negligibly small. After a brief transient period the system enters the steady-state flow at a shear stress of σ. Below T_0 a yielding peak at σ_{yield} is observed before entering the steady state. The viscosity of the system was calculated from the steady-state stress by $\eta = \sigma/\dot{\gamma}$. It has been shown for a model of granular materials the viscosity-stress data can be nicely collapsed by scaling them with density [6]. We found that our data also collapse when η and σ are scaled with temperature [7] instead of density as a variable,

$$\eta_T = \eta \left| \frac{T}{T_0} - 1 \right|^{\alpha}, \qquad \sigma_T = \sigma \left| \frac{T}{T_0} - 1 \right|^{-\beta}, \quad (1)$$

with $\alpha = 1.23$, $\beta = 0.6$, and $1/\eta_T$ is plotted against σ_T in Fig. 2. This result shows that granular systems and metallic glasses share the common nature of the flow characteristics in the vicinity of $T = T_0$ and $\sigma = 0$, through appropriate scaling of temperature or density. At high temperatures
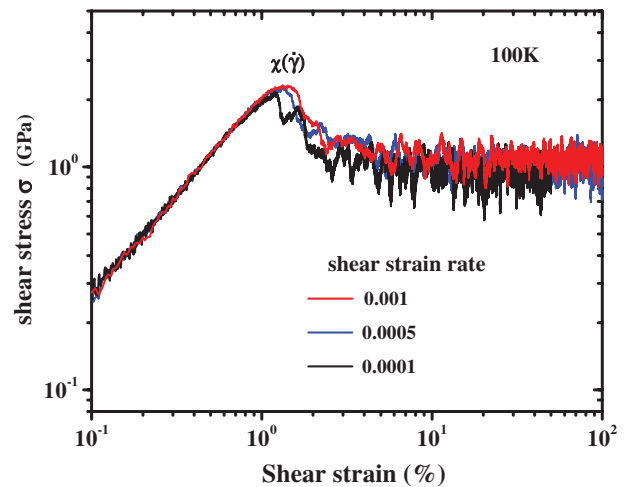

FIG. 1 (color).   Typical stress-strain relations at various shear rates (in the units of ps^{-1}) at $T = 100$ K. The simulation was done for constant volume.
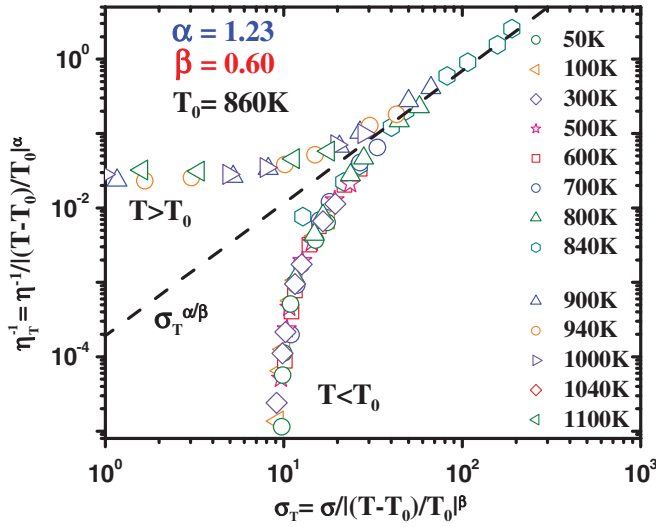
FIG. 2 (color). The plot of scaled viscosity $1/\eta_T$ vs scaled shear stress σ_T for different temperatures.

viscosity usually follows the Vogel-Fulcher-Tammann law, but near the glass transition temperature it often crosses over to the power law as in Eq. (1) [8,9].

In this scaling T_0 is a singular point playing the role of the critical temperature in the conventional critical phenomena. However, the glass transition temperature may not be a true critical point. It is also well known that temperature and stress are intimately coupled in the liquid state, as seen, for instance, in shear thinning, which is observed in all liquids [10,11]. Therefore we enlarged our view and considered the entire T-σ field, rather than focusing on the vicinity of the glass transition at $T = T_0$ and $\sigma = 0$. Figure 3 shows the viscosity as a two-dimensional function of T/T_0 and σ/σ_0, where σ_0 is the critical stress where η diverges upon extrapolation to $T = 0$. Such a plot was attempted for a binary Lennard-Jones glass [12], over a somewhat limited range of temperature. By plotting the results over a wider range of temperature it becomes evident that the curves of constant viscosity are self-similar.

Moreover, it was found that the curves of constant viscosity are given by a surprisingly simple expression,

$$\frac{T}{T_0(\eta)} + \left(\frac{\sigma}{\sigma_0(\eta)}\right)^2 = 1, \qquad (2)$$

where the definitions of T_0 and σ_0 are expanded to be dependent on viscosity. $T_0(\eta)$ is the temperature where viscosity is equal to η when $\sigma = 0$, and $\sigma_0(\eta)$ is the shear flow stress where viscosity is equal to η when $T = 0$. Note that $T_0 = T_0(\infty)$ and $\sigma_0 = \sigma_0(\infty)$. The even (quadratic) power of σ in Eq. (2) is compatible with the symmetry that the sign inversion on $\sigma(\sigma \to -\sigma)$ keeps the physics unchanged. From the scaling in Eq. (1) we obtain

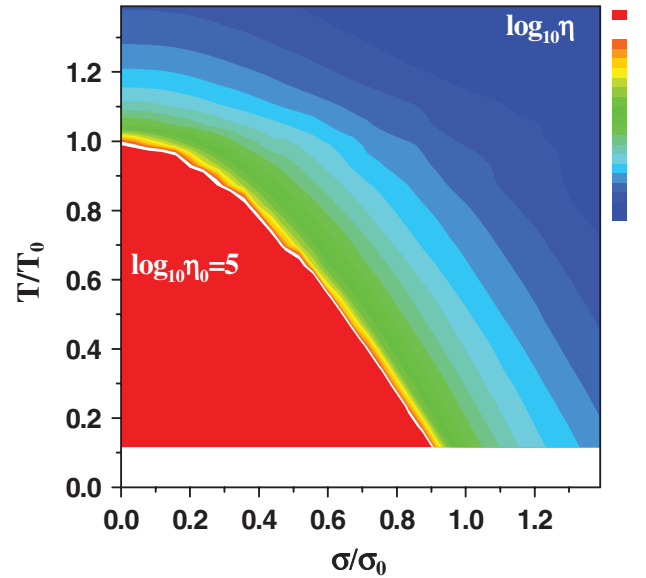FIG. 3 (color). Two-dimensional plot of viscosity as a function of T/T_0 and σ/σ_0, where T_0 is as defined in Fig. 2 and σ_0 is the critical stress where η_0 diverges in extrapolation to $T = 0$ K. The white curve shows the line for $\log_{10}\eta_0 = 5$. Note that the lines with a constant value of η are self-similar.

$$T_0(\eta) = T_0\left[1 + \left(\frac{\eta}{\eta_{T,\sigma=0}}\right)^{-1/\alpha}\right], \qquad (3)$$

where $\eta_{T,\sigma=0}$ is the value of η_T extrapolated to $\sigma = 0$. Also from the data in Fig. 3 extrapolated to $T = 0$ we obtain,

$$\sigma_0(\eta) = \sigma_0\left[1 + \left(\frac{\eta}{\eta_{\sigma,T=0}}\right)^{-1/\gamma}\right], \qquad (4)$$

where $\gamma = 0.93$, and $\eta_{\sigma,T=0}$ is a fitting parameter for the stress extrapolated to $T = 0$. The viscosity data collapse into one master curve when we plot $T/T_0(\eta)$ against $\sigma/\sigma_0(\eta)$, as shown in Fig. 4, where the dashed line represents Eq. (2).

Note that in Fig. 2 the data above and below T_0 converge to two different curves, as in general critical phenomena. However, in Fig. 4 the data for the whole T-σ field collapse into a single curve. In Fig. 3 T_0 is not a singular point, and for $\sigma > 0$ the curves for constant viscosity are smooth through T_0. Thus, although these two scaling relationships are mathematically related, they represent very different points of view. The first scaling by Eq. (1) describes the behavior of the stress-viscosity relation as it crosses over from the mean-field regime to the fluctuation dominated regime around T_0. The second scaling by Eq. (2) does not assume T_0 to be a singular point, but regards the T-σ line defined by $\eta \to \infty$ as a critical line, and describes the viscosity behavior as the system approaches in the T-σ field toward this line. This critical line corresponds to a cut in the jamming phase diagram [13,14] showing the phase boundary at a constant density. A cut at a constant tem-
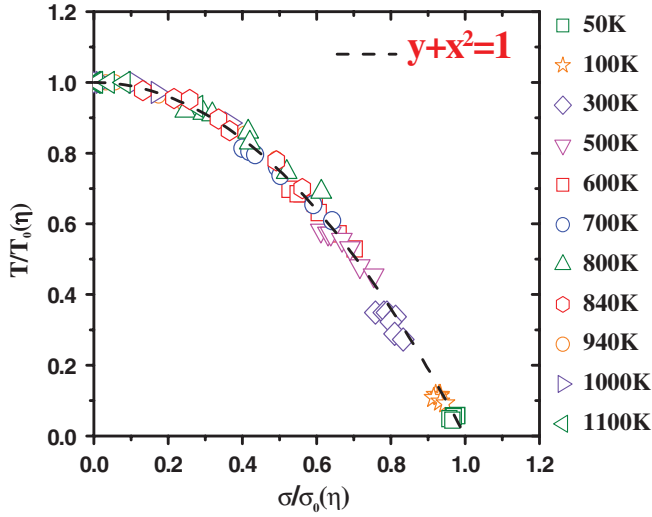
FIG. 4 (color).   Plot of $\frac{T}{T_0(\eta)}$ vs $\frac{\sigma}{\sigma_0(\eta)}$ for different temperatures T and stress σ with various shear strain rates $\dot{\gamma}$, demonstrating the scaling relationship, Eq. (2).

perature above T_0 in Fig. 3 shows that stress reduces viscosity, describing the phenomenon of shear thinning [10,11].

Reference [7], as well as other references (for example Ref. [15]), interpreted the effect of the external stress on the flow in terms of the effective temperature [16]. Equation (2), however, suggests an interesting alternative interpretation of the effects of the external stress. For instance, we may assume a simple activation model of viscosity [17], $\eta = \eta_0 \exp(E_a/kT)$, where E_a is the apparent activation energy. Then viscosity is constant at different temperatures when $E_a(T, \sigma)/kT \equiv E_0/kT_0(\eta) = \text{const}$. On the other hand by multiplying Eq. (2) through E_0/kT we obtain

$$\frac{E_0 - E_0(\frac{\sigma}{\sigma_0(\eta)})^2}{kT} = \frac{E_0}{kT_0(\eta)} = \text{const}, \qquad (5)$$

which can be rearranged as

$$E_a(T, \sigma) = \frac{T}{T_0(\eta)}E_0 = E_0 - E_0\left(\frac{\sigma}{\sigma_0(\eta)}\right)^2. \qquad (6)$$

Furthermore, by rewriting $E_0/\sigma_0^2(\eta)$ as $V_{\text{eff}}/2G$, where G is the instantaneous shear elastic constant and V_{eff} is the effective volume of the flow unit, we obtain

$$E_a(T, \sigma) = E_0 - \frac{V_{\text{eff}}}{2G}\sigma^2. \qquad (7)$$

Equation (7) suggests that the effect of the external stress is to reduce the apparent activation energy barrier in the local energy landscape through the self-energy of the external stress and to reduce viscosity, thereby changing a glass into a liquid. The evaluation of V_{eff} will be discussed elsewhere.

The result shown here suggests that mechanical failure is indeed the glass transition induced by applied stress, at a temperature lower than the glass transition temperature. This observation allows us to connect the microscopic mechanism of deformation in the steady-state flow with that of the glass transition. If we consider locally fluctuating stress fields, such as the atomic level stresses [18], and assume that there is a threshold for local atomic level strain due to topological instability [19] which defines the unstable (liquidlike) atomic sites and stable (solidlike) atomic sites. The density of the liquidlike atomic sites decreases with decreasing temperature, and when it reaches the percolation limit glass transition would occur [20]. Recently the glass transition temperature calculated with this scenario was shown to agree well with experimental values [21]. The same logic could be applied here in the following hypothesis: the applied stress biases the stress distribution and thus the local energy landscape, and makes some atoms unstable, or liquidlike. If the density of the liquidlike atoms exceeds the percolation limit the system flows like a liquid, thus inducing the glass-to-liquid transition.

The process of establishing the state of steady-state flow through yielding is spatially inhomogeneous, which requires mesoscopic description [22–24]. Also, because the MD simulation is classical, quantum corrections are necessary at low temperatures. Specifically $\sigma_0(\eta)$ will be independent of temperature at low temperatures. Consequently the curve of constant η will be vertical at $T = 0$, making the shape of the constant η curve closer to the schematics given in Ref. [13].

In conclusion, through simulation on a metallic glass system with a set of realistic potentials we have quantified the intimate coupling of temperature and shear stress in inducing liquidlike flow in glasses. The observed effect is closely related to the jamming phase diagram. A new temperature-stress scaling for constant viscosity was found, providing new insights about the mechanism of atomic flow under stress in the glassy and liquid state.

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