

Supporting Information

Viscoelasticity and Relaxations in $\text{Cu}_{64}\text{Zr}_{36}$ Bulk Metallic Glass and Nanoglass

Param Punj Singh[†]; Dhyanesh Baskaran[†], Omar Adjaoud[‡], Karsten Albe[‡], and
Raghavan Ranganathan^{†*}

[†]Department of Materials Engineering, Indian Institute of Technology Gandhinagar,
India-382355

[‡]Institute of Materials Science, Technische Universität Darmstadt, Otto-Berndt-Str.3,
64287, Darmstadt, Germany *Corresponding author. E-mail: rraghav@iitgn.ac.in

Table of contents

1. **Figure S1:** Evolution of potential energy in $\text{Cu}_{64}\text{Zr}_{36}$ MG and NG over 20 cycles of oscillatory.
2. **Figure S2:** String-like motion in NG within the temperature range of 400–480 K.
3. **Figure S3:** Visualization of atomic displacements in MG and NG at varying temperatures and their evolution during shear cycles.
4. **Figure S4:** Contoured plots of non-affine atomic displacements in MG and NG at 640 K, 680 K, and 720 K.

Figure S1 illustrates the evolution of potential energy as a function of oscillatory shear simulation time. The simulations were performed with an oscillation period of 0.1 ns, corresponding to 25 shear cycles. The per-atom potential energy of both systems was computed using the `pe/atom` command in LAMMPS and recorded at intervals of 2 ps.

For temperatures in the range of 640–720 K, corresponding to the β -relaxation regime, NG exhibits a higher potential energy compared to MG. However, during oscillatory shear, the potential energy of NG decreases at all temperatures, indicating enhanced relaxation toward a more stable, lower-energy state, a behavior not observed in MG.

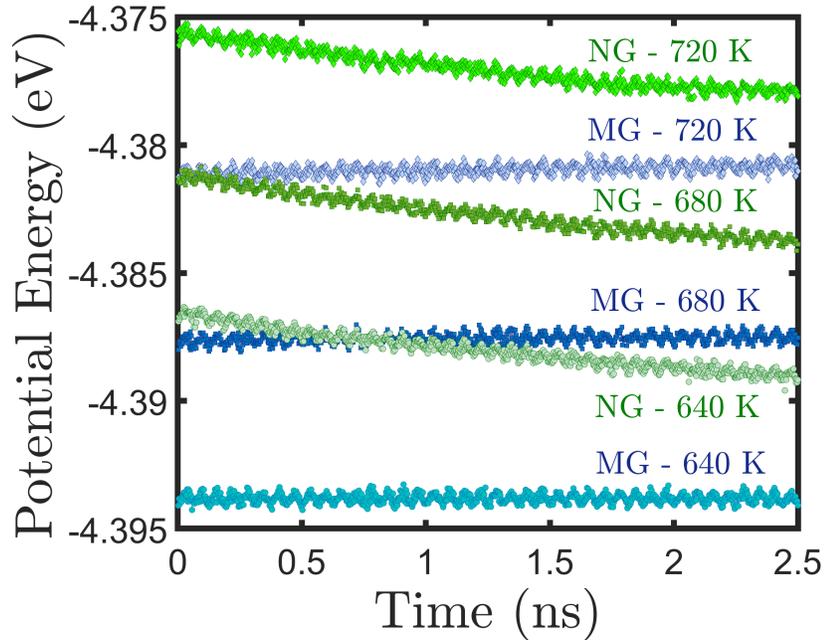


Figure S1: Evolution of potential energy in MG and NG over 20 cycles of oscillatory deformation at varying temperatures.

The G'' of NG exhibits distinct peaks at 400 K and 480 K, and the following analysis explains the mechanisms underlying these relaxations. As shown in Figure S2a, string-like cooperative motions are observed in NG even at moderately low temperatures (400–480 K), whereas such behavior is absent in MG. Furthermore, when comparing across these temperatures, it is evident that the extent of string-like motion is reduced at 440 K relative to 400 K and 480 K. Figure S2(b,c) presents the histograms of N_{string} for oscillation periods of 0.1 ns and 1 ns, respectively. The results show that small string clusters ($N_{string} = 2$) are more prevalent at 400 K, decrease at 440 K, and increase again at 480 K. This analysis is consistent with the relaxation behavior of NG within this temperature range, giving rise to the small peaks observed in G'' .

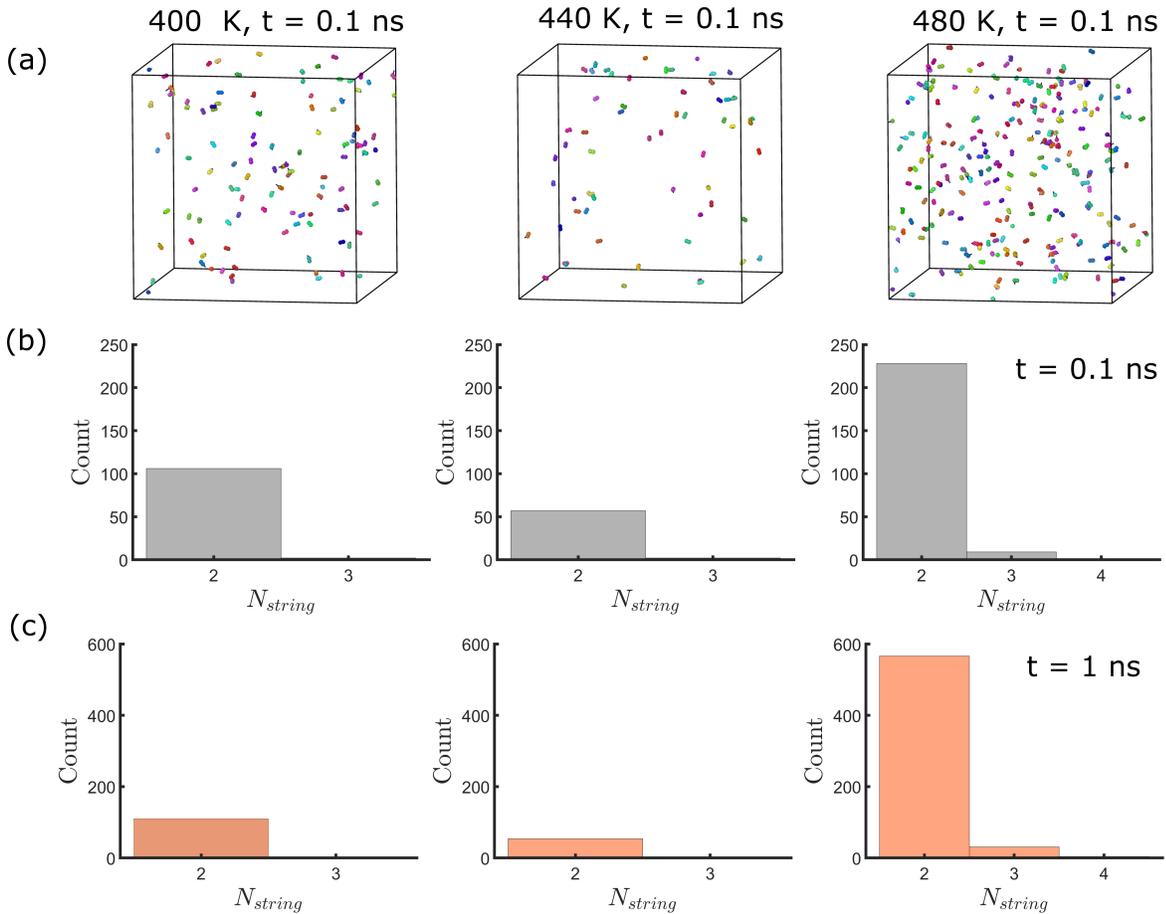


Figure S2: (a) String-like motion in NG at 400 K, 440 K, and 480 K, and (b and c) histogram of string count for $t = 0.1$ and 1 ns, respectively.

The 2D atomic displacement maps of MG and NG at varying temperatures are shown in Figure S3(a,b), respectively. A cutoff of 2.75 \AA , corresponding to the first nearest-neighbor distance, was applied. Displacement vectors above this threshold are shown in red, while those below are shown in blue. Comparison of the maps reveals that NG contains a larger number of atoms with displacements exceeding the nearest-neighbor distance. Notably, in NG, such nearest-neighbor jumps are predominantly initiated at the interfacial regions at lower temperatures. Figure S3c shows the displacement evolution in NG during an oscillating shear loading. Interfacial atoms are found to be more prone to jump easily than the atoms present in the grain interiors.

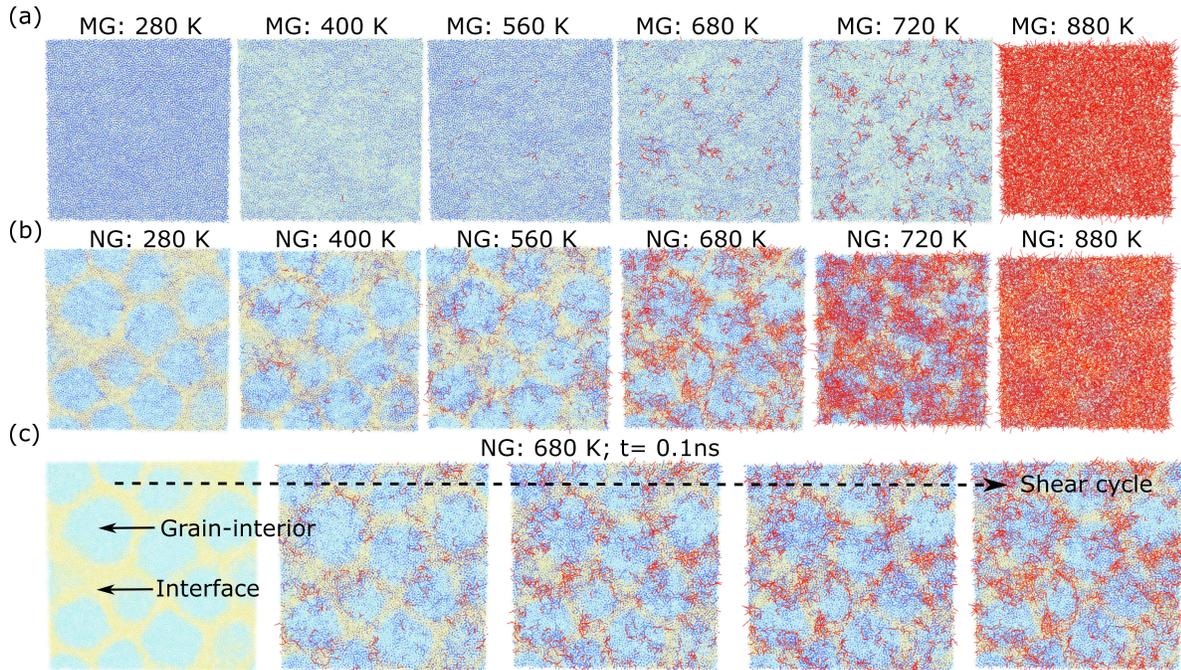


Figure S3: (a,b) Atomic displacement vectors (red lines) in MG and NG at varying temperatures. (c) Atomic displacements in NG for an oscillation period of 0.1 ns at 680 K.

The D_{min}^2 parameter is employed to identify atoms that undergo shear transformations by tracking sudden changes in its value. The calculation begins with determining the locally affine transformation matrix, J_i , which best maps the reference bond vectors d_{ji}^0 to the current bond vectors d_{ji} for all nearest neighbors N . The locally non-affine displacement of the central atom i relative to its nearest neighbors is then defined as:

$$D_{min}^2 = \frac{1}{N} \left(\vec{r}_j(t) - \vec{r}_i(t) - J_i [\vec{r}_j(t - \Delta t) - \vec{r}_i(t - \Delta t)] \right)^2 \quad (S1)$$

Figure S4(a,b) exhibits the contoured plot of D_{min}^2 in MG and NG after 20 shear cycles at temperatures 640 K, 680 K, and 720 K. NG exhibits a larger shear-prone zone, indicated by the cluster of atoms in yellow, compared to MG across all temperatures. This suggests that NG experiences higher energy dissipation due to shear deformation.

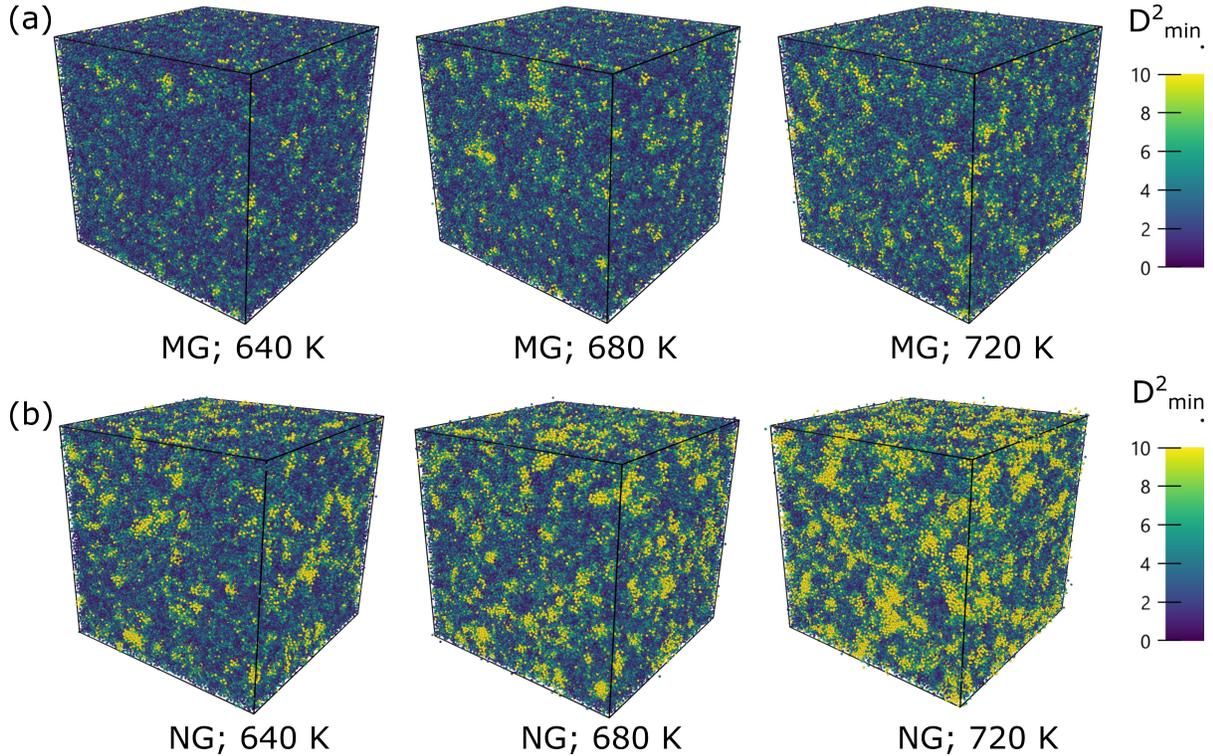


Figure S4: Contoured map of non-affine displacement D_{min}^2 of MG and NG at 640 K, 680 K, and 720 K.