Electronic supplementary information

Effects of atomic interaction stiffness on low-temperature relaxation of amorphous solids

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Section S1. Simulation details

All simulations were performed using LAMMPS package.[1] The simulation boxes are cuboid with the ratio of length in x, y, z direction maintaining 5:5:1 throughout all the simulation processes. Periodic boundary conditions are applied in all directions. All samples are first melted and equilibrated at T=5 in NPT (constant number, pressure, temperature) ensemble, followed by a cooling to the designated temperature with a constant cooling rate of 10^5, and then annealed at the temperature for 5×10^5 simulation steps. The pressure is set to be 1 during sample preparations. After sample preparation, MD-DMAs were performed in NVT (constant number, volume, temperature) ensemble, during which a sinusoidal shear strain $\varepsilon(t) = \varepsilon_A \sin(2\pi t/T_\omega)$ is applied along *xy* direction. The period T_ω is selected to be 10000 simulation steps, and the maximum strain ε_A=0.02, 40 cycles were performed at each test temperature. For each system at each temperature, the atoms were assigned 10 different initial velocities according to Gaussian distribution, and 10 independent MD-DMA simulations were performed to improve the statistics. The phase difference between the applied strain and the detected stress gives information on the internal friction of the system.

Section S2. Voronoi analysis

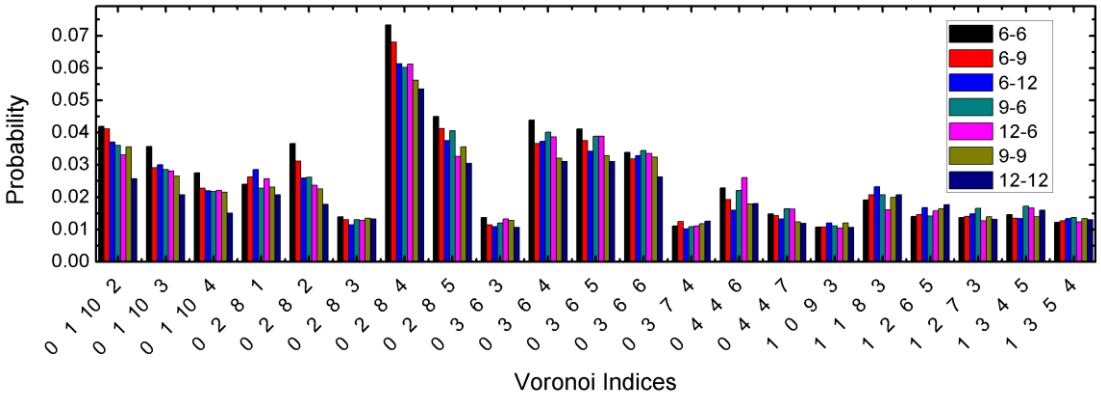

Fig. S1. Voronoi analysis for model 6-6, 6-9, 6-12, 9-6, 12-6, 9-9, 12-12. Only Voronoi polyhedra with fraction larger than 1% in all models are shown. All models share similar atomic motifs.

Section S3. Shear modulus *E*, phase difference δ, storage modulus *E'* and loss modulus *E"*

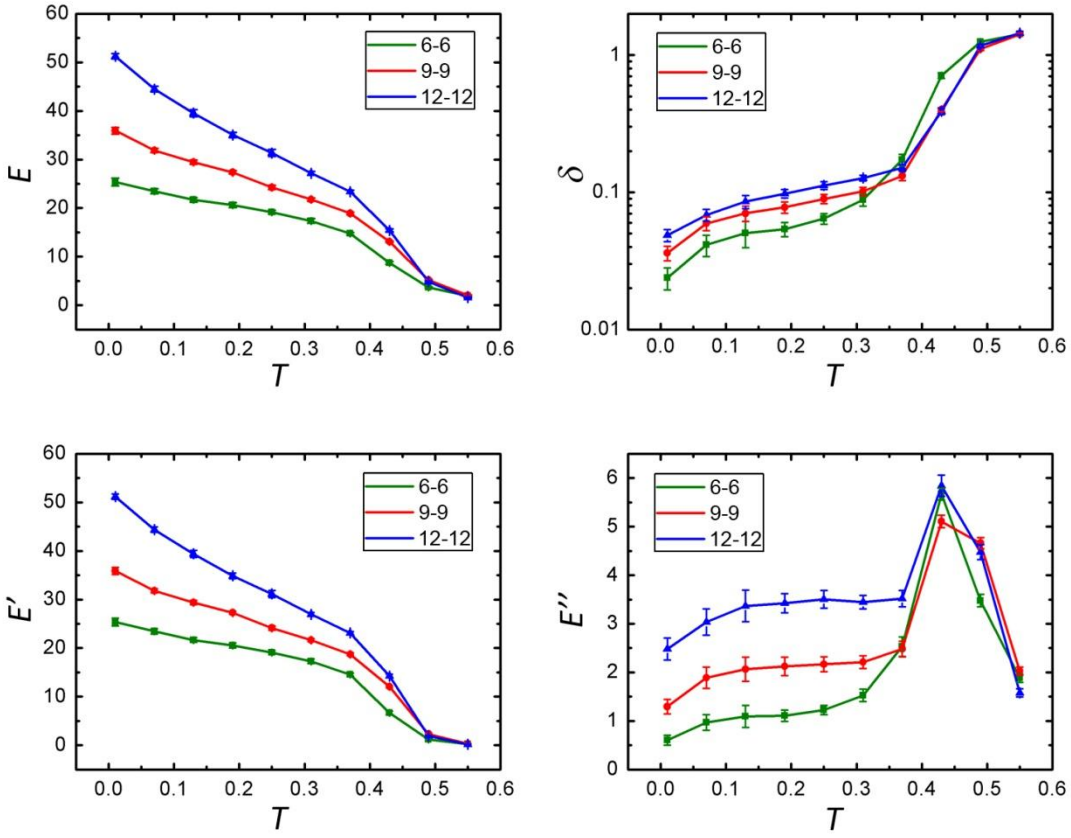

Fig. S2. Shear modulus *E*, phase difference δ, storage modulus *E'* and loss modulus *E"* for model 6-6, 9-9, 12-12. In this case, the loss modulus *E"* is not able to reflect the relaxation dynamics of these models, due to the increase of shear modulus *E* as the atomic interaction stiffness increases. The phase difference δ, as an indicator of the relation between *E'* and *E"*, reveals the internal friction of these models.

Section S4. Calculation of non-affine displacement D^2

The non-affine displacement of a particle (denoted as 0) during a time interval of T_ω is defined as

$$D^2\left(t,T_\omega\right)=\sum_n\sum_i\left(\mathbf{r}_n^i\left(t+T_\omega\right)-\mathbf{r}_0^i\left(t+T_\omega\right)-\sum_j\left(\delta_{ij}+\varepsilon_{ij}\right)\times\left[\mathbf{r}_n^j\left(t\right)-\mathbf{r}_0^j\left(t\right)\right]\right)^2,$$

$$X_{ij}=\sum_n\left[\mathbf{r}_n^i\left(t+T_\omega\right)-\mathbf{r}_0^i\left(t+T_\omega\right)\right]\times\left[\mathbf{r}_n^j\left(t\right)-\mathbf{r}_0^j\left(t\right)\right],$$

$$Y_{ij}=\sum_n\left[\mathbf{r}_n^i\left(t\right)-\mathbf{r}_0^i\left(t\right)\right]\times\left[\mathbf{r}_n^j\left(t\right)-\mathbf{r}_0^j\left(t\right)\right],$$

$$\varepsilon_{ij}=\sum_k X_{ik}Y_{jk}^{-1}-\delta_{ij},$$

where indices *i, j* denotes the spatial coordinates so that $\mathbf{r}_n^i\left(t\right)$ indicates the *i*th component of the *n*th particle at time *t*, and the index *n* runs over the neighbor particles within the cutoff distance of atomic interaction [2].

Section S5. Experimental data for molar volume and bulk modulus of 62 metallic glasses

Table S1. Experimental data for molar volume and bulk modulus of 62 metallic glasses.

Compositions	Molar volume (cm^3/mol)	Bulk modulus (GPa)	References
$(Ce_{20}La_{80})_{68}Al_{10}Cu_{20}Co_2$	16.78	32.9	[3]
$(Ce_{80}La_{20})_{68}Al_{10}Cu_{20}Co_2$	16.69	31.8	[3]
$Ce_{68}Al_{10}Cu_{20}Co_2$	16.57	30.3	[4]
$Ce_{68}Al_{10}Cu_{20}Fe_2$	16.59	31.4	[5, 6]
$Ce_{68}Al_{10}Cu_{20}Nb_2$	16.70	30.1	[4, 6]
$Ce_{68}Al_{10}Cu_{20}Ni_2$	16.57	31.8	[4, 6]
$Ce_{70}Al_{10}Cu_{10}Ni_{10}$	16.94	27.0	[7, 8]
$Ce_{70}Al_{10}Cu_{20}$	16.94	29.2	[4, 6]
$Dy_{55}Al_{25}Co_{20}$	13.85	52.2	[9-11]
$Er_{55}Al_{25}Co_{20}$	13.87	60.7	[10, 11]
$Gd_{55}Al_{25}Co_{20}$	14.15	60.1	[3, 11]
$Ho_{55}Al_{25}Co_{20}$	13.56	58.8	[10, 11]
$La_{55}Al_{25}Cu_{10}Ni_5Co_5$	15.90	44.2	[7, 8]
$La_{55}Al_{25}Co_{20}$	15.08	39.3	[10, 11]
$Lu_{39}Y_{16}Al_{25}Co_{20}$	13.30	71.3	[3]
$Lu_{45}Y_{10}Al_{25}Co_{20}$	13.25	70.2	[3]
$Lu_{55}Al_{25}Co_{20}$	13.20	69.2	[3]
$Nd_{60}Al_{10}Fe_{20}Co_{10}$	15.18	46.5	[7, 12-14]
$Nd_{60}Al_{10}Ni_{10}Cu_{20}$	16.12	42.8	[3]
$Tb_{55}Al_{25}Co_{20}$	14.28	50.2	[10, 11]
$Tm_{39}Y_{16}Al_{25}Co_{20}$	13.51	66.1	[3]
$Tm_{55}Al_{25}Co_{20}$	13.47	62.0	[3]
$Yb_{62.5}Zn_{15}Mg_{17.5}Cu_5$	19.24	19.8	[3]
$Pr_{55}Al_{25}Co_{20}$	14.86	43.5	[10, 11]
$Pr_{60}Al_{10}Ni_{10}Cu_{20}$	15.33	45.2	[7, 15]
$Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$	10.00	114.7	[13, 16]
$Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$	10.17	114.1	[7, 8]
$Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5}$	10.21	111.9	[7, 9, 13]
$Zr_{48}Nb_8Cu_{12}Fe_8Be_{24}$	10.18	113.6	[7, 17]
$Zr_{48}Nb_8Ni_{12}Cu_{14}Be_{18}$	10.26	118.3	[7, 8]
$(Zr_{59}Ti_6Cu_{22}Ni_{13})_{85.7}Al_{14.3}$	10.74	112.6	[17]
$Zr_{50}Cu_{30}Ni_{10}Al_{10}$	10.66	124.1	[15]
$Zr_{52.5}Al_{10}Ti_5Cu_{17.9}Ni_{14.6}$	11.02	120.7	[10]
$Zr_{55}Al_{10}Ni_5Cu_{30}$	10.97	113.1	[18]
$Zr_{55}Al_{19}Co_{19}Cu_7$	11.44	114.9	[7, 8]
$Zr_{55}Ti_5Cu_{20}Ni_{10}Al_{10}$	11.15	118.0	[7, 8]
$Zr_{57}Nb_5Cu_{15.4}Ni_{12.6}Al_{10}$	11.44	107.7	[16]
$Zr_{57}Ti_5Cu_{20}Ni_8Al_{10}$	11.40	99.2	[10, 13, 16]

$Zr_{57.5}Nb_5Cu_{15.5}Ni_{12}Al_{10}$	11.80	117.6	[7]
$Zr_{61.88}Cu_{18}Ni_{10.12}Al_{10}$	11.51	108.3	[19]
$Zr_{64.13}Cu_{15.75}Ni_{10.12}Al_{10}$	11.68	106.6	[19]
$Zr_{65}Al_{10}Ni_{10}Cu_{15}$	11.65	106.7	[13]
$Cu_{45}Zr_{45}Al_7Gd_3$	10.71	117.1	[5, 9]
$Cu_{46}Zr_{42}Al_7Y_5$	10.20	104.1	[7, 8]
$Cu_{46}Zr_{54}$	10.30	128.5	[7, 8]
$Cu_{50}Zr_{50}$	10.45	101.2	[5, 10, 12]
$Cu_{50}Hf_{43}Al_7$	10.04	132.8	[7, 8]
$Cu_{57.5}Hf_{27.5}Ti_{15}$	9.36	117.5	[7, 8]
$Cu_{60}Hf_{10}Zr_{20}Ti_{10}$	9.52	128.2	[4, 13]
$Pd_{35}Cu_{30}Ni_{10}Fe_5P_{20}$	7.75	173.5	[7]
$Pd_{39}Ni_{10}Cu_{30}P_{21}$	7.96	159.1	[7, 17]
$Pd_{40}Ni_{10}Cu_{30}P_{20}$	8.03	159.1	[7, 13]
$Pd_{40}Ni_{40}P_{20}$	7.68	185.0	[7, 13, 20]
$Pd_{40}Cu_{40}P_{20}$	8.00	158.0	[7, 8]
$Pd_{48}Ni_{32}P_{20}$	7.73	176.7	[7, 20, 21]
$Pd_{56}Fe_{24}P_{20}$	8.00	161.2	[7, 21]
$Pd_{60}Cu_{20}P_{20}$	7.81	167.0	[7, 8]
$Pd_{60}Fe_{20}P_{20}$	8.15	164.5	[7, 21]
$Pd_{64}Fe_{16}P_{20}$	8.32	161.9	[7, 21]
$Pd_{64}Ni_{16}P_{20}$	8.29	166.0	[7, 8, 21]
$Pd_{68}Fe_{12}P_{20}$	8.44	158.1	[7, 21]
$Pd_{77.5}Cu_6Si_{16.5}$	8.74	174.7	[7, 13]

The rear earth, Zr, Cu and Pd based metallic glasses are studied for their adequate data for the analysis of atomic interaction stiffness. Other series of metallic glasses do not shown clear relation between bulk modulus and molar volume, due to the deficiency of data[22].

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