Elastic Properties and Glass Forming Ability of the $Zr_{50}Cu_{40}Ag_{10}$ Metallic Alloy

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The elastic properties of the $Zr_{50}Cu_{40}Ag_{10}$ metallic alloy, such as the bulk modulus B, the shear modulus G, the Young's modulus E and the Poisson's ratio σ , are investigated by molecular dynamics simulation in the temperature range T = 250 - 2000 K and at an external pressure of p = 1.0 bar. It is shown that the liquid-glass transition is accompanied by a considerable increase in the shear modulus G and the Young's modulus E (by more than 50%). The temperature dependence of the Poisson's ratio exhibits a sharp fall from typical values for metals of approximately 0.32 - 0.33 to low values (close to zero), which are characteristic for brittle bulk metallic glasses. Non-monotonic temperature dependence of the longitudinal and transverse sound velocity near the liquid-glass transition is also observed. The glass forming ability of the alloy is evaluated in terms of the fragility index m. As found, its value is $m \approx 64$ for the $Zr_{50}Cu_{40}Ag_{10}$ metallic glasses, that is in a good agreement with the experimental data for the Zr-Cu-based metallic glasses.

I. INTRODUCTION

Interest to the amorphous metallic alloys is provoked by the obvious advantages of their potential applications in various fields of techniques, engineering and materials science. Amorphous alloys or metallic glasses are a new class of materials with unique physical and mechanical properties, which differ from the properties of their crystalline analogs [1, 2]. So, for example, non-crystalline metallic alloys are characterized by high strength and elasticity, as well as good ductility under strong deformation effects. As a rule, metallic glasses are multicomponent systems with high glass forming ability, the disordered phase of which can be formed by means of cooling the equilibrium melt with the cooling rates of $\gamma > 10^4$ K/s [3]. Despite a significant number of experimental and numerical studies, there is still no clear understanding of the mechanisms of formation of structural heterogeneities and their influence on the elastic and mechanical properties of metallic glasses.

The aim of this work was a thorough study of the elastic properties of the $Zr_{50}Cu_{40}Ag_{10}$ -alloy in a wide temperature range, which includes "liquid-glass" transition.

II. SIMULATION DETAILS

Molecular dynamics (MD) simulations of the $Zr_{50}Cu_{40}Ag_{10}$ melt were carried out with an isothermalisobaric (*NPT-*) ensemble in the temperature range T = 250 - 2000 K at an external pressure of p = 1.0 bar. To stabilize the temperature and the pressure of the system, the Nosé-Hoover thermostat with a relaxation parameter of 0.1 ps and the barostat with damping parameter of 1.0 ps were applied. The system was consisted of N = 32000 atoms enclosed in a cubic cell with the periodic boundary conditions. The interactions between atoms are described by means of the EAM model potential [4]. Initial configurations were equilibrated $Zr_{50}Cu_{40}Ag_{10}$ melt at T = 2000 K. Then, the system was cooled down to T = 250 K with the cooling rate 1.0 K/ps [5]. The integration of the equations of motion of atoms was carried out using the velocity Verlet algorithm with a time step of $\tau = 1.0$ fs. To bring the system to a state of thermodynamic equilibrium, we performed 10^6 time steps and then 10^7 steps to calculate the mechanical properties and characteristics. All simulations were performed in the LAMMPS simulation package [6].

III. RESULTS AND DISCUSSIONS

Fig. 1 (a-c) shows the partial components of the radial distribution function for the $\operatorname{Zr}_{50}\operatorname{Cu}_{40}\operatorname{Ag}_{10}$ melt at various temperatures. As seen from the figure, all partial components $g_{\alpha,\beta}(r)$ (where $\alpha, \beta \in \{\operatorname{Zr}, \operatorname{Cu}, \operatorname{Ag}\}$) with decreasing temperature manifest the structural transformations in the system due to the transition from the equilibrium melt to the amorphous glassy state [7]. The critical temperature T_c associated with liquid-glass transition was determined using the translational order parameter [2]. The temperature dependence of this parameter in the semilogarithmic coordinates has two segments with an intersection point at $T_c \approx 750$ K (see Fig. 1d).

The elastic properties of the $Zr_{50}Cu_{40}Ag_{10}$ metal melt were studied by means of evaluation of the elastic (bulk and shear) moduli [8, 9]. For the case of the *NPT*ensemble, the isothermal bulk modulus is related to the mean-square fluctuations σ_V^2 of the volume V of the simulation cell by the expression:

$$B = \frac{k_B T V}{\sigma_V^2}$$



Figure 1: Partial components of the radial distribution function and the translational order parameter for $Zr_{50}Cu_{40}Ag_{10}$ melt at different temperatures.

The shear modulus was calculated by the formula

$$G = \frac{V}{k_B T} \left\langle \left| p_{xy}(0) \right|^2 \right\rangle,$$

where angle brackets mean averaging over time samples and $p_{xy}(0)$ are the non-diagonal components of the pressure tensor.

To assess the deformation properties of materials, it is necessary to know the equilibrium elastic modulus (the so-called Young's modulus) and the transverse strain coefficient (the so-called Poisson's ratio). The Young's modulus E and the Poisson's ratio σ are related with the elastic moduli as follows

$$E = \frac{9BG}{3B+G}, \quad \sigma = \frac{3B-2G}{6B+2G}$$



Figure 2: Temperature dependences of the bulk modulus B, the shear modulus G, the Poisson's ratio σ and the Young's modulus E for the $Zr_{50}Cu_{40}Ag_{10}$ metallic alloy.

The temperature dependences of the bulk modulus B, the shear modulus G, the Poisson's ratio σ and the

Young's modulus E for the $Zr_{50}Cu_{40}Ag_{10}$ metallic alloy are plotted in Fig. 2. As seen from this figure, the temperature dependences of the shear modulus G, the Poisson's ratio σ and the Young's modulus E are characterized by a sharp change near the liquid-glass transition. At the same time, the bulk modulus grows monotonically with decreasing temperature. The importance of the Poisson's ratio in the study of glasses was pointed out by Novikov and Sokolov [10]. They found that the Poisson's ratio is related to the fragility index m of the glass forming liquid, which can be used to account for the glass forming ability (GFA) properties of a melt. So, for example, the Poisson's ratio for brittle bulk metallic glasses demonstrates a low value. It can be seen from the Fig. 2 that glassy $Zr_{50}Cu_{40}Ag_{10}$ -alloy at the temperature T = 250 K is characterized by a Poisson's ratio close to zero. The Poisson's ratio takes values close to zero showing very little lateral expansion when compressed. The fragility index m was calculated by the formula [10]

$$m = 17 + 29(B/G - 1),$$

where 17 is the lowest value expected for m and B/Gin glasses is not expected to be lower than 1 [11]. For the $Zr_{50}Cu_{40}Ag_{10}$ -system, the calculated value of the fragility index is $m = 64\pm 4$, which is in a good agreement with the experimental data for the Zr-Cu-based metallic glasses (see Table I).

Table I: Experimental data on the fragility index for amorphous Zr-Cu-based metallic alloys [12, 13].

Alloy composition	Fragility index, m
$\mathrm{Zr}_{50}\mathrm{Cu}_{50}$	76 ± 10
$Zr_{44}Cu_{50}Ti_6$	48 ± 4
$Zr_{46}Cu_{46}Al_8$	63 ± 7
$Zr_{47}Cu_{47}Al_6$	53 ± 5

The elastic characteristics and the velocities of the longitudinal and transverse ultrasonic waves for an isotropic medium are related by the expressions [14–16]

$$\vartheta_L = \sqrt{\frac{B+4/3G}{\rho}}, \quad \vartheta_T = \sqrt{\frac{G}{\rho}}$$

where ρ is the mass density of the system. Fig. 3 depicts the obtained temperature dependences of the speed of sound for $Zr_{50}Cu_{40}Ag_{10}$ metallic alloy. As seen from the figure, that the temperature dependences of the longitudinal and transverse sound velocities for the equilibrium melt phase are monotonic. At the same time, near the liquid-glass transition, the changes in the temperature dependencies of both the rates ϑ_L and ϑ_T are observed.

IV. SUMMARY

In the present work, using molecular dynamics simulations, the elastic properties and glass forming ability of



the ternary metallic alloy are investigated. The temperature dependences of bulk modulus B, the shear modulus G, the Young's modulus E and the Poisson's ratio σ , for the $Zr_{50}Cu_{40}Ag_{10}$ metallic alloy were obtained for the first time. It is shown that the liquid-glass transition is accompanied by a considerable increase in the shear modulus G and the Young's modulus E (by more than 50%). Temperature dependence of the Poisson's ratio for the $Zr_{50}Cu_{40}Ag_{10}$ -system exhibits a sharp fall from typical values for metals of approximately 0.32 - 0.33to low values (close to zero), which are characteristic for brittle bulk metallic glasses. It was also found that the temperature dependence of the sound velocities undergo significant changes near the liquid-glass transition. The calculated value of fragility index $m \approx 64$ for the $Zr_{50}Cu_{40}Ag_{10}$ -system is in a good agreement with the experimental data for the Zr-Cu-based metallic glasses.

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Figure 3: Temperature dependences of the longitudinal and transverse sound velocities for $Zr_{50}Cu_{40}Ag_{10}$ metallic alloy.

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