

Local Structural Order and Single-Particle Dynamics in Metallic Glass

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Abstract—The atomic dynamics of the amorphous metallic alloy $\text{Al}_{50}\text{Cu}_{50}$ is investigated in this work. The critical glass-forming temperature is defined from the behavior change of the Wendt–Abraham parameter and the pair-correlation entropy, and is $T_c \approx 500$ K. It is shown that the power-fractional memory function allows us to describe in the framework of the mode-coupling theory the time dependences of the incoherent scattering functions obtained from the atomic dynamic simulations for the wide region of temperatures ($T = 100$ – 2000 K).

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INTRODUCTION

Amorphous metallic alloys (or metallic glasses) were first obtained in 1950s and continue to attract wide interest due to their unique complex of physico-mechanical properties, which differs from their crystalline counterparts [1–6]. An important feature of metallic glasses is that their atomic ensemble is characterized by topological and/or compositional short-order range, kept within five to six interatomic distances (i.e., within distances of approximately several nanometers); such systems do not possess the long-order range typical of crystals. The atoms are forming disordered local condensations and rarefactions allowing also to consider metallic glasses as non-uniform and disordered nanostructures [7].

Metals are usually crystallized rapidly at crystallization temperature T_m as a result of melt cooling. Sufficiently fast melt cooling (as a rule at cooling rates higher than 10^5 K/s) allows us to avoid crystallization and obtain amorphous metallic alloys characterized by disordered arrangement of the atoms [3]. Metallic glasses are very hard and plastic. In spite of the fact that the density of amorphous alloys being 1–2% less than that of their crystalline analogues, the strength of metallic glasses is 5–10 times greater [4–6]. High strength combined with plasticity, increased fatigue strength, and radiation damage durability distinguish metallic glasses as a promising class of materials with a wide range of applications. Despite the wide practical applications of metallic glasses, there is still no comprehensive understanding of the physical mechanisms occurring in these systems. This is partially due to the absence of the unified theory describing the dynamic processes in glasses (metallic, polymeric, spin) and of explanations for the appearance of the glass phase itself and its typical anomalous features in transport properties (anomalous subdiffusion),

vibration properties (the bozon peak), and relaxation dynamics (dynamic heterogeneity) [8].

This work presents the results from investigating the structural features and dynamic properties of metallic glass $\text{Al}_{50}\text{Cu}_{50}$ obtained by the atomic dynamics simulation. The critical glass transition temperature is defined on the basis of obtained temperature dependences of the Wendt–Abraham parameter and two-particle correlation entropy. We describe the space–time dependences of the partial incoherent scattering function in metallic glass $\text{Al}_{50}\text{Cu}_{50}$ at temperature $T=100$ K within the framework of the mode-coupling theory.

DETAILS OF ATOMIC DYNAMICS SIMULATION

$\text{Al}_{50}\text{Cu}_{50}$ alloy was selected as the researched system, and its atomic interaction was modeled by the multipartial *EAM*-potentials, which were suggested in the work [9]. The total interaction energy of the particles is denoted as:

$$E_{\text{tot}} = \sum_i U(\rho_i) + \frac{1}{2} \sum_{i \neq j} \phi_{ij}(r_{ij}), \quad (1)$$

where $\phi_{ij}(r_{ij})$ is the pair interaction potential between particles i and j , located at distance r_{ij} from one to another ($i, j \in \{\text{Al}, \text{Cu}\}$), $U(\rho_i)$ is the many-particle interaction energy, and ρ_i is the electron density of i -th atom. The forms of functions $U(\rho_i)$ and ρ_i in expression (1) were obtained in the work [9].

The modeled system represented initially a cubic cell with the periodic boundary conditions. The size of the system was $N = 4000$ particles, with the numerical density of $n = N/V = 0.0701 \text{ \AA}^{-3}$, where V is the cell volume. The system with initial temperature $T = 2000$ K

was reduced to the amorphous state at temperature $T = 100$ K by rapid cooling at rate $dT/dt = 10^{12}$ K/s. The numeric integration of particle motion equations was performed with the velocity Verlet algorithm with the time step 10^{-15} s [10].

RADIAL DISTRIBUTION FUNCTION OF PARTICLES

One of the quantities characterizing the structure of the system under study is the radial distribution function of particles $g(r)$. This magnitude is associated with the static structural factor $S(k)$, measured by means of integral Fourier transformation in experiments on scattering neutrons and X-rays. For dense disordered single-component systems, function $g(r)$ is characterized by a prominent first peak at values r comparable to particle sizes, and by rapid attenuating oscillations. Oscillation maxima correspond to the coordination spheres around a selected atom. During structural ordering, strongly pronounced peaks that become narrow as the degree of order increases appear instead of oscillation.

In the case of binary systems, we can introduce partial radial distribution functions of particles

$$g_{\alpha\beta}(r) = \frac{L^3}{N_\alpha N_\beta} \left\langle \sum_{j=1}^{N_\alpha} \frac{n_{j\beta}(r)}{4\pi r^2 \Delta r} \right\rangle, \quad \alpha, \beta \in \{\text{Al}, \text{Cu}\}, \quad (2)$$

calculated directly from the modeling data of atomic dynamics, or from Monte Carlo modeling data [11]. Here, $g_{\alpha\beta}(r)$ is the probability of an atom being located in the range r to $r + \Delta r$, L is the edge length of the modeled cell, and N_α and N_β are the numbers of α - and β -type atoms, respectively. Magnitude $n_{j\beta}(r)$ defines the number of β -particles in a spherical layer with the thickness Δr at the distance r from the j -th particle. Figure 1 shows partial radial distribution functions of particles $g_{\alpha\beta}(r)$, calculated by the atomic dynamics simulations at temperatures $T = 2000$ K (the liquid state) and $T = 100$ K (the amorphous state). The peaks in the partial radial distribution functions are obviously more pronounced for the system at temperature $T = 100$ K, and the second maximum is split into two peaks. As is well known, such peculiarities in the behavior of a radial function testify to the formation of local structures and are typical of amorphous materials [12].

WENDT–ABRAHAM PARAMETER AND TWO-PARTICLE CORRELATION ENTROPY

Wendt–Abraham parameter r^{WA} [13] and two-particle correlation entropy S_2 [14, 15] may be considered

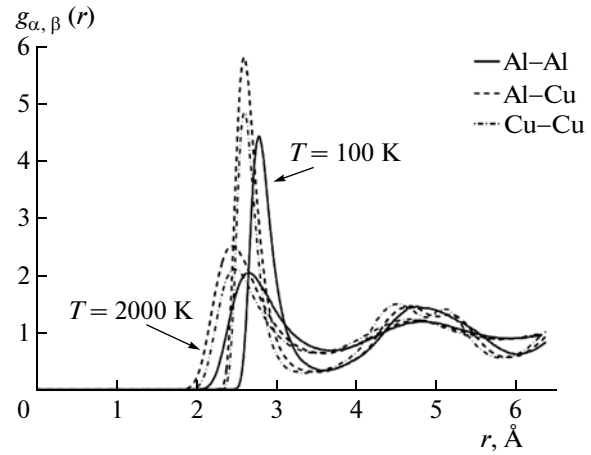


Fig. 1. Partial radial distribution functions of particles $g_{\alpha\beta}(r)$ of metallic alloy $\text{Al}_{50}\text{Cu}_{50}$ at temperatures $T = 2000$ and 100 K.

as additional factors characterizing local structural ordering:

$$r^{WA} = g_{\min}(r)/g_{\max}(r),$$

$$S_2 = -2\pi n \int_0^\infty \{g(r) \ln(g(r)) - [g(r) - 1]\} r^2 dr. \quad (3)$$

Here, $g_{\max}(r)$ and $g_{\min}(r)$ represent values of the first maximum and the first minimum of the radial distribution function of particles, respectively. A decrease in the values of these parameters will obviously lead to an increase in the degree of local structural order in the system. Figure 2 shows the temperature dependences of the Wendt–Abraham parameter and two-particle correlation entropy for different component pairs Al–Al, Al–Cu, and Cu–Cu. Two regions with different linear temperature dependences of the parameters are clearly seen, and critical glass transition of $\text{Al}_{50}\text{Cu}_{50}$ alloy temperature $T_c \approx 500$ K is defined by the intersection of the interpolation lines in the Wendt–Abraham parameter and by the change in the inclination of the temperature dependence of entropy.

SINGLE-PARTICLE DYNAMICS

Information on the single-particle dynamics observed at nanoscopic spatial scales is contained in incoherent scattering function

$$F_s(k, t) = \frac{1}{N} \left\langle \sum_{j=1}^N e^{-ik(\mathbf{r}_j(t) - \mathbf{r}_j(0))} \right\rangle, \quad (4)$$

the Fourier transform for which was measured in experiments on the scattering of slow neutrons [16]. Here, $\mathbf{r}_j(t)$ is the radius vector of the j -th particle at the moment of time t , and brackets $\langle \dots \rangle$ denote averaging over the ensemble of particles. Scattering function $F_s(k, t)$ was calculated on the basis of data on the

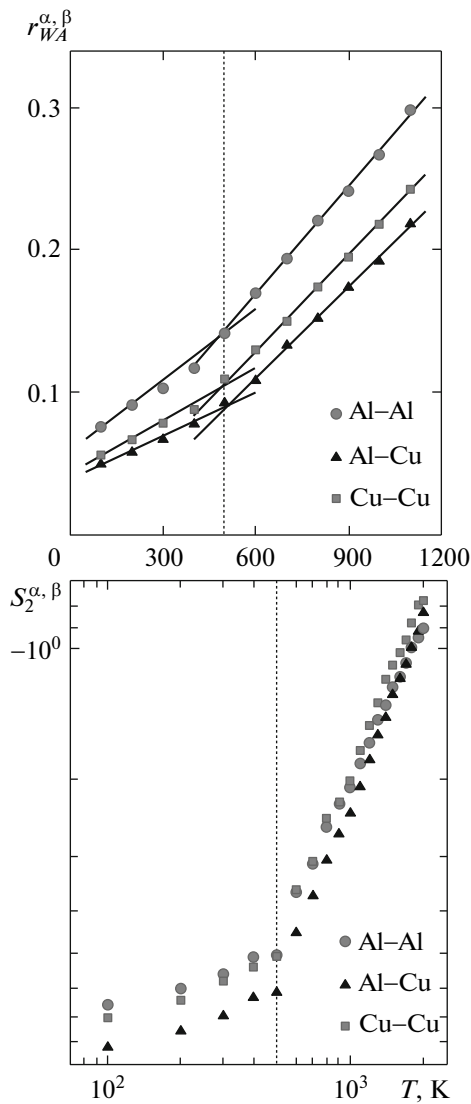


Fig. 2. Temperature dependences of the Wëndt–Abraham parameter and two-particle correlation entropy for different components of the system.

atomic dynamics simulations of $\text{Al}_{50}\text{Cu}_{50}$ binary system at the wave-number $k = 3.05 \text{ \AA}^{-1}$ near the maximum in the static structure factor $S(k)$ at the temperatures $T = 100, 500, 600, 1000,$ and 2000 K . The figure shows that with decreasing the temperature below T_c leads to so-called two-step relaxation with the appearance of a plateau in the incoherent scattering function. Using the Zwanzig–Mori projection operator technique [17], we can show that the behavior of incoherent scattering function $F_s(k, t)$ is described by the integro-differential equation

$$\ddot{F}_s(k, t) + \gamma \dot{F}_s(k, t) + \Omega_1^2 F_s(k, t) + \Omega_1^2 \int_0^t dt' M(k, t-t') \dot{F}_s(k, t') = 0, \quad (5)$$

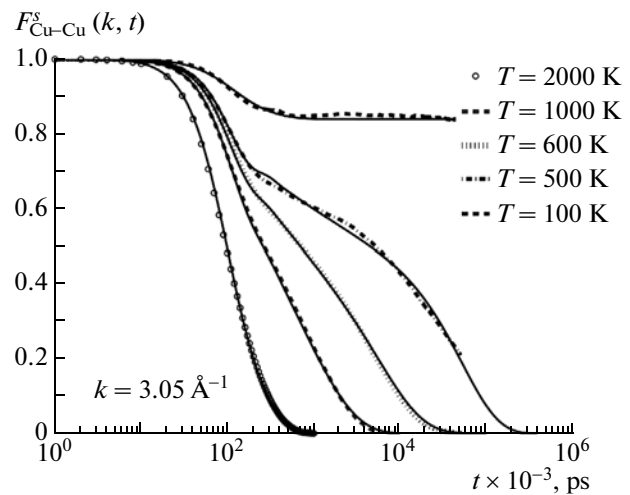


Fig. 3. Time dependences of incoherent scattering function of copper at the wave-number $k = 3.05 \text{ \AA}^{-1}$ for the liquid state ($T = 2000$ and 1000 K), supercooled liquid ($T = 600$ and 500 K), and the glass state ($T = 100 \text{ K}$). The solid line denotes theoretical results; markers denote modelling data.

where $\Omega_1^2 = k_B T / m_\alpha$ is the frequency relaxation parameter ($\alpha = \text{Al}, \text{Cu}$), $M(k, t)$ is the memory function, and γ is some constant value. According to the mode-coupling theory [18], we write memory function $M(k, t)$ as the polynomial

$$M(k, t) = v_1 F_s(k, t)^p + v_2 F_s(k, t)^q, \quad p, q \geq 0. \quad (6)$$

Here, coefficients v_1 and v_2 are defined as

$$v_1 = \frac{2\lambda - 1}{\lambda^2} (1 + \varepsilon), \quad v_2 = \frac{1}{\lambda^2} (1 + \varepsilon), \quad (7)$$

where $0.5 \leq \lambda < 1$, $\varepsilon = \pm(1/4)^n$ are the so-called interacting parameter. Since $\varepsilon = 0$, we have a transition from the ergodic state to nonergodic [18]. Substituting expression (6) into (5), we obtain an equation for function $F_s(k, t)$ that is solved numerically. Figure 3 shows a comparison of the theoretical results with the modeling data on atomic dynamics. It is evident that the theory perfectly describes the behavior of function $F_s(k, t)$ over the range of the temperature region under consideration. The theory allows us to adequately reproduce the changes in time dependence $F_s(k, t)$ associated with the transition in the region of the supercooled liquid and glass phase. In particular, the theoretical curves show the same features in long-time behavior of the incoherent scattering function with results of the atomic dynamics simulations, and appearance of continuous correlations in glass state.

CONCLUSIONS

This work presents the results of calculations the structural and dynamic properties of $\text{Al}_{50}\text{Cu}_{50}$ alloy, obtained by the atomic dynamics simulation. The crit-

ical glass transition temperature of Al₅₀Cu₅₀-alloy ($T_c \approx 500$ K) was defined on the basis of the temperature dependence analysis of the Wendt–Abraham parameter and the two-particle correlation entropy. It was established that the mode-coupling theory with a fractional–exponential memory function is applicable to describing the dynamic processes that occur on nanospatial scales in amorphous metallic alloys. The proposed model correctly reproduces the time dependences of the incoherent scattering function of the Al₅₀Cu₅₀ system, obtained from the atomic dynamics simulation for a wide temperature range.

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REFERENCES

1. Frenkel, Y.I., *Vvedenie v teoriyu metallov* (Introduction into the Theory of Metals), Leningrad: Nauka, 1972.
2. Waseda, Y., *The Structure of Non-Crystalline Materials: Liquids and Amorphous Solids*, New York: McGraw–Hill, 1980.
3. Suzuki, K., Fudzimoru, K., and Hashimoto, K., *Amorfnye metally* (Amorphous Metals), Moscow: Metallurgiya, 1987.
4. Clement, W., Willens, R.H. and Duwez, P., *Nature*, 1960, vol. 187, p. 869.
5. Greer, A.L., *Science*, 1995, vol. 267, p. 1947.
6. Jonson, W.L., *MRS Bull.*, 1999, vol. 24, p. 42.
7. Glezer, A.M., *Ros. Khim. Zh.*, 2002, vol. XLVI, p. 57.
8. Cugliandolo, L.F., *Physica A*, 2007, vol. 386, no. 2, p. 597.
9. Cai, J. and Ye, Y.Y., *Phys. Rev. B*, 1996, vol. 54, p. 8398.
10. Khusnutdinoff, R.M., Mokshin, A.V., and Yulmetev, R.M., *Zh. Eksp. Teoret. Fiz.*, 2009, vol. 135, p. 477 [*JETP* (Engl. Transl.), 2009, vol. 108, no. 3, p. 417].
11. Allen, M.P. and Tildesley, D.J., *Computer Simulation of Liquids*, Oxford: Clarendon Press, 1987.
12. Sarkisov, G.N., *Usp. Fiz. Nauk*, 2002, vol. 172, p. 647.
13. Wendt, H.R. and Abraham, F.F., *Phys. Rev. Lett.*, 1978, vol. 41, p. 1244.
14. Truskett, T.M., Torquato, S., Sastry, S., et al., *Phys. Rev. E*, 1998, vol. 58, p. 3083.
15. Nettleton, R.E. and Green, M.S., *J. Chem. Phys.*, 1958, vol. 29, p. 1365.
16. Turchin, V.F., *Medlennye neitrony* (Slow Neutrons), Moscow: Gosatomizdat, 1963.
17. Zwanzig, R., *Nonequilibrium Statistical Mechanics*, Oxford: Oxford Univ. Press, 2001.
18. Götze, W., *Liquids, Freezing, and the Glass Transition*, Amsterdam: North–Holland, 1991.