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Shear Influence on the Structural Ordering in a Model Metallic Glass[1]

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Abstract—Nonequilibrium molecular dynamics simulations of a model amorphous system are performed with the aim of studying the structural transformations induced by external shear influence. We reveal that the shear drive has both positive and negative effects on the structural ordering processes. The dependence of the phase transition rate versus the strain rate at three different temperatures is found.

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INTRODUCTION

Glasses as well as liquids are characterized by a lack of long-range order in the positions of their atoms. At the same time, a glassy state is out of thermodynamical equilibrium, which is why the processes of structural relaxation move the glassy system to a more advantageous energy state. Structural rearrangements in glasses are very slow because of their high viscosity, which suppresses the processes of the appearance of crystalline nuclei and their further growth [1, 2]. These external influences, which lead to an increase of atom mobility in a glass, must therefore be governed by crystallization in an amorphous system.

The present work gives the results of nonequilibrium molecular dynamics simulations—modeling the influence of shear on the structural ordering in a one-component metallic glass. With the help of the mean-first passage time technique and by computing the orientational order parameter for the deformation of a system with different rates, we determine the transition rates from amorphous phase into an ordered phase.

NONEQUILIBRIUM MOLECULAR DYNAMICS

Nonequilibrium Molecular Dynamics. The interaction of the atoms in the system occurs via the pair potential

$$U(r^*)/\varepsilon = A(r^{*-m} - B)\exp\left(\frac{c}{r^* - a}\right)\Theta(a - r^*) + B\exp\left(\frac{d}{r^* - b}\right)\Theta(b - r^*), \quad r^* = r/\sigma, \tag{1}$$

which was originally suggested in [3] as a model for a simple glass-forming liquid metal. The quantities ε and σ included in Eq. (1) define the unit energy and length, respectively; the potential parameters have the following values: $A = 5.82$, $a = 1.87$, $B = 1.28$, $b = 1.94$, $c = 1.1$, $d = 0.27$ and $m = 16$, and $\Theta(\ldots)$ is the Heaviside function. A minimum in potential (1) is observed at $r^* = 1.13$ with a form identical to that of the minimum in the Lennard–Jones model, while the maximum is located at $r^* = 1.628$. The presence of this additional repulsive contribution in interpartical interaction leads to the suppression of the crystalline structures with FCC and BCC lattices, but it favors the formation of quasicrystals under certain conditions [3]. A feature of this one-component system is its ability to generate an amorphous state at low temperatures. The equilibrium diagram of the system was studied in detail over a wide interval of temperatures and pressures in [4]. Below, the values of the lengths and energies will be given in units of σ and ε, respectively, and time will be given in units of $\tau = \sigma\sqrt{m/\varepsilon}$, where m is the mass of the atom [5].

The system initially took the form of a cubic cell with volume $V = L^3$ ($L = 28.55\sigma$ is the length of the cube edge), inside of which $N = 19652$ identical atoms interacted via potential (1). Integration of the equations of motion was performed by means of the Verle algorithm with the application of periodic boundary conditions in the x-, y-, z-directions for the simulation cell [6]. The system was equilibrated at the thermodynamic state with temperature $T = 2.0\varepsilon/k_B$ and zero pressure, which corresponds to the liquid state according to the phase diagram [4]. The system was then rapidly supercooled by 96–99% to states with the temperatures $T = 0.01$, 0.03 and $0.06\varepsilon/k_B$. The obtained glassy state at these temperatures was verified by the

[1] The article was translated by the authors.

virtual absence of atom diffusion and the radial atom distribution typical for an amorphous systems (the lower curve on Fig. 1).

Simulation of the influence of shear on the system was performed via two amorphous walls created at the cell faces in the direction of the y-axis. The thickness of the each wall was three atomic diameters. The top wall was moved along the x-axis at the rate $\mathbf{\upsilon}(t) = \dot{\gamma} L_y(t)\mathbf{e}_x$ with fixed shear rate $\dot{\gamma}$ and constant external pressure $P_{yy} = 7.62\varepsilon/\sigma^3$. The term $L_y(t)$ denotes the distance between the walls. The bottom wall was fixed [8]. The temperature control of the simulation cell upon the influence of shear was performed by rescaling the z-component of the atom velocities [7], while the periodic boundary conditions were applied in the -x and z-directions only.

Results of the Structural Analysis. The structural analysis was performed on the basis of the radial distribution of the atoms in the system,

$$g(r,t) = \frac{1}{N\rho}\left\langle \sum_i \sum_{j\neq i} \delta(\mathbf{r} - \mathbf{r}_{ij}, t) \right\rangle, \qquad (2)$$

and the orientational order parameter [9]

$$Q_6 = \left(\frac{4\pi}{13} \sum_{m=-6}^{6} \left| \frac{\sum_{i=1}^{N} \sum_{j=1}^{N_b(i)} Y_{6m}(\theta_{ij}, \varphi_{ij})}{\sum_{i=1}^{N} N_b(i)} \right|^2 \right)^{1/2}, \qquad (3)$$

where $\rho = N/V$ is the numerical density, $Y_{6m}(\theta_{ij},\varphi_{ij})$ are the spherical harmonics, θ_{ij} and φ_{ij} are the polar and azimuthal angles between radius vector $\mathbf{r}_{ij}$ and the selected direction, and N_b is the number of particles forming the nearest neighborhood of particle i. The nearest neighborhood is the particles located inside a sphere of radius $r = 1.5\sigma$, where the center of the sphere coincides with the center of particle i. For a completely disordered system, we have $Q_6 \to 0$; for an FCC structure, we obtain $Q_6 = 0.5745$; and for a BCC structure, we find $Q_6 = 0.5106$. The increase in values of the order parameter Q_6 thus indicates the orientational ordering in the system, while the appearance of the translational order is reflected in the well-defined peaks in the radial distribution $g(r,t)$ at large r.

Variation over time in the radial particle distribution $g(r,t)$ and of the orientational order parameter $Q_6(t)$ in the system upon the influence of shear at a rate of $\dot{\gamma} = 0.005\tau^{-1}$ is presented in Fig. 1. It is clearly seen that at $t \approx 500\tau$, pronounced peaks in the radial distribution function $g(r,t)$ appear at distances $r > 2\sigma$, evidence of the structural ordering in the system. The increase in the orientational order parameter $Q_6(t)$, observed at $t \approx 500\tau$, and the following attainment of saturation, $Q_6(t \geq 1100\tau) \approx \text{const}$, are also evidence of

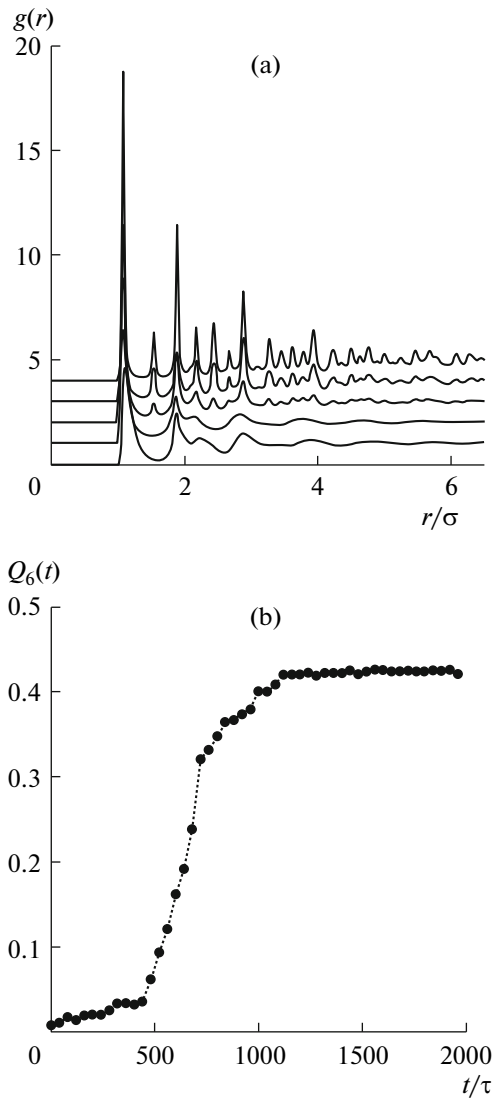

Fig. 1. Structural characteristics of the system at temperature $T = 0.03\varepsilon/k_B$ under external shear influence with shear rate $\dot{\gamma} = 0.005\tau^{-1}$. (a) Radial distribution of atoms at the different times: t= 0, 400, 600, 800 and 1600τ (from bottom to top). (The curves move upwards for the sake of clarity.) The form of the lower curve, reflecting the particle distribution before the influence of shear ($t = 0$), is typical for an amorphous system [7]. (b) Evolution of the orientational order parameter Q_6.

the system's transition to an ordered phase [8]. It should be noted that the free evolution of the system (without external influences) does not reveal a phase transition within such a time scale [8].

Phase Transition Rate. The influence of shear with rate $\dot{\gamma}$ on the transition can be characterized by the rate of the observed transition $\tau_{tr}^{-1}(\dot{\gamma},T)$. To estimate the phase transition rate, we use the so-called method of "mean first-passage time" [10, 11]. According to this method, it is necessary to define the

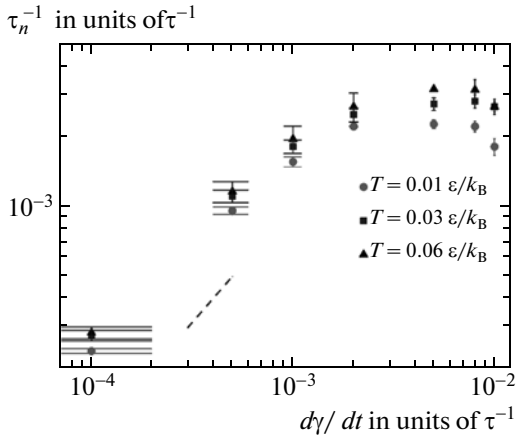

Fig. 2. Dependence of the transition rate τ_{tr}^{-1} versus shear rate $\dot{\gamma}$ at three different temperatures $T = 0.01$, 0.03 and $0.06\varepsilon/k_B$. The dashed line corresponds to the unit slope.

average time $\bar{\tau}$ during which the system first attains a state with some value of the order parameter Q_6): $Q_6 \to Q_6^{(1)}, Q_6^{(2)}, Q_6^{(3)},\ldots$, where $Q_6^{(i)} \in R$, R is the domain of states [11]. For transitions with high activation barriers, distribution $\bar{\tau}(Q_6)$ has assumes a sigmoidal form with a pronounced plateau $\bar{\tau}_{pl}$ that indicates the system's attaining of some stable state. Distribution averaging $\bar{\tau}(Q_6)$ is performed over the set of independent experiments. The transition rate into this state can then be determined directly as $\tau_{tr}^{-1} = \bar{\tau}_{pl}^{-1}$ [8].

The dependences of the found transition rates versus the shear rate $\dot{\gamma}$ at three different temperatures $T = 0.01$, 0.03 and $0.06\varepsilon/k_B$ are presented in Fig. 2. It can be seen that linear growth of the values of transition rate τ_{tr}^{-1} is observed at low values of $\dot{\gamma}$ for all the considered values of temperature, $\tau_{tr}^{-1} \sim \dot{\gamma}^{\alpha}$, where $\alpha \approx 1$. Nevertheless, starting with some critical value of the shear rate, the values of the transition rate cease growth and begin to decline somewhat. Such nontrivial behavior indicates the dual impact of external shear on the structural ordering in the amorphous system. On the one hand, insignificant shear influence leads to an increase in atom mobility and, as a result, to the formation of crystalline clusters, which accelerates the transition of the system to a more advantageous state, energy-wise [5]. On the other hand, strong shear influence leads to the fracture of the forming crystalline structures. It should be noted in conclusion that a similar dependence of the nucleation rate versus the shear rate was recently observed for the case of the two-dimensional Ising model under the influence of shear [12].

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REFERENCES

1. Skripov, V.P. and Faizullin, M.Z., *Crystal-Liquid-Gas Phase Transitions and Thermodynamic Similarity*, Moscow: Fizmatlit Publishers, 2003, p. 160.
2. Schmelzer, J.W.P., *Nucleation Theory and Applications*, Weinheim: Wiley, 2005, p. 469.
3. Dzugutov, M., *Phys. Rev. A*, 1992, vol. 46, p. R2984.
4. Roth, J. and Denton, A.R., *Phys. Rev. E,* 2000, vol. 61, p. 1063.
5. Mokshin, A.V. and Barrat, J.-L., *Phys. Rev. E*, 2008, vol. 77, p. 021505.
6. Allen, M.P. and Tildesley, D.J., *Computer Simulation of Liquids*, Oxford Clarendon Press., 1987, p. 387.
7. Binder, K. and Kob, W., *Glassy Materials and Disordered Solids,* London: World Sci. Publ., 2005, p. 405.
8. Mokshin, A.V. and Barrat, J.-L., *J. Chem. Phys.,* 2009, vol. 130, p. 034502.
9. Steinhardt, P.J., Nelson, D.R., and Ronchetti, M., *Phys. Rev. B*, 1983, vol. 28, p. 784.
10. Pontryagin, L.S., Andronov, A., and Vitt, A.A., *JETP*, 1933, vol. 3, p. 165.
11. Hänggi, P., Talkner, P., and Borkovec, M., *Rev. Mod. Phys.*, 1990, vol. 63, p. 251.
12. Allen, R.J., et al., *J. Chem. Phys.*, 2008, vol. 129, p. 134704.