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Three-particle correlations in liquid and amorphous aluminium

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HIGHLIGHTS

- The original method of three-particle structural analysis is proposed.
- The original three-particle correlation function is introduced.
- The time evolution of three-particle correlations is evaluated for liquid and amorphous aluminium.
- The "stable" disordered structures are detected, that are formed by triplets with various configurations.
- Transitions between the various configurations of triplets are observed.

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ABSTRACT

Analysis of three-particle correlations is performed on the basis of simulation data of atomic dynamics in liquid and amorphous aluminium. A three-particle correlation function is introduced to characterize the relative positions of various three particles—the so-called triplets. Various configurations of triplets are found by calculation of pair and three-particle correlation functions. It was found that in the case of liquid aluminium with temperatures 1000 K, 1500 K, and 2000 K the three-particle correlations are more pronounced within the spatial scales, comparable with a size of the second coordination sphere. In the case of amorphous aluminium with temperatures 50 K, 100 K, and 150 K these correlations in the mutual arrangement of three particles are manifested up to spatial scales, which are comparable with a size of the third coordination sphere. Temporal evolution of three-particle correlations is analyzed by using a time-dependent three-particle correlation function, for which an integro-differential equation of type of the generalized Langevin equation is output with help of projection operators technique. A solution of this equation by means of mode-coupling theory is compared with our simulation results. It was found that this solution correctly reproduces the behavior of the time-dependent three-particle correlation functions for liquid and amorphous aluminium.

1. Introduction

At present time the main attention is paid to study the structure of condensed systems, which are in equilibrium or in metastable states [1–4]. Often, the traditional experimental methods of structural analysis do not allow to correctly identify the presence of some structures in bulk systems due to their small sizes, either low concentrations in the system, or due to relatively short lifetimes. Usually, information about the structure of condensed systems is extracted by using microscopic

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methods or by methods of X-ray and neutron diffraction. Here, the static structure factor determined from the experimental data is critical quantity, which is related with the pair distribution function, $g(r)$ [2,4]. At the same time, the pair distribution function $g(r)$ can be determined on the basis of atomic/molecular dynamics simulations, and then it can be compared with experimental data.

In addition, the three-particle correlations has an essential impact for different processes in condensed systems. Thus, an account for three-particle correlations is required to explain the dynamic heterogeneity in liquids [5,6], to describe of transport properties in chemical reactions [4,7], to study the structural heterogeneity of materials at mechanical deformations [8,9], to detect the nuclei of on ordered phase (i.e. crystalline, quasicrystalline) [10,11], to describe the amorphization of liquids at rapid cooling [2,4,12]. Direct evaluation of three-particle correlations by means of experimental measurements is extremely difficult problem [4,13,14]. Here, the special methods must be adopted to extract such information [4,15,16]. On the other hand, detailed information about the three-particle correlations can be obtained on the basis of atomic/molecular dynamics simulations data. Note that early studies of the three-particle correlations were focused mainly on simple liquids such as the Lennard-Jones fluid, the hard spheres system, and the colloidal systems [4, 17–20]. In some recent studies, on the basis of data of the molecular dynamics simulation the three-particle correlations were estimated in such systems as carbon nanotubes, electrolytes, metallic melts and alloys [2,21,22]. In these studies, the information about three-particle correlations is usually extracted from the time evolution of two and more parameters characterized the positions and trajectories of the particles relative each other.

In the present work, the original method of three-particle structural analysis and evaluation of time-dependent three-particle correlations is proposed, where the arbitrary trajectories of motion of the various three particles (that will be denoted as triplets) are considered. The method allows one to identify the presence of ordered crystalline and "stable" disordered structures, which are difficult to be detected by conventional methods of structural analysis (for example, such as the Voronoi's tessellation method [23], the Delaunay's triangulation method [24], the bond-orientational order parameters [3]). The applicability of this method will be demonstrated for the case of the liquid and amorphous aluminium.

2. Simulation details

We performed the atomic dynamics simulation of liquid and amorphous aluminium. The system contains $N = 864$ atoms, located into a cubic simulation cell with periodic boundary conditions in all directions. The interatomic forces are calculated through EAM-potential [25,26]. The velocities and coordinates of atoms are determined through the integrating Newton's equations of motion by using Velocity-Verlet algorithm with the time-step $\Delta t = 1$ fs.

Initially, a crystalline sample with *fcc* lattice and numerical density $\rho = 1.23\,\sigma^{-3}$ (or mass density 2300 kg/m^3) was prepared, where $\sigma = 2.86$ Å is the effective diameter of the aluminium atom. Further, the system was melted to the temperatures $T = 1000$, 1500, and 2000 K. The amorphous samples were generated through the fast cooling with the rate 10^{12} K/c of a melt at the temperature 2000 K to the temperatures $T = 50$, 100, and 150 K (the melting temperature is $T_m \simeq 934$ K). The simulations were performed in NpT-ensemble at constant pressure $p = 1$ atm.

3. Methods

3.1. Three-particle correlation function

Let us consider a system, consisting of N classical particles with same masses m, which are located into the simulation cubic cell with a volume V. From the geometric point of view, locations of any three particles generate a triangle (i.e. triplet). This triplet is characterized by the area, S. Then, the area of ith triplet, S_i, at time t (here $i \in \{1, 2, \ldots, N_T\}$, N_T is the number of all possible triplets in the system) is defined by

$$S_i(t) = \left\{ l_i(t) \cdot [l_i(t) - r_i^{(12)}(t)] \cdot [l_i(t) - r_i^{(23)}(t)] \cdot [l_i(t) - r_i^{(31)}(t)] \right\}^{1/2}. \tag{1}$$

Here, $l_i(t) = [r_i^{(12)}(t) + r_i^{(23)}(t) + r_i^{(31)}(t)]/2$ is the semiperimeter of ith triplet; $r_i^{(12)}$, $r_i^{(23)}$, and $r_i^{(31)}$ are the distances between the vertices of ith triplet with conditional labels 1, 2, and 3. It follows from Eq. (1) that the area of ith triplet, S_i, takes positive values and values closed to zero. Different triplets can be correlated to the same values S_i, and the triplets are independent from each other (i.e. have no common vertices) or interconnected (i.e. one or two vertices are mutual). To estimate the quantity S_i one vertex of triplet is considered as central (i.e. fixed), relative to which the positions of the other vertices must be defined (see Fig. 1).

To determine the probability of emergence of the triplets with the area S, the three-particle correlation function is introduced

$$g(S) = \frac{1}{N_T} \sum_{i=1}^{N_T} \delta(S - S_i). \tag{2}$$