ANISOTROPIC X-RAY SCATTERING FROM SIMULATED AMORPHOUS STRUCTURES

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Two 800-atom cluster models of amorphous copper were constructed utilizing the method of continuous static relaxation of nonrigid spheres developed by the authors. The Morse and Ballog potentials were used to construct and relax the structures. The angular dependence of the x-ray scattering was simulated for both clusters, and a rather appreciable anisotropy was found for them. In order to elucidate the causes of the observed anisotropy, a distribution of atoms in thin sections perpendicular to the direction of the main peak was investigated. Quasiperiodic fluctuations in the occupation numbers of the sections with a period roughly corresponding to the radius of the first coordination sphere were found. A special numerical—graphical analysis showed that the atoms contributing to the main peak form pseudoplanes corresponding to fluctuations in the occupation numbers of the sections. In spite of appreciable differences in the character of interatomic interactions, the density, and the microstructure of the modeled clusters, the pseudocrystalline motifs in both clusters were very similar. A comparison was performed of the results obtained from the scattering pattern for an 800-atom unrelaxed stochastic cluster especially generated for the investigation of the influence of relaxation on the anisotropy. It is shown that the relaxation somewhat increases the anisotropy and orders the displacement of the atoms within the sections although on the whole the scattering pattern in all cases is similar. The results obtained correlate well with published data.

The microscopic structure of met-glasses is of particular interest, inasmuch as the spatial distribution of atoms together with the assigned interaction potentials completely determine all the properties of modeled materials. Contemporary procedures for the computer simulation of topologically disordered structures randomly pack some region of space with atoms. Such packing, generally speaking, is inhomogeneous, but macroscopically ought to be isotropic. Also, the interatomic interaction, which in met-glasses is always spherically symmetric, is not a source of anisotropy. Nevertheless, careful analysis of the angular dependence of x-ray scattering from simulated structures has established anisotropy in the distribution of scattered radiation [1, 2]. Beyond this, the anisotropy present in real amorphous structures has been observed by precision electron microscopy [3, 4].

Here, the angular dependence of x-ray scattering for two 800-atom copper clusters constructed by the method of continuous static relaxation was investigated [5]. For the construction and relaxation of the first cluster an empirical Morse potential was employed; and for the second cluster the Ballog potential was employed. For brevity, the first cluster from here on will be referred to as the Morse Model (MM), and the second cluster as the Ballog Model (BM). A detailed description of the potentials and the scheme for choosing the adjustable parameters are discussed in [6]. Here we are limited to just a short description of the potentials under study.

A Morse potential was chosen with the form

\[ V_M(R) = D\beta \exp(-\alpha R) (\beta \exp(-\alpha R) - 2), \quad (1) \]

where \( D = 0.3611 \text{ eV}, \alpha = 1.282 \text{ Å}^{-1}; \beta = 36.76 \) is the adjustable parameter determined from experimental data for crystalline copper [6].

The composite Ballog potential differs from zero in the interval \( 0 \leq R \leq 4.42 \text{ Å} \) and is a superposition of ten cubic polynomials, each of which has its own region of definition:

\[ V_B(R) = A_k (R - R_k)^3 + B_k (R - R_k)^2 + C_k (R - R_k) + D_k, \quad (2) \]

where \( A_k, B_k, C_k, D_k \) are the coefficients determined from experimental data for crystalline copper [6].
Fig. 1. Dependence of the Morse and Ballog potentials on distance.

Fig. 2. Structure factors of the simulated clusters. The solid line is for MM, the dashed line is for BM.

\[ V(R) = 0 \]

Here the simulation of the structures is accomplished by a method originally developed by the authors [5], in which the hard-sphere approximation characteristic of classical methods [7-10] is generally not used. Modeling of the structure was performed as follows. Each atom from the very start was represented by some previously chosen potential with an interaction radius \( R_{\text{int}} \) determined from the form of the potential for \( R > R_{\text{int}} \), \( V(R) = 0 \). The first atom was placed at the origin and the second was placed at the position of the minimum in the potential of the first; the third and all subsequent atoms were placed at the minimum of the summed potentials of all the atoms falling inside the interaction sphere with radius \( R_{\text{int}} \) constructed around the added atom. The procedure was continued until the number of atoms in the model reached the assigned value. Then the simulated structures were relaxed using a simple gradientless minimization of the total energy of the system by the method of nonlinear simplexes [11].

Relaxation of the cluster was accomplished as follows. A sphere with radius \( R_{\text{int}} \) was constructed successively around each atom; atoms were chosen falling within this sphere of interaction; their total potential was calculated, and the central atom was displaced to the minimum of the summed potential. After running over all the atoms (global iteration), the configurational energy as a function of the total pair interactions was calculated. In the relaxation process the energy of each configuration was compared with the preceding energy, and the relaxation was stopped when the relative change of energies fell to some fraction of a percent.

The simulation procedure developed by the authors corresponds to the real process of the formation of amorphous materials by precipitation from the gas phase at low temperatures. The specific scattering intensity in electron units for an ensemble of \( N \) scatterers, located at the points \( \mathbf{R}_i \) can be written as follows [1]

\[ I(\kappa) = \frac{|f|^2}{N} \sum_i f \exp \left( i\kappa \mathbf{R}_i \right)^2, \]  

where \( \kappa \) is the scattering vector, \( f \) is the atomic form-factor. It is convenient to transform from the Cartesian coordinates \( \kappa_x, \kappa_y, \) and \( \kappa_z \) to spherical coordinates \( r, \theta, \) and \( \phi \); in addition \( r \) was chosen equal to the ordinate of the main peak for the average scattering calculated according to Debye’s equation

\[ I(\kappa) = \sum_{ij} f_i(\kappa) f_j(\kappa) \frac{\sin \left( \kappa R_{ij} \right)}{\kappa R_{ij}}, \]  

where \( R_{ij} \) is the distance between atoms \( i \) and \( j \).

In Fig. 2, the dependence of \( I(\kappa) \) for clusters constructed using the potentials (1) and (2), respectively, are shown; moreover, for the Morse model \( \kappa_{\text{max}} = 3.2 \, \text{Å}^{-1} \) and \( I_{\text{max}}^D = 3.49 \), and for the Ballog model \( \kappa_{\text{max}} = 2.92 \, \text{Å}^{-1} \) and \( I_{\text{max}}^D = 3.75 \). The position of the main peaks is well correlated with the behavior of the potentials (1) and (2). The long-range attraction of the