Available online at www.sciencedirect.com
ScienceDirect
Acta Materialia 58 (2010) 615–625
Acta MATERIALIA
www.elsevier.com/locate/actamat

The geometry, topology and structure of amorphous solids

Cheuk-Yu Lee [a], Zbigniew H. Stachurski [a,*], T. Richard Welberry [b]

[a] College of Engineering and Computer Science, Australian National University, Canberra, ACT 0200, Australia
[b] Research School of Chemistry, Australian National University, Canberra, ACT 0200, Australia

Received 21 July 2009; received in revised form 21 September 2009; accepted 21 September 2009
Available online 21 October 2009

Abstract

Clusters of atoms can be divided into three categories depending on their topology. One of the categories provides the basis for development of a model of a perfectly random structure (ideal amorphous solid) using the non-equilateral triangle topology in the coordination shell. Metallic glasses solidify as amorphous solids with random arrangement of atoms. A model of Zr-based metallic glass has been constructed and described in terms of cluster topology, and compared with a recently published dynamic molecular model of the same alloy. It is shown that the pair distribution function for the ideal amorphous model relates to the pair correlation function obtained from the dynamic model. Debye X-ray scattering computations reveal the presence of vacancies and other flaws relative to the ideal amorphous solid. A shift in the peak position can be predicted using the Erhenfest formula. Two atomic displacement mechanisms involving a five-atom sub-cluster are identified as the fundamental means of compositional redistribution between clusters in the alloy.

Keywords: Cluster topology; Multiatomic glass; Debye scattering; Amorphousness

1. Introduction

The understanding of the atomic-scale structure of solids (from which modern technology and society benefit so greatly) has come about to a large degree because of the development of the methods of geometry and X-ray crystallography. From the first discovery of the diffraction of X-rays by a crystal in 1912 [1] to the present day, when structures of large protein molecules are determined routinely, crystallography has developed from small beginnings to become an enormously successful and powerful tool. Of pivotal importance in this success is the fact that the methods of crystallography are based on the concept of an ideal (perfect) crystal in which unit cells or building blocks of the material are stacked in perfectly repeating rows and columns to form a periodic array of atoms or molecules (of infinite extent). This same ideal is assumed for the very simplest to the most complex of structures. Although real crystals only ever approximate this ideal (some not very closely), it is this ideal structure which is always used as a permanent baseline relative to which real crystalline materials are compared and can be understood.

In 1984 a new class of solid materials were discovered [2] that possessed long-range orientational order but no translational symmetry. These so-called quasicrystals gave sharp diffraction peaks like crystals but had symmetries incompatible with those found for normal crystals. As this field has developed over the last 25 years, a model that has been used extensively to understand the structure of these novel materials is the Penrose tiling model [3]. This structure is envisaged to be made up of two different types of building blocks (tiles), which take the place of the single unit cell of the ideal crystal model. Although real quasicrystals do not conform exactly to the ideal Penrose model, it nevertheless plays a role of prime importance in providing the same kind of baseline relative to which real quasicrystals may be understood.

For the third class of solid materials, namely amorphous or glassy materials, the situation has not been so

* Corresponding author. Tel.: +61 2 6125 5681; fax: +61 2 6125 0506.
E-mail address: zbigniew.stachurski@anu.edu.au (Z.H. Stachurski).

1359-6454/$36.00 © 2009 Acta Materialia Inc. Published by Elsevier Ltd. All rights reserved.
doi:10.1016/j.actamat.2009.09.040

satisfactory and there is still much debate concerning the exact nature of these materials. In a recent article, Sheng et al. [4] say that "the atomic arrangements in amorphous alloys remain mysterious at present". A search of recently published literature on atomic structure of metallic glassy alloys will reveal that the main effort towards solving this problem is directed towards atomistic molecular dynamics simulations. Since glassy materials are non-equilibrium structures, one should anticipate that the results of each simulation, carried out in different laboratories, will be different; no asymptotically unique structure can be achieved. In light of the above, one can conjecture that this is not the right approach to define the ideal baseline model for the structure of amorphous metals. Instead, a geometrical model should be sought. Such a geometrical ideal amorphous solid (IAS) for mono-sized spheres has been described in detail elsewhere [5], and we follow that approach here to describe the ideal amorphous structure of a Zr-based metallic glass. Coincidentally, a molecular dynamics model of the very same Zr-based metallic glass has been just published by Hui et al. [6], and this gives a special opportunity for comparison and analysis of what are at present the best atomistic dynamic model, with the best theoretical baseline model for this material.

2. Geometric simulations

2.1. IAS model of $[Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}]$ metallic glass

According to the specific IAS rules [5], the construction of random packing of spheres of m types is as follows:

Step 1: Begin by setting up m virtual bins, each bin containing enough of each type of sphere.
Step 2: Place one sphere of any type at the origin ($x = y = z = 0$).
Step 3: Place k spheres (normally $\bar{k}$) in contact with the sphere at the centre, forming a cluster of random configuration. It is essential to use the method of division of the sphere's surface into equal areas [7] to avoid bias (see Fig. 1). Pick up spheres from the bins with a frequency in proportion to the required composition.
Step 4: Identify all three-adjacent-sphere sites formed on the surface of the created cluster and sort the sites in ascending order of distance from the origin.
Step 5: Add spheres of m type on identified sites corresponding to the required composition and in order from the closest to the furthermost from the centre. Reject overlapping spheres.
Step 6: Repeat the previous two steps as many times as required; in principle, the additions can be carried out to infinity; in practice, the simulation ends with a cell of finite dimensions.

The outcome of this computational process is a three-dimensional geometrical pattern of randomly packed spheres, called a "Round Cell" if it is of finite dimensions or an IAS if it is of infinite extent. The essential information about the Cell is stored in a matrix, $[\mathbf{x}_n, m]$, where the vectors $\mathbf{x}_n$ define the positions of all atoms/spheres with respect to the origin and m identifies the corresponding type of the sphere, including its radius. It is a structure of special geometrical and topological properties. As a rule, the centres of any three adjacent spheres form triangles of unequal sides due to (i) some/all spheres not touching and (ii) different sphere radii. There is not a single incidence of four adjacent spheres that are coplanar, in direct contrast to any of the crystallographic Bravais lattices. Consequently, there is no translational symmetry in this structure. The geometrical construction of IAS lends itself readily to description and analysis by Voronoi tessellation and associated Delauny simplexes. This includes structure

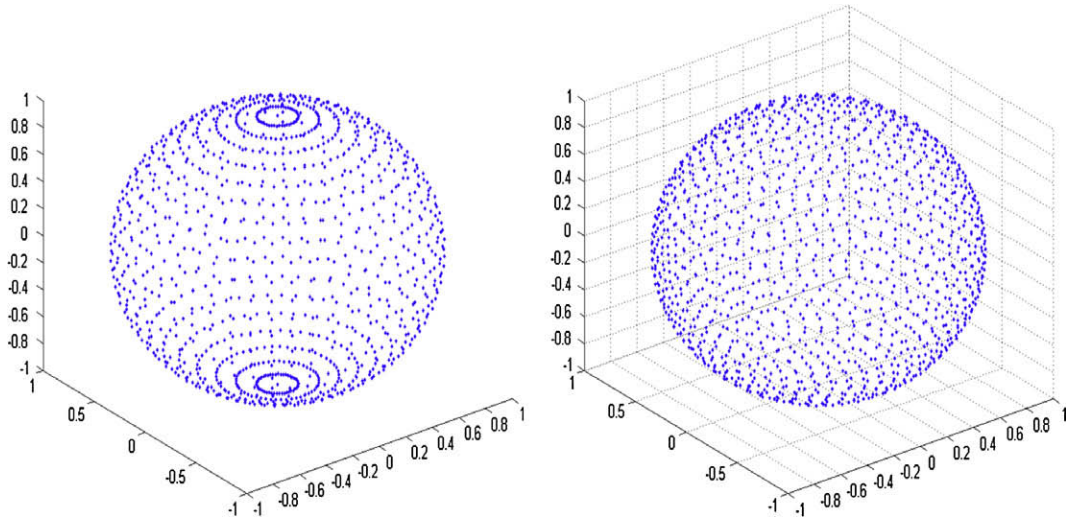

Fig. 1. Density of points on the surface of a sphere: (a) according to the equal area scheme [7] and (b) according to equal probability for the spherical variables: $0 \leqslant \Theta \leqslant \pi, 0 \leqslant \phi \leqslant 2\pi$.