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## X-ray scattering from amorphous solids

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## ABSTRACT

The main objective of this work is to verify the proposed models by comparing computed outcomes with experimental results. For metallic glasses the novel ideal amorphous solid model is used to simulate the structure and the atomic positions which are input into the Debye equation. Computations predict structure factor or scattered intensity which agree well with the experimentally obtained data. For covalent materials, such as amorphous silica or amorphous polyethylene with short range order the Warren approach offers a simple method to predict X-ray scattering in good agreement with experimental data. Neither of the two above methods works well for chalcogenide glasses which require calculations involving spherical harmonics.

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## 1. Introduction

The pioneering findings of von Laue and Braggs on X-ray diffraction led rapidly to the determination of precise locations of atoms in crystalline minerals and materials. Studies of space group symmetry provided theoretical basis for definition and description of crystal structures, corroborated by the diffraction methods. For amorphous solids the geometrical methods are less well developed. The positions of atoms are usually not known *a priori* and the vision of atomic arrangements is difficult to perceive clearly [1]. However, if the positions of atoms are known, say, at  $r_0, r_1, r_2, \dots, r_N$  with their corresponding atomic scattering factors,  $f_1, f_2, \dots, f_N$ , then the scattered intensity emanating from such a solid can be predicted by the Debye equation [2]:

$$I(s) = \sum_i \sum_j f_i(s)f_j(s) \frac{\sin(2\pi sr_{ij})}{2\pi sr_{ij}}, \text{ for } i \text{ and } j \text{ from } 0 \text{ to } N, \quad (1)$$

where:  $r_{ij} = |r_j - r_i|$  is the distance between scattering centers, and  $s = |(S - S_0)|/\lambda$  is the magnitude of the scattering vector. The phase difference between any two scattered rays is  $\delta = 2\pi sr_{ij}$ , determined only by  $r_{ij}$  for a fixed angle of scattering, with an equal probability of phase difference having any value between 0 and  $2\pi$ .

It is assumed that a model for the atomic-scale arrangements exists. Then, the main objective is to verify the proposed model by a trial-and-error method [3]. A crucial test is to compare the predicted

scattering trace with experimentally derived data. Then there are three possible outcomes: (i) the agreement is good in which case the model is accepted, (ii) the agreement is not good, in which case it is rejected, or (iii) the model is modified to improve the agreement until case (i) is reached. The final acceptance of the atomic arrangement is achieved only in corroboration with other complementary methods.

## 2. Experimental methods

## 2.1. Preparations of samples

## 2.1.1. Metallic glasses

The range of samples covered in this study is listed in Table 1. The Fe–B alloy samples were between 25 and 30  $\mu\text{m}$  in thickness, the Ni–Si–B were 35  $\mu\text{m}$ , and the Ti–Cu–Zr were  $\sim 30 \mu\text{m}$ , respectively. All the ribbon-like samples were made by melt spinning technique with no further annealing. All the other metallic glasses were made in bulk. Ingots of the alloys were prepared by arc melting a mixture of pure metal elements in a titanium-gettered argon atmosphere, followed by suction casting into copper molds to form bulk metallic glass (BMG) solid samples [4]. For each metallic glass an IAS Round Cell model of  $10^6$  spheres has been generated, and the coordinates of the sphere centers have been used to predict X-ray scattering by a program based on Eq. (1).

## 2.1.2. Covalent glasses

Plate glass or powder samples from pure silica have been prepared by grinding. Amorphous polyethylene has been researched extensively in the past and some relevant data from published literature has been used in this work.

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**Table 1**  
Range of studied glass samples and their characteristics.

Class type	Glass composition [%]	Assumed atomic radii [pm]	$2\theta$ exp. [ $\pm 0.5^\circ$ ]	$2\theta$ IAS [ $\pm 0.3^\circ$ ]	$2\theta$ Ehr
Metallic	Ni68–Nb32	[125] [146]	42°	41.9°	42.1°
	Mg65–Cu25–Gd10	[146] [128] [175]	37.0°	37.3°	38.3°
	Zr50–Cu50	[159] [128]	37.5°	37.8°	38.5°
	Zr40–Cu60	[159] [128]	38.7°	38.5°	39.5°
	Zr36–Cu64	[159] [128]	38.8°	38.5°	39.7°
	Zr45–Cu50–Al5	[159] [128] [143]	36.6°	36.7°	37.3°
	Zr45–Cu50–Ti5	[159] [128] [147]	36.2°	36.4°	37.3°
	Zr35–Cu30–Ti35	[159] [128] [147]	36.5°	37.0°	38.0°
	Zr52–Ti5–Cu18–Ni15–Al10	[159] [147] [128][125] [143]	37.2°	37.8°	37.8°
	Zr43–Ti14–Cu12–Ni10–Be23	[159] [147] [128][125] [112]	38.2°	38.4°	38.8°
Mixed	Fe40–Ni40–B20	[126] [125] [85]	45.0°	42.0°	47.4°
	Fe80–B20	[126] [85]	37.6°	37.6°	37.3°
	Fe81–B14–Si3–C2	[126] [85] [105] [77]	37.8°	25°	47.1°
	Ni78–Si8–B14	[125] [105] [85]	46.0°	42.0°	47.4°
Covalent	SiO <sub>2</sub> , silica glass powder	[105] [55]	21.2°	21°	Warren
	CH <sub>2</sub> a-polyethylene	[71] [38]	18°	18°	Warren
Chalcog.	Ge33–As33–Se34, bulk	[246] [243] [238]	29°	44°	22.6°
	Ge33–As33–Se34, annealed	[246] [243] [238]	29°	44°	22.6°

### 2.1.3. Chalcogenide glasses

Commercially available AMTIR-1 in bulk solid form, with composition Ge33–As12–Se55, was used as a thermal evaporation source. Films of thickness several micrometers were evaporated on both Si substrates and X-ray transparent Kapton, the former first coated with a release layer. Some samples were annealed at 350 °C to induce structural relaxation. As-deposited and thermally annealed films were separated from the Si substrate by dissolution of the release layer, finely crushed and loaded into capillaries. For extended X-ray absorption fine structure (EXAFS) measurements, films (on Kapton) were stacked together to achieve  $\mu t \approx 1$  at each of the Ge, As and Se K-edges, where  $\mu$  is the mass attenuation coefficient and  $t$  is the layer thickness.

### 2.2. Measurement of X-ray scattering and EXAFS

For the metallic glasses diffractograms were obtained with synchrotron wide angle X-ray scattering (WAXS) performed in transmission geometry at the Australian Synchrotron (X-ray energies of 12 and 20 keV). A LaB<sub>6</sub> standard was used for spatial calibration. Alternatively, a high resolution Phillips PANalytical X'Pert PRO MRD system was used in powder diffraction configuration with Cu-K $\alpha$  radiation (8.02 keV, 30 kV, 10 mA,  $2\theta$  varying from 10° to 80°). A separate set of measurements has been carried out on the chalcogenide glasses, using the Phillips PANalytical system with Co-K $\alpha$  radiation, 40 kV, 30 mA, graphite monochromator, with 20 s/step rate of data acquisition.

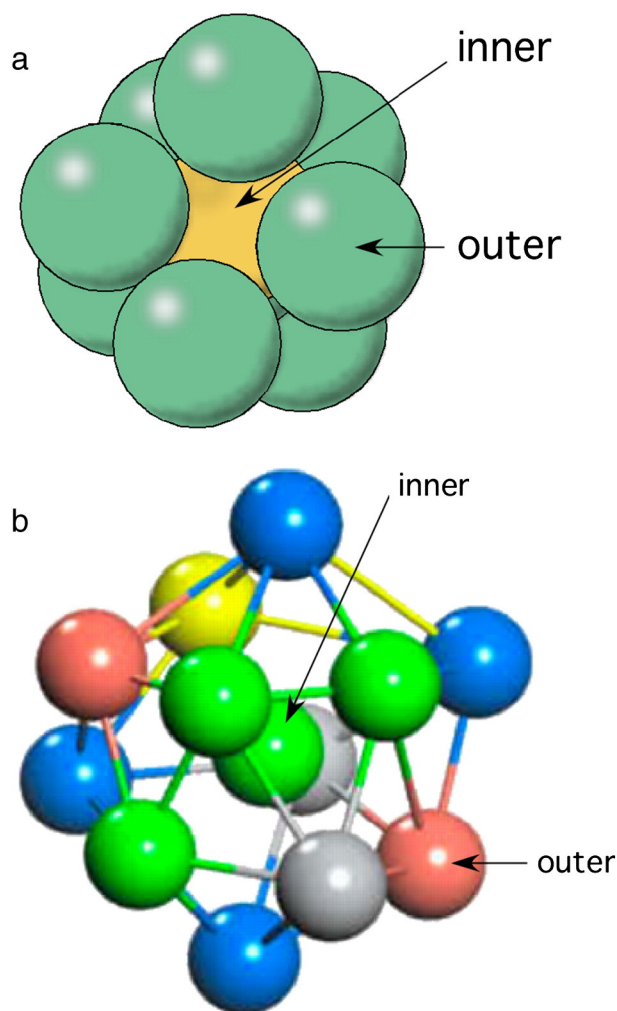
EXAFS measurements were carried out in transmission mode at the Australian Synchrotron with the samples maintained at a temperature of 12 K to minimize thermal vibrations. To extract the structural parameters (coordination number, bond length and Debye–Waller factor) about the Ge, As and Se absorbers, EXAFS spectra were analyzed with the IFFEFIT code [5].

For covalent glasses the measured scattering patterns were obtained on a Siemens D5000 X-ray diffractometer, using collimated Cu-K $\alpha$  radiation and  $2\theta$  goniometer. Typical conditions were: 30 kV, 10 mA, 1 hour scanning, with angle  $2\theta$  varying from 10° to 80°. In some cases the experimental X-ray scattering traces have been obtained from other sources as indicated in the text.

## 3. Models of amorphous structure

### 3.1. Ideal amorphous solid (IAS)

The ideal amorphous solid (IAS) is a geometric model of atomic arrangement composed of contacting hard spheres packed in a perfectly



**Fig. 1.** (a) An irregular cluster from IAS model with inner sphere surrounded by outer spheres all touching the inner sphere; (b) a cluster from molecular dynamics simulations showing central (inner) atom surrounded by a number of outer atoms with uneven distances to the center [10]. In molecular dynamics representation of atoms is symbolic since the size of atoms is undefined.

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