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An efficient method to generate amorphous structures based on local geometry

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ABSTRACT

We propose an efficient method that generates amorphous structures based on information on the short-range order such as bond lengths and coordination numbers. The base amorphous structure is constructed in the "seed-and-coordinate" style, which conforms to the given short-range order. The structure is further annealed to relax the atomic bonding and establish the medium-range order. The computational cost of the proposed method is less than 10% of the standard melt-quench approach. We combine this method with *ab initio* calculations and generate amorphous structures for various materials such as *a*-Si, *a*-SiO_2, *a*-$Ge_2Sb_2Te_5$, and *a*-$InGaZnO_4$. The obtained structures are close to those from melt-quench simulations in terms of atomic and electronic structures. To substantiate the computational efficiency, we generate *a*-GeSe with 512 atoms in the unit cell. The large cell allows us for identifying the mobility edge clearly.

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1. Introduction

In many applications, crystalline materials are usually favored owing to good stability and well-defined physical properties. Nevertheless, the metastability or non-equilibrium state of amorphous structures also leads to useful applications. For example, the structural flexibility of amorphous SiO_2 (*a*-SiO_2) has been used for shaping glasses in various forms since long time ago. More recently, the hydrogenated amorphous Si (*a*-Si:H) has been widely used in electronic devices that require a large-area growth of electrically active materials [1,2]. Furthermore, *a*-Si is now challenged by another amorphous material, In–Ga–Zn–O, which shows larger electron mobilities [3–8] Lastly, the phase-change memory that exploits reversible switching between amorphous and crystalline phases of chalcogenides such as $Ge_2Sb_2Te_5$, is a leading contender for the next-generation nonvolatile memory [9–12].

The conventional spectroscopic analysis is seriously limited when studying amorphous materials due to the lack of lattice periodicity. This made the computational modeling on amorphous phases very informative because it can provide structural and electronic data that are not accessible in experiment. However, in contrast to the crystalline phase, detailed atomic arrangements in amorphous materials are not available prior to the simulation. This is a serious hurdle against the atomistic simulation, in which the knowledge on atomic positions is a prerequisite. So far, several methods have been developed to generate amorphous structures theoretically [13,14]. One example is the bond-switching method [15], which was successfully applied to generate the continuous random network (CRN) structures for sp^3 semiconductors like *a*-Si, *a*-Ge, and amorphous diamond. However, this method is rather limited to CRN structures with strong covalent characters. In addition, the ideal CRN model is sometimes inconsistent with experiment [16]. Activation-relaxation technique (ART) is another well-established approach [17–20]. In this case, the atomic positions are activated from the local energy minimum to an adjacent saddle point and then relaxed to another local minimum following the downhill paths in the energy landscape.

Perhaps, the most favored method at present is the melt-quench (MQ) approach based on molecular dynamics (MD) simulation. This mimics the actual MQ process in experiment. If a reliable force field or interatomic potential is available, the classical MD simulation will be able to produce amorphous structures very efficiently [21–24]. However, the classical force field is highly limited in the choice of material classes. Such a limitation does not exist in the *ab initio* MD simulation based on the density functional theory (DFT) in which the atomic forces are obtained through quantum-mechanical description of electronic states. Therefore, the MQ method based on DFT-MD has been applied to generating amorphous structures of a wide range of materials in a reliable way [25–33]. However, the computational cost of DFT-MD is so expensive that supercells comprise only about 100–200 atoms in most

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cases. With a typical size of modern computational clusters, it takes weeks of computational time to obtain one amorphous structure including 100–200 atoms in the unit cell. If one employs a relatively big supercell with >500 atoms [34] or generates multiple amorphous structures for statistical average [35], the computational time easily scales up to several months.

In this work, we propose a new computational scheme to produce the amorphous structure, which utilizes DFT but is faster than the conventional DFT-based MQ method by more than ten folds. Our method starts with the premise that if the detailed information on the short-range order is available beforehand, one can quickly generate a reasonable amorphous model in one-shot and only short time annealing would be necessary to refine the structure. The reference local order could be obtained experimentally through the reverse Monte Carlo method [36,37] or theoretically from the MQ amorphous structure. In the latter case, we note that the short-range order is well addressed in relatively small supercells consisting of less than 100 atoms. Therefore, the basic strategy of the present method is that one performs the MQ simulation on a small cell and extracts the detailed local order, which is used in generating amorphous structures on a large scale or multiple times for the statistical average, within a reasonable computational time.

The paper is organized as follows. In Section 2.1, we define various parameters required for generating the structure. The detailed algorithm (so called “seed-coordinate-anneal”) is put forward in Section 2.2. Test calculations on various amorphous materials (Si, SiO_2, $Ge_2Sb_2Te_5$, and $InGaZnO_4$) are presented in Section 3 with comparison with reference structures and experiment. Finally, we summarize and conclude in Section 4.

2. Computational method

2.1. Input parameters defining the local geometry

In the following, we assume that the detailed information on the short-range order is available from either the reference calculations involving a small number of atoms or reverse Monte Carlo analysis on the experimental diffraction data. We then define variables specifying the local geometry of the reference structures. The basic bonding information is determined by the atom-resolved radial distribution function (RDF). For easy understanding, the procedure will be described with the specific example of *a*-GeSe as shown in Fig. 1. The topmost figures in Fig. 1(a) and (b) are partial RDFs around Ge and Se atoms with the first-dip position indicated by the dashed line. If the fully resolved RDF between *X* and *Y* (*X*, *Y* = Ge or Se) has the first peak within the dashed line, it is assumed that bonds are formed between *X* and *Y*. According to this criterion, bonds are not formed between Se atoms as the first peak in the bottom picture of Fig. 1(b) lies outside the dashed line. The bonding ranges for Ge–Ge, Ge–Se and Se–Ge pairs are parameterized by $d^{X}_{\mathrm{min}}(Y)$ and $d^{X}_{\mathrm{max}}(Y)$, where $d^{X}_{\mathrm{max}}(Y)$ is set to the first-dip position in partial RDF between *X* and *Y* while $d^{X}_{\mathrm{min}}(Y)$ corresponds to a point where the partial RDF reaches 0.01 (see Fig. 1). By symmetry, $d^{X}_{\mathrm{min\,(max)}}(Y) = d^{Y}_{\mathrm{min\,(max)}}(X)$.

With the determined bonding ranges, one can obtain the probability of atom type *Y* of bonded neighbors, $P^{X}_{\mathrm{type}}(Y)$, and that of coordination number (CN), $P^{X}_{\mathrm{CN}}(N)$, around a certain type *X*. The results for *a*-GeSe are shown in Fig. 2. Obviously, $\sum_{Y} P^{X}_{\mathrm{type}}(Y) = \sum_{N} P^{X}_{\mathrm{CN}}(N) = 1$ for every *X*.

2.2. Generation of amorphous structures

With $d^{X}_{\mathrm{min(max)}}(Y)$, $P^{X}_{\mathrm{type}}(Y)$, and $P^{X}_{\mathrm{CN}}(N)$ extracted from the reference configurations, we proceed on how to distribute atoms in a way that is consistent with these input parameters. The procedure is somewhat involved, and for the easy understanding, the flow chart of the schematic procedure is presented in Fig. 3. Broadly speaking, the present method consists of two steps in seed-and-coordinate style. In the first step (the upper box in Fig. 3), a “seed” atom (indexed with *i*) is introduced into the simulation box or it is simply selected if already exists in the box. Then, the seed atom is coordinated with other atoms (indexed with *j*) in a way consistent with the input parameters. This second step corresponds to the “coordination” mode (the lower box in Fig. 3). In the below, we provide further details for each step.

(i) *Seeding step:* if $i = j$, this atom is not in the simulation box and should be first put inside the box. This new seed atom is assigned the atom type $\alpha(i)$ that is randomly chosen within the given stoichiometry. The CN of this atom ($\mathrm{CN}_0(i)$) is selected on the basis of $P^{\alpha(i)}_{\mathrm{CN}}$ in a probabilistic way. Then, this *i*th atom is put at a random position $\mathbf{R}(i)$ that is sufficiently away from the other atoms present in the box,

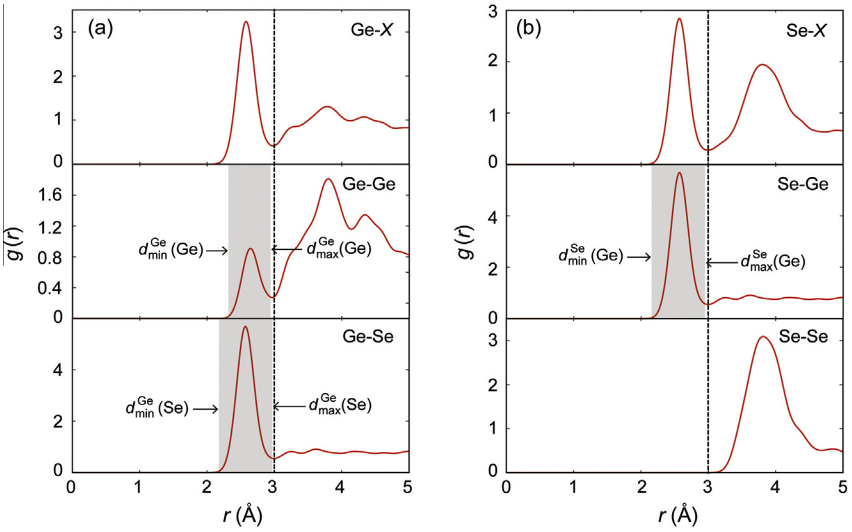

Fig. 1. The atom-resolved radial distribution function, $g(r)$, of (a) Ge and (b) Se in *a*-GeSe. The dashed line and shaded areas indicate the cutoff distance for defining the atomic bond and the ranges of the bond lengths between atom types, respectively.

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