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Review

Long-range n-body potential and applied to atomistic modeling the formation of ternary metallic glasses

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ABSTRACT

In this review article, a brief summary is firstly presented concerning the currently used n-body potentials for the metal systems, i.e. the second moment approximation of tight-binding, Finnis-Sinclair potentials and embedded atom method. Secondly, a long-range n-body potential is proposed and proven to be not only applicable for the three major crystalline structured, i.e. bcc, fcc and hcp, metals and their alloys, but also able to distinguish the energy differences between fcc and hcp structures of the metals. Furthermore, both the energy and force reproduced by the proposed potential could go smoothly to zero at cutoff distance, without appearing some unphysical phenomena frequently observed in atomistic simulations with other n-body potentials. Thirdly, for some selected ternary metal systems, the long-range n-body potentials are constructed with the aid of *ab initio* calculations and applied in molecular dynamics and Monte Carlo simulations to study the metallic glass formation. The simulation results not only clarified that the underlying physics of the metallic glass formation is the crystalline lattice collapsing of the solid solution when the solute concentration exceeds the critical solid solubility, but also predicted, for a ternary system, a quantitative composition region within which the metallic glass formation is energetically favored. Fourthly, the energy difference between the solid solution and the amorphous counterpart is defined as the driving force for the crystal-to-amorphous transition, and the amount of the driving force could thus be considered as a comparative measure of the glass-forming ability of the amorphous alloy. It follows that in a ternary metal system, the largest driving force could be correlated to the optimized composition, of which the metallic glass is the most stable or easiest one to be produced in practice. It turns out that the predictions directly from the constructed potential through atomistic simulations are well compatible with the experimental observations reported so far in the literature, leading firm support to the relevance of the predicted glass-forming regions and the optimized compositions of the respective ternary metal systems, as well as to the validity of the proposed long-range n-body potential.

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1. Introduction

In 1954, Buckel and Hilsch found that condensation of metal vapor onto a cooled substrate could result in the formation of an amorphous alloy [1,2]. A few years later, Duwez et al. obtained an Au–Si amorphous alloy using a technique named liquid melt quenching [3,4]. These pioneering works explored a new category of metallic materials, i.e. amorphous alloys or later named metallic glasses. The metallic glasses, with a non-crystalline structure, differ significantly from those widely used crystalline alloys, such as steels, aluminum alloys, titanium alloys [5–7]. Since the 1960s, liquid melt quenching has been extensively used to produce a large number of metallic glasses in binary, ternary as well as multicomponent metal systems [8–10]. In the early stage, metallic glasses obtained by liquid melt quenching were mostly thin films or foils. Since the late 1980s, researchers have found a number of ternary and multicomponent metal systems, in which liquid melt quenching could readily produce the so-called bulk metallic glasses with a geometrical size of up to a magnitude of centimeter [11–16].

In the field of metallic glasses, one of the fundamental scientific issues is the so-called glass-forming ability (GFA), which reflects the easiness or difficulty of the metallic glass formation. This issue has been widely discussed generally in two ways [17–19]. First, consideration is given to the specific alloy and metallic glass

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formation is considered as the frustration of the crystallization, or the consequence of possibly enhanced stability of a liquid-like state down to low temperatures. Second, consideration is given to, for example, a specific binary metal system and in this situation, the glass-forming range (GFR) is used to indicate an alloy composition range within which the metallic glasses could be obtained in the system by certain non-equilibrium materials producing techniques. In general, the issues of the GFA/GFR could be approached either by experiment or atomistic simulation [13,20].

An interatomic potential of a system is used to describe the atomic interactions of the system and, in principle, if the potential is known, then most of the properties or behaviors of the system could be obtained directly from the potential, e.g. through computation and simulation. In this sense, the GFA/GFR of the system could surely be derived from the potential. However, the ideal potential, which could commonly be valid, is not available. In fact, some approximations and assumptions have to be made to the interatomic interaction. In the very early stage, Mie has proposed a phenomeno-logical model to describe the interatomic interactions [21]. In Mie's model, the interatomic interactions could be decomposed into two terms: one represents the repulsion and the other the attraction between the two atoms. Based on Mie's work, Lennard and Jones proposed in 1925 a simple form of the pair potential, i.e. the Lennard–Jones, or L–J, 6–12 potential [22]. A few years later, some variants of the L–J potential, such as the Morse [23] and Born–Mayer potentials [24] were proposed. The unique feature of these potentials is their convenient mathematical forms to describe the properties of real materials. Because these pair potentials do not incorporate the many-body effect, there are some intrinsic drawbacks, e.g. the dilemma of the cohesive energy and vacancy formation energy. For a pair potential, if the cohesive energy is correctly described, then the vacancy formation energy is not, and vice versa. Besides, for cubic metals, the Cauchy pressures derived from the pair potentials are always zero, yet the real Cauchy pressures are usually non-zero [25,26].

In the 1980s, based on the concept of the local electron density, a significant progress was made by developing the so-called n-body potentials [27]. The main physical idea of the n-body potentials is that the bonds would become weaker when the local environment becomes more crowded. Consequently, a plot of the cohesive energy as a function of coordination should not decrease linearly [28]. This means that the cohesive energy of an atom is largely governed by the local atomic configuration at the site where the atom is located. In the literature, several n-body potentials of different forms have been proposed for the transition metals and their alloys. The most widely used ones are the second-moment approximation of tight-binding (TB-SMA) potential [29–35], the Finnis-Sinclair (F-S) potential [36], the embedded-atom method (EAM) potential and their various modifications [37–43]. In the past decades, a great efforts have been made to improve the performance of these n-body potentials. For example, Guellil and Adams proposed a polynomial truncation function for the electron density in the EAM potential [44]. Nonetheless, because a logarithmic function is adopted for the embedding function, the potential energy goes to infinity instead of converging to zero when the electron density approaches to zero, probably resulting in some unphysical phenomena in atomistic simulations [39,45]. Wadley et al. proposed a segmented embedding function by introducing about 20 parameters in the EAM potential [46]. Sutton and Chen proposed a Long-range F-S potential for fcc structured metals [47,48].

In recent years, by combining the unique features of the TB-SMA and F-S potentials, Li, Dai and Dai proposed a long-range n-body potential (abbreviated as the LDD potential for convenience). The proposed LDD potential has been proven to be suitable for bcc, fcc

and hcp structured metals and their alloys [45,49–51]. The LDD potential also solved the problem related to the appearance of some unphysical phenomena that occurred with other n-body potentials, because the energy and force derived from the LDD potential both smoothly converge to zero at the cutoff distance in the simulations. Furthermore, the LDD potential is also able to well distinguish energy difference between the fcc and hcp structures of the transition metals [52–54]. In addition, the LDD potential has also been proven to be able to reproduce the static as well as dynamic physical properties of the transition metals and alloys.

In the present review, a brief discussion is first presented concerning the currently available n-body potentials that are widely used for the metal systems. Second, the newly proposed LDD potential is introduced in detail, including its form, unique feature as well as the performance in reproducing the static and dynamic physical properties of metals and alloys. Third, we applied the constructed LDD potential to model the formation of ternary metallic glasses, clarifying the underlying physics of metallic glass formation and to derive the related GFA/GFR for some representative ternary transition metal systems through atomistic simulations. Finally, a few concluding remarks and prospects are given to end the present review.

2. Typical n-body potentials of the transition metal systems

For transition metal systems, several n-body potentials have been developed and are currently used in the field of computational materials science. These potentials share a similar form, yet frequently result in rather different parameterization for the same material. In many cases, the researchers could make a guess of the functions and fit the parameters to the experimental data, whereas for some special cases, the researchers could derive the functions and parameters by fitting the data acquired from the *ab initio* calculations. In general, these n-body potentials still belong to empirical approaches and hence have some limitations and drawbacks. In this section, a brief discussion is firstly presented concerning the typical n-body potentials used in transition metal systems, i.e. the EAM, TB-SMA and F-S potentials.

2.1. Embedded atom method

According to Daw and Bask [37,41,42], the basic principle of the EAM is that each atom can be viewed as an impurity embedded in a host created by its neighboring atoms and the energy of a system consisting of N atoms can be expressed by

$$E_{\text{total}} = \frac{1}{2} \sum_{j \neq i} \phi_{ij}(r_{ij}) + \sum_i F_i(\rho_i), \qquad (1)$$

where r_{ij} is the distance between atoms i and j and $\phi_{ij}(r_{ij})$ is the pair interaction accounting for the electrostatic repulsion between the atoms. The local electron density, ρ_i, is the electron density of the host at the site of atom i. The embedding energy of atom i, $F_i(\rho_i)$, incorporating the many-body contributions, reflects the interaction between the embedded atom and the background electron gas. The local electron density may vary from site to site, depending on the local atomic configurations, and can be approximated by the superposition of the contributions from the neighboring atoms, i.e.

$$\rho_i = \sum_{j \neq i} \psi_j(r_{ij}), \qquad (2)$$

where $\psi_j(r_{ij})$ is the contribution of the neighboring atom j to the local electron density ρ_i. It can also be considered as the electron density of the atom j at the site of the atom i. The embedding