Physica B 427 (2013) 110–117

Contents lists available at SciVerse ScienceDirect

Physica B

journal homepage: www.elsevier.com/locate/physb

Atomistic simulation of site preference, Curie temperature and lattice vibration of $ZrT_{12-x}M_x$ (T=Fe, Co; M=Al, Ga)

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ARTICLE INFO

Article history:
Received 13 April 2013
Received in revised form
4 July 2013
Accepted 7 July 2013
Available online 11 July 2013

Keywords:
Interatomic potentials
Magnetic properties
Phase stability
Site occupancy

ABSTRACT

A series of lattice inversion pair potentials are used to evaluate the phase stability and site preference of $ZrT_{12-x}M_x$ (T=Co, Fe; M=Al, Ga; x=6.0, 6.5, 7.0) compounds. The calculated preferential occupation site of the M atom is found to be the 8i site, which is in good agreement with experimental results. And the calculated lattice constants coincide quite well with experimental values. Further, the energy DOS of the relaxed structures are calculated and the variation in Curie temperature is explained qualitatively by the spin-fluctuation theory. Meanwhile, the phonon densities of states and Debye temperature are evaluated for the $ZrT_{12-x}M_x$ compounds.

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1. Introduction

Due to their high Curie temperature and large magnetic moment, among the rare earth–transition metal compounds, the study of $RT_{12-x}M_x$ has been one of the major topics, where M represents a stabilizing element [1–8].

Recently, intermetallic compounds which combine 3d transition metals carrying large magnetic moment with heavier 4d or 5d transition metals have attracted considerable attention. As the heavy transition metals replace the rare-earths, the eliminating of rare-earth elements from devices that rely on permanent magnets will be realized. Research shows that the formation of isostructure analogs with 4d or 5d metal instead of rare-earth elements is difficult to achieve, probably because the size of rare-earth is much larger than that of the 4d or 5d metals [9]. Up to present, some kinds of intermetallic compounds in $ThMn_{12}$ structure-type have been found [10–15].

There is a previous report of magnetism for $RFe_{12-x}Al_x$, where R is a rare-earth element, include compounds with A=Sc, Y, La, Ce, and Lu [16]. Lately, an investigation of crystal structure and magnetic properties of compounds $ZrFe_{12-x}Al_x$ has been reported [17].

The purpose of this work is to provide a description of some properties such as site preference and Debye temperature of these $ZrT_{12-x}M_x$ (where T=Co, Fe; M=Al, Ga) compounds.

2. Methodology

2.1. Brief introduction of the inverted potentials

In the mid-1990s, based on Möbius Inversion in number theory, Chen et al. presented a new principle [18–23], which developed into the lattice inversion technique to obtain pair potentials. A brief introduction to this lattice inversion method is given below.

Suppose that the crystal cohesive energy can be expressed as the sum of interatomic pair potentials

$$E(x)=\frac{1}{2}\sum_{r_i\neq 0}\Phi(\boldsymbol{r}_i)=\frac{1}{2}\sum_{n=1}^{\infty}r_0(n)\Phi[b_0(n)x] \tag{1}$$

where x is the nearest-neighbor distance, $\boldsymbol{r}_i$ is the position vector of the ith atom, $b_0(n)x$ is the nth coordination number. We extend the series $\{b_0(n)\}$ into a multiplicative semigroup $\{b(n)\}$, such that, for any two integers m and n, there always exists an integer k such that

$$b(k)=b(m)b(n) \tag{2}$$

Eq. (1) can then be rewritten as

$$E(x)=\frac{1}{2}\sum_{n=1}^{\infty}r(n)\Phi[b(n)x] \tag{3}$$

where

$$r(n)=\begin{cases} r_0(b_0^{-1}[b(n)]) & \text{if}\quad b(n)\in\{b_0(n)\} \\ 0 & \text{if}\quad b(n)\notin\{b_0(n)\} \end{cases} \tag{4}$$

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http://dx.doi.org/10.1016/j.physb.2013.07.004

Thus the pair potential $\Phi(x)$ can be written as

$$\Phi(x) = 2\sum_{n=1}^{\infty} I(n)E[b(n)x] \tag{5}$$

where $I(n)$ is determined by

$$\sum_{b(n)|b(k)} I(n)r\left(b^{-1}\left[\frac{b(k)}{b(n)}\right]\right) = \delta_{k,1} \quad (k=1,\ 2, \ldots\infty) \tag{6}$$

$I(n)$ is related only to crystal geometrical structure, not to concrete element category, *i.e.* $I(n)$ is uniquely determined by $\{r_0(n)\}$ and $\{b_0(n)\}$. Thus the inverted potentials can be obtained from the known cohesive energy function.

In principle, all the potentials can be obtained from cohesive energy curves. In our work, the cohesive energy has been acquired by *ab initio* calculation, and all the potentials are extracted by using lattice inversion method.

In this work, we considered the virtual bcc Zr as B_2 structure with two simple cubic sub-lattices Zr_1 and Zr_2. Thus we calculate the partially cohesive energy as

$$\begin{aligned} E(x) &= E_{Zr}^{bcc}(x) - E_{Zr_1}^{sc}(x) - E_{Zr_2}^{sc}(x) \\ &= \sum_{i,j,k\neq 0}^{\infty} \Phi_{\text{Zr–Zr}}\left(\sqrt{\frac{4}{3}\left[\left(i-\frac{1}{2}\right)^2 + \left(j-\frac{1}{2}\right)^2 + \left(k-\frac{1}{2}\right)^2\right]}x\right) \end{aligned} \tag{7}$$

where x is the nearest neighbor distance in the bcc structure, $E_{Zr}^{bcc}(x)$ represents the total energy of Zr with bcc structure, $E_{Zr_1}^{sc}(x)$ and $E_{Zr_2}^{sc}(x)$ is the total energy of Zr with simple cubic structure. Now $E(x)$ automatically becomes the cohesive energy function of one Zr_1 atom with all the Zr_2 atoms. Here the Zr_2 atoms form a simple cubic structure, and only one Zr_1 atom is located at the center of each cube. Then the pair potentials $\Phi_{\text{Zr–Zr}}(x)$ between identical atoms can be obtained directly by using Chen's lattice inversion technique.

The *ab initio* calculation of total energy curve related to $\Phi_{\text{Zr–Fe}}(x)$ is hard to perform. The lattice constant module constant and total energy of Zr_3Fe with $L1_2$ structure near equilibrium position has been calculated. Then the partial cohesive energy of Zr–Fe can be obtained by subtracting the contribution of Zr–Zr and Fe–Fe in the structure respectively. Subsequently, the $\Phi_{\text{Zr–Fe}}(x)$ can be deduced from lattice inversion method.

The obtained pair potential can be described by Morse function

$$\varphi(x) = D_0\left(e^{-\gamma(x/R_0-1)} - 2e^{-(\gamma/2)(x/R_0-1)}\right) \tag{8}$$

where x indicates the distance between two atoms, D_0, γ and R_0 are the parameters. The calculated potential parameters used in this work are given in Table 1. For the readers' convenience, potentials in the system of $ZrT_{12-x}M_x$ are shown in Fig. 1.

Table 1
Parameters of the inverted potentials in this work.

Potential types	R_0 (Å)	D_0 (eV)	γ
$\Phi_{\text{Zr–Zr}}$	3.6209	0.7847	8.9154
$\Phi_{\text{Zr–Fe}}$	3.0916	0.8251	8.4045
$\Phi_{\text{Zr–Co}}$	3.0852	0.7838	8.4076
$\Phi_{\text{Zr–Al}}$	3.3194	0.5697	8.7459
$\Phi_{\text{Zr–Ga}}$	3.2454	0.5300	8.6985
$\Phi_{\text{Fe–Fe}}$	2.7361	0.7640	8.7529
$\Phi_{\text{Fe–Al}}$	2.7405	0.6462	9.0968
$\Phi_{\text{Fe–Ga}}$	2.6882	0.5690	9.6878
$\Phi_{\text{Co–Co}}$	2.7087	0.6766	8.9030
$\Phi_{\text{Co–Al}}$	2.6970	0.6477	9.1129
$\Phi_{\text{Co–Ga}}$	2.6609	0.5564	9.6663
$\Phi_{\text{Al–Al}}$	3.0059	0.4232	8.9191
$\Phi_{\text{Ga–Ga}}$	3.0857	0.3706	8.7343

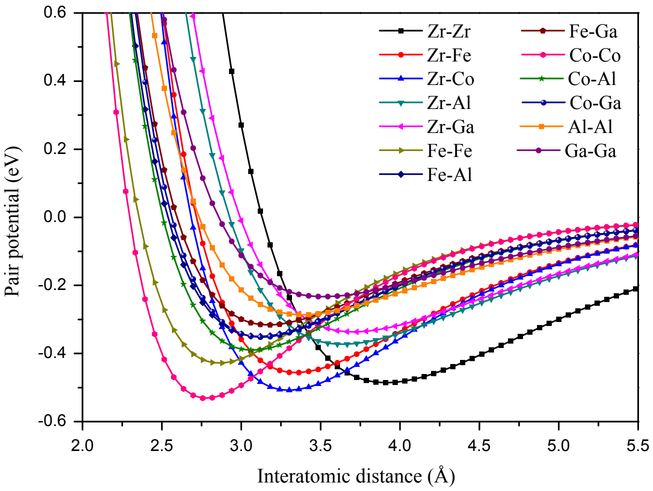

Fig. 1. Potentials in the system of $ZrT_{12-x}M_x$.

It is true that the intermetallic pair potential and the cohesive energy are related to the various neighbor-atom configurations for multielement systems, thus the pair potential and the cohesive energy will not be the same after the M atoms (M=Al, Ga) substitution for T atoms (T=Fe, Co) in ZrT_{12} compounds. But most of the atomistic simulation methods for complex systems are unfortunately quite complicated and time consuming especially when there are too many adjustment parameters in the calculations. In our semiempirical simulation based on lattice-inversion method, as a practical approximation, the intermetallic pair potential obtained from simple structures such as B_2 and $L1_2$ are assumed to be able to transfer to other complex intermetallic structures without any adjustment parameter. The validity of the transferability of intermetallic pair potentials has been supported by our previous work [24–29]. It has been established that the inverted pair potentials are quite reliable for calculations of the cohesive energy and the structural properties of intermetallics. Practically we can simulate the multielement systems with large cell, *e.g.* $((ZrT_{12-x}M_x)_2)_{4\times4\times4}$ (T=Fe, Co; M=Al, Ga) in this paper, and take the relaxation effect into account, avoiding the difficulties of quantum mechanics encountered when treating a large cell.

2.2. Calculation method for phonon DOS and the Debye temperature

In the harmonic approximation of the lattice dynamics, the secular equation of the lattice vibrations can be written as

$$\det\left|D_{\alpha\beta,k\mu}(\boldsymbol{q}) - \omega^2\delta_{\alpha\beta}\delta_{k\mu}\right| = 0 \tag{9}$$

where ω is the angular frequency and $\boldsymbol{q}$ is the wave vector. $D_{\alpha\beta,k\mu}(\boldsymbol{q})$ is the wave vector. $D_{\alpha\beta,k\mu}(\boldsymbol{q})$ is a dynamical matrix and its elements are written as

$$D_{\alpha\beta,k\mu} = (M_k M_\mu)^{-1/2}\sum_{R_i-R_j}\Phi_{\alpha\beta,ik,j\mu}\exp[-i\boldsymbol{q}(\boldsymbol{R}_i-\boldsymbol{R}_j)] \tag{10}$$

where $\Phi_{\alpha\beta,\ ik,\ j\mu}$ is the force constant, $\boldsymbol{R}_i$, $\boldsymbol{R}_j$ are positions in the ith and jth cell, and k, μ represent different atoms in the same cell. Using the interatomic pair potential, the force constant can be expressed as

$$\Phi_{\alpha\beta,ik,j\mu} = \begin{cases} \frac{1}{2}\left[\frac{R_\alpha R_\beta}{R^3}\Phi'_{k,\mu}(R) - \frac{R_\alpha R_\beta}{R^2}\Phi''_{k,\mu}(R) - \frac{1}{R}\Phi'_{k,\mu}(R)\delta_{\alpha\beta}\right] & \text{if} \quad R\neq 0 \\ -\sum\limits_{k\neq\mu}\Phi_{\alpha\beta,i\mu,ik} - \sum\limits_{i\neq j}\sum\limits_{k}\Phi_{\alpha\beta,ik,i\mu} & \text{if} \quad R=0 \end{cases} \tag{11}$$

where $R=|\boldsymbol{R}|=|\boldsymbol{R}_i-\boldsymbol{R}_j+\boldsymbol{r}_k-\boldsymbol{r}_\mu|$, $\boldsymbol{r}_k$, $\boldsymbol{r}_\mu$ are the relative positions of the different atoms in the same cell, $\Phi_{k,\ \mu}(R)$ is the pair potential between the kth and μth atoms, and Φ' and Φ'' are the first and second derivative of Φ, respectively.

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