Atomistic study on the structure and Curie temperature for Nd$_2$Fe$_{17-x}$Cr$_x$

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Abstract

The structural stability of ternary compounds Nd$_2$Fe$_{17-x}$Cr$_x$ is evaluated by using a series of quasi-ab initio interatomic potentials. The results show that the substitution of Cr atoms for Fe almost does not change the crystal symmetry significantly and the calculated structural parameters of Nd$_2$Fe$_{17-x}$Cr$_x$ correspond well to experimental data. The site preference of Cr atom is further evaluated and the order is given as 6c, 18f, 18h and 9d, which are close to experimental results. Moreover, the relaxed structure together with the site preference of Cr atoms could give some simple explanation on the variation behavior of Curie temperature. All results indicate that the present calculated potentials are effective for studying some structural properties of this kind of intermetallics. © 2002 Elsevier Science B.V. All rights reserved.

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Keywords: Structure determination; Site preference; Interatomic potentials; Lattice inversion, Curie temperature

1. Introduction

As permanent magnets, rare-earth iron-rich compounds are widely used in many fields [1]. Many rare-earth compounds such as R$_2$(Fe,T)$_{17}$ [2,3], R(Fe,T)$_{12}$ [4], R$_3$(Fe,T)$_{29}$ [1,5,6] received extensive attention. In these compounds, the applications of R$_2$Fe$_{17}$ are restricted by their lower Curie temperatures and poor magnetocrystalline anisotropy at room temperature. Therefore, many efforts have been made by either substituting ternary element or introducing interstitial atom to improve the magnetic properties [7,8]. In theory, there are few works about rare-earth compounds with 4f electron. Ab-initio calculations by Sabiryanov indicated that the nitrogenation of Sm$_2$Fe$_{17}$ more than doubles the Curie temperature ($T_c$), but lowers $T_c$ of Sm$_2$Co$_{17}$ [9]. They also calculated the exchange parameters ($J_{ij}$) of Sm$_2$Fe$_{16}$A (A = Ga, Si) by self-consistent spin-polarized LMTO, and simulated the Curie temperature using Monte Carlo method [10]. Phenomenological studies have been performed for R$_2$(Fe,T)$_{17}$ [11,12] on the ternary substitution mechanism, in which three term contributions including chemical term, electronic term and elastic term are considered. Recently, Chen et al.
have performed a theoretical study on the phase
stability, site preference and lattice parameters for
Gd(Fe, Ti)_{12} [13], Sm(Fe, T)_{12} [14], Nd(Fe, Ti)_{12}N_x,
and Nd_{2}Fe_{29–x}T_x [15], suggesting the validity of
ab initio potentials on the structure simulation of a
rare-earth compound.

In the present work, a series of heuristic
interatomic potentials is used to systematically
simulate the structural properties of Nd_{2}Fe_{17–x}Cr_x
system. The computation for lattice inversion
and structure selection for quasi-ab initio procedure are discussed in Section 2. The calculated
results for a variety of structural properties are shown in Sections 3 and 4 with comparison to
experimental data. The good agreement between
calculated and experiment indicates the adapt-
ability of the quasi-ab initio interatomic (pair)
potentials on rare-earth iron-based intermetallics.
As a consequence, the variation of atomic sites
with content of Cr atoms is evaluated for
explaining the behavior of Curie temperature.
The conclusion and discussion are presented in
Section 5.

2. Calculation method

In the beginning of 1980s, Carlsson et al. [16]
reported that a pair potential could be obtained by
ab initio cohesive energy curve, but the expression
is indigestible for including infinite summations,
each of which contains infinite terms. Different
from above method, Chen’s lattice inversion
formula based on ab initio cohesive energy
[17–20] is a rigorous and concise technique to
obtain interatomic potentials, which can express
the inversion coefficient of materials with identical
structure concisely and uniformly, making it
convenient for analysis.

2.1. Chen’s lattice-inversion technique

Here, we take a single element crystal as an
example to explain how to use Chen’s lattice-
inversion theorem to obtain the interatomic pair
potential from the first-principle cohesive energy
curve. Suppose that the crystal cohesive energy
obtained by first principle calculation can be
expressed as

\[ E(x) = \frac{1}{2} \sum_{n=1}^{\infty} r_0(n) \Phi(b_0(n)x), \]  \hspace{1cm} (1)

where \( x \) is the nearest-neighbor distance, \( r_0(n) \) is
the \( n \)th neighbor coordination number, \( b_0(n)x \) is
the distance between the reference central atom
and its \( n \)th neighbor, and \( \Phi(x) \) is the pair potential.

By self-multiplicative process from \( \{ b_0(n) \} \), the
multiplicative closed semi-group \( \{ b(n) \} \) is formed.
This implies that a lot of virtual lattice points are
involved, but the corresponding virtual coordina-
tion number is zero. In \( \{ b(n) \} \), there is a sole
integer \( k \) such that \( b(k) = b(m)b(n) \) for any two
integers \( m \) and \( n \). Hence, Eq. (1) can be re-
written as

\[ E(x) = \frac{1}{2} \sum_{n=1}^{\infty} r(n) \Phi(b(n)x), \] \hspace{1cm} (2)

where

\[ r(n) = \begin{cases} r_0(1/b_0^{-1}[b(n)]), & \text{if } b(n) \in \{ b_0(n) \}, \\ 0, & \text{if } b(n) \notin \{ b_0(n) \}. \end{cases} \] \hspace{1cm} (3)

Then the general equation for the interatomic
pair potential obtained from inversion can be ex-
pressed as

\[ \Phi(x) = 2 \sum_{n=1}^{\infty} I(n)E(b(n)x), \] \hspace{1cm} (4)

where \( I(n) \) has the characteristics of

\[ \sum_{b(a)b(n)} I(d)r(1/b_0^{-1}[b(n)/b(d)]) = \delta_{n1}. \] \hspace{1cm} (5)

\( I(n) \) is uniquely determined by crystal geometrical
structure, not related to concrete element category.
Thus, the interatomic pair potentials can be
obtained from the known cohesive energy function
\( E(x) \).

The interatomic pair potential between distinct
atoms can also be obtained by the same inversion
method, for a virtual structure for which the
corresponding cohesive energy only contains this
potential between distinct atoms.
2.2. Method for obtaining quasi-ab initio cohesive energy curve

Once the cohesive energy curves $E(x)$ are known, the corresponding effective interatomic potentials can be obtained by Eq. (4). But how can we obtain the cohesive energy curve, $E(x)$? Therefore, the searching and design of some simple and virtual structures for covering the necessary interatomic potentials are important for us. For example, in order to obtain potentials $\Phi_{\text{Fe} \rightarrow \text{Fe}}(x)$, $\Phi_{\text{Nd} \rightarrow \text{Nd}}(x)$ and $\Phi_{\text{Nd} \rightarrow \text{Fe}}(x)$, we consider three structures, respectively, as follows.

First, let us consider the structure of BCC Fe as B2 or CsCl structure with two simple cubic sublattices, Fe1 and Fe2. Thus, we calculate the energy of the formation of BCC Fe from cubic Fe as

\[
E(x) = E_{\text{Fe}}^{\text{BCC}}(x) - E_{\text{Fe}}^{\text{sc}}(x) - E_{\text{Fe}}^{\text{sc}}(x) = \sum_{i,j,k \neq 0} \Phi_{\text{Fe} \rightarrow \text{Fe}}(x) \times \left( \frac{4}{3} \left[ (i - 1/2)^2 + (j - 1/2)^2 + (k - 1/2)^2 \right] x \right),
\]

where $x$ is the nearest neighbor distance in the BCC structure, $E_{\text{Fe}}(x)$ represents the total energy curve with BCC structure and $E_{\text{Fe}}^{\text{sc}}(x)$ is the total energy function with virtual cubic structure. The calculation was performed by ESCOS code from MSI. Now, $E(x)$ automatically becomes the cohesive energy function of one Fe1 atom with all the Fe2 atoms. Here, the Fe2 atoms form a simple cubic structure, and only one Fe1 atom is located at the center of each cube. Then $\Phi_{\text{Fe} \rightarrow \text{Fe}}(x)$ can be given directly by using Chen’s lattice-inversion technique.

The total energy calculation for Nd with FCC structure can be done by norm-conserving pseudopotential method (CASTEP). Thus, the $\Phi_{\text{Nd} \rightarrow \text{Nd}}(x)$ can be also given by Chen’s lattice-inversion technique.

The ab initio calculation of total energy curve related to $\Phi_{\text{Nd} \rightarrow \text{Fe}}(x)$ is very hard to perform. The lattice constant, module constant and total energy of Nd3Fe with L12 structure near equilibrium position has been calculated. Then the partial cohesive energy of Nd–Fe can be obtained by subtracting the contribution of Nd–Nd and Fe–Fe in the structure, respectively. Subsequently, $\Phi_{\text{Nd} \rightarrow \text{Fe}}(x)$ can be deduced from lattice inversion.

Note that in the conventional lattice-inversion technique [16], the cohesive energy curves are taken from ab initio calculation for some real existing structures. In the present work, the direct calculation for a real structure is impossible. Therefore, the ab initio calculations are based on virtual structures. In fact, the structures of FCC-Nd and L12-Nd3Fe are virtual. This is the reason why we call the resultant potentials quasi-ab initio.

For the above-converted potentials $\Phi_{\text{Nd} \rightarrow \text{Fe}}(x)$, $\Phi_{\text{Nd} \rightarrow \text{Nd}}(x)$ and $\Phi_{\text{Fe} \rightarrow \text{Fe}}(x)$, we found that they can be simply expressed by the Morse approximation. The analytic expression of Morse function for potential versus the distance between two atoms is given by

\[
\phi(x) = D_0 \left( e^{-\gamma[(x/R_0)-1]} - 2e^{-(\gamma/2)((x/R_0)-1)} \right),
\]

Table 1

<table>
<thead>
<tr>
<th>The parameters of some important potentials</th>
<th>$R_0$ (Å)</th>
<th>$D_0$ (eV)</th>
<th>$\gamma$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe–Fe</td>
<td>2.736</td>
<td>0.7636</td>
<td>8.753</td>
</tr>
<tr>
<td>Fe–Nd</td>
<td>3.188</td>
<td>0.6033</td>
<td>10.496</td>
</tr>
<tr>
<td>Fe–Cr</td>
<td>2.820</td>
<td>0.8081</td>
<td>8.474</td>
</tr>
<tr>
<td>Nd–Cr</td>
<td>3.288</td>
<td>0.5275</td>
<td>8.783</td>
</tr>
</tbody>
</table>
where $D_0$, $R_0$, $\gamma$ are potential parameters. Some important potentials are shown in Fig. 1 and the corresponding parameters are listed in Table 1.

### 3. Binary compound Nd$_2$Fe$_{17}$

The structures of Nd$_2$Fe$_{17}$ and Nd$_2$Fe$_{17-x}$Cr$_x$ are simulated by the energy minimization process, which being realized by conjugate gradient methods and the cut-off radius of potentials is taken as 14 Å. To avoid statistic fluctuation, we adopt the model of $2 \times 2 \times 2$ supercell with 456 atoms. Under the control of ab initio potentials, the structure can be relaxed into Th$_2$Zn$_{17}$-type, and the structure parameters are similar to the neutron diffraction data. In this part, the structural stability is tested by some methods including atom-random shift, global deformation and high temperature disturbance. The atom-random shift means moving each atom deviating from equilibrium position in random direction, while the global deformation means making some operations on the model such as stretching, compressing, shearing and the mixture of these ways.

The Th$_2$Zn$_{17}$ structure is disturbed by atom random shift by 0.5 Å. Each atom in a disturbed cell can recover its equilibrium position under the interaction of ab initio potentials. After the relaxation, the structure can be identified as Th$_2$Zn$_{17}$ structure with R$3m$ space group. The atom sites are closer to the neutron diffraction data [21] as shown in Table 2.

For the case of global deformation, the relaxed compounds can recover their initial structure. The calculated results are listed in Table 3. Notice that there is no symmetry restriction in the relaxation process. Moreover, many samples can stabilize to a uniform final structure with identical energy after the energy minimization, even though the average energy of the deformed structure may be very high.

Temperature effect and the dynamic properties are neglected in the above calculations. To check the most stable Nd$_2$Fe$_{17}$ structure, the simulated annealing method is applied to NPT ensemble $P = 0.0001$ GPa, $T = 1200$ K. After 10 cycles, the compound still can be identified as Th$_2$Zn$_{17}$ structure.

The calculated results show that the distorted system can restore the same final structure as Th$_2$Zn$_{17}$. So the potentials can be thought of as possessing long-range character for the restoration of the structure undergoing global deformation, and the process of restoration from the random shift atom model may be considered as liquid–solid phase transformation. High temperature disturbance embodies the dynamic equilibrium properties at different temperatures. It is

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**Table 2**

Comparison between the calculated and experimental [21] structure parameters of Nd$_2$Fe$_{17}$

<table>
<thead>
<tr>
<th>Nd$<em>2$Fe$</em>{17}$</th>
<th>Calc.</th>
<th>Exp. [21]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a$ (Å)</td>
<td>8.582</td>
<td>8.5815</td>
</tr>
<tr>
<td>$c$ (Å)</td>
<td>12.574</td>
<td>12.4546</td>
</tr>
<tr>
<td>Nd (6c)</td>
<td>0.0, 0.338</td>
<td>0.0, 0.3429</td>
</tr>
<tr>
<td>Fe (6c)</td>
<td>0.0, 0.098</td>
<td>0.0, 0.0961</td>
</tr>
<tr>
<td>Fe (18f)</td>
<td>0.293, 0.0</td>
<td>0.2875, 0.0</td>
</tr>
<tr>
<td>Fe (18h)</td>
<td>0.499, –0.02, 0.149</td>
<td>0.5024, 0.0, 0.1557</td>
</tr>
<tr>
<td>Fe (9d)</td>
<td>0.5, 0, 0.5</td>
<td>—</td>
</tr>
</tbody>
</table>

**Table 3**

Determination of the lattice parameters of Nd$_2$Fe$_{17}$

<table>
<thead>
<tr>
<th>Deformed state</th>
<th>$a$, $b$, $c$(Å)</th>
<th>$\alpha$, $\beta$, $\gamma$(deg)</th>
<th>Energy (eV/atom)</th>
<th>Relaxed state under the calculated potentials</th>
<th>$a$, $b$, $c$(Å)</th>
<th>$\alpha$, $\beta$, $\gamma$(deg)</th>
<th>Energy (eV/atom)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10, 15, 20</td>
<td>90, 90, 120</td>
<td>1.43913</td>
<td></td>
<td>8.582, 8.582, 12.574</td>
<td>90, 90, 120</td>
<td>–5.90622</td>
<td></td>
</tr>
<tr>
<td>3.5, 3.5, 6</td>
<td>90, 90, 120</td>
<td>1055.91425</td>
<td></td>
<td>8.582, 8.582, 12.574</td>
<td>90, 90, 120</td>
<td>–5.90622</td>
<td></td>
</tr>
<tr>
<td>8.582, 8.582, 12.574</td>
<td>80, 70, 135</td>
<td>19.26625</td>
<td></td>
<td>8.582, 8.582, 12.574</td>
<td>90, 90, 120</td>
<td>–5.90622</td>
<td></td>
</tr>
<tr>
<td>8.582, 8.582, 12.574</td>
<td>110, 68, 90</td>
<td>1.71509</td>
<td></td>
<td>8.582, 8.582, 12.574</td>
<td>90, 90, 120</td>
<td>–5.90622</td>
<td></td>
</tr>
<tr>
<td>5, 6, 5.5</td>
<td>70, 120, 88</td>
<td>447.39503</td>
<td></td>
<td>8.582, 8.582, 12.574</td>
<td>90, 90, 120</td>
<td>–5.90622</td>
<td></td>
</tr>
<tr>
<td>13, 12, 17</td>
<td>65, 70, 100</td>
<td>–2.02082</td>
<td></td>
<td>8.582, 8.582, 12.574</td>
<td>90, 90, 120</td>
<td>–5.90622</td>
<td></td>
</tr>
</tbody>
</table>
reasonable to conclude that the potentials inverted from the cohesive energy curves of virtual structures in large scale reflect not only the character of equilibrium state but also the non-equilibrium properties to some extent. All the above cases suggest that it is effective for these potentials to simulate different kinds of rare-earth compounds.

4. Ternary compounds Nd$_2$Fe$_{17-x}$Cr$_x$

4.1. Site preference

For the site preference of Cr substitution in Nd$_2$Fe$_{17-x}$Cr$_x$, Girt [21] gave a phenomenological explanation using size effects and enthalpy effects. In this part, the site preference of Cr atom is studied using the calculated potentials. Firstly, Fe atoms in each site are substituted by Cr atoms with different concentrations. Then energy minimization process is applied to relax the ternary system under the interaction of the potentials. Thus, the average energy of final structure can be investigated and compared. Results shown in Fig. 2 suggest that Cr atoms strongly prefer the 6c sites and medially substitute for Fe atom in the 18f sites. The order of site preference is 6c, 18f, 18h and 9d. They correspond well with that of Girt [21], whose work has taken into consideration the chemical, electronic and elastic terms. To avoid the accidental error, we calculate 20 stochastic samples for the arithmetic average. The symbol “I” denotes the range of error bar.

The site preference occupation of the Cr atom is analyzed by the benefit factors as shown in Table 4. The first column includes the sites occupied by the Cr atom, the second column shows the number of Fe atoms within the sphere centered at the Cr atom with a radius of 2.7 Å. Notice that for more Fe atoms in this range, more energy is unfavorable, since $\Phi_{Fe-Cr(r)} > \Phi_{Fe-Fe(r)}$, so there is a negative sign. The third column shows the number of Fe atoms within the range of 2.7–4.4 Å, where $\Phi_{Fe-Cr(r)} < \Phi_{Fe-Fe(r)}$, more Fe atoms in this range, more energy is favorable, so there is a positive sign. Nd atoms shown in the fourth column provide a negative sign, because of $\Phi_{Nd-Cr(r)} > \Phi_{Nd-Fe(r)}$, which is unfavorable for energy decrease. Thus, it can be concluded as the preferential occupation sequence, from Table 4. In fact, the beneficial factors in atom environments reflect both size effects and energy effects.

4.2. Structure of Nd$_2$Fe$_{17-x}$Cr$_x$

The ab initio potentials are applied to check the structure of ternary compound Nd$_2$Fe$_{17-x}$Cr$_x$,
with $x = 0.5, 1, 1.5$, respectively. For $x = 1$, according to the site preference of Cr atom, compound Nd$_2$Fe$_{16}$Cr with Cr atom in 6c site is adopted to check the structure stability using the same method. Results (Table 5) show that the structure can be stabilized to Th$_2$Zn$_{17}$-type even while undergoing either global deformation or atom random shift by 0.5 Å. Furthermore, the atom sites of Nd$_2$Fe$_{17-x}$Cr$_x$ with different $x$ values are evaluated and compared with the neutron diffraction data. The results listed in Table 6 suggest that the structure of Nd$_2$Fe$_{17-x}$Cr$_x$ does not change with the Cr atom introduction, and the lattice parameters are in good agreement with the observed values [21].

4.3. Explanation on the variation of Curie temperature

Based on the site preference of Cr atom in Nd$_2$Fe$_{17}$ and the calculated structures shown in Table 6, the bond lengths of Fe–Fe which play an important role in magnetic properties can be evaluated. In this subsection, the dependence of Curie temperature on the Cr concentration is analyzed qualitatively.

Experimental results [21] show that upon Cr substitution in Nd$_2$Fe$_{17}$, the Curie temperature increases from 327 K up to 374 K at $x = 1$ and then decreases to 356 K for $x = 1.9$. Just as Givord et al. [22] have pointed out, there are two types of exchange interactions, positive and negative, which depend on the length of Fe-dumbbells. All the exchange interactions could be positive except the case associated with 6c–6c, 9d–18f, since the distances between these iron atoms are small.

In order to study the trend of Curie temperature variation, the distance between special sites and the related exchange interaction should be investigated. Fig. 3 presents the bond lengths in Nd$_2$Fe$_{17-x}$Cr$_x$, indicating that the lengths of 6c–6c, 9d–18f are shorter than 2.46 Å which can be considered as the threshold of switching-exchange interaction. The distances between 6c–6c and 9d–18f decrease with increasing Cr content, which are similar to that in the

| Table 5 |
| Determination of the lattice constants of Nd$_2$Fe$_{16}$Cr |

<table>
<thead>
<tr>
<th>Deformed state</th>
<th>Final state under the potentials</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a$, $b$, $c$ (Å)</td>
<td>$x$, $\beta$, $\gamma$ (deg)</td>
</tr>
<tr>
<td>4, 3.5, 6</td>
<td>90, 90, 120</td>
</tr>
<tr>
<td>13, 15, 20</td>
<td>90, 90, 120</td>
</tr>
<tr>
<td>8.599, 8.599, 12.570</td>
<td>78, 80, 140</td>
</tr>
<tr>
<td>8.599, 8.599, 12.570</td>
<td>120, 70, 100</td>
</tr>
<tr>
<td>6, 5.5, 9</td>
<td>80, 110, 100</td>
</tr>
<tr>
<td>14, 15, 22</td>
<td>80, 110, 130</td>
</tr>
</tbody>
</table>

| Table 6 |
| Comparison between calculated and experimental [21] structure parameters of Nd$_2$Fe$_{17-x}$Cr$_x$ |

<table>
<thead>
<tr>
<th>Nd$<em>2$Fe$</em>{17-x}$Cr$_x$</th>
<th>$x = 0.5$ Calc./(Exp.)</th>
<th>$x = 1$ Calc./(Exp.)</th>
<th>$x = 1.9$ Calc./(Exp.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a$ (Å)</td>
<td>8.591/(8.5747)</td>
<td>8.598/(8.5712)</td>
<td>8.614/(8.5656)</td>
</tr>
<tr>
<td>Nd (6c), z</td>
<td>0.331/(0.3433)</td>
<td>0.338/(0.3433)</td>
<td>0.338/(0.3439)</td>
</tr>
<tr>
<td>Fe/Cr (6c), z</td>
<td>0.098/(0.0956)</td>
<td>0.100/(0.0951)</td>
<td>0.100/(0.0949)</td>
</tr>
<tr>
<td>Fe (18f), x</td>
<td>0.292/(0.2878)</td>
<td>0.294/(0.2878)</td>
<td>0.294/(0.2882)</td>
</tr>
<tr>
<td>Fe (18h), x</td>
<td>0.498/(0.5026)</td>
<td>0.496/(0.5028)</td>
<td>0.496/(0.5028)</td>
</tr>
<tr>
<td>Fe (18h), z</td>
<td>0.148/(0.1559)</td>
<td>0.150/(0.1557)</td>
<td>0.150/(0.1559)</td>
</tr>
</tbody>
</table>
experimental trend. Notice that the bond lengths of 9d–18f decrease mildly at the range of 0–0.5, and move steadily from $x = 0.5$ to 1.5, then the variation of exchange interaction from 9d–18f pairs can be neglected. As far as iron pairs except 6c–6c and 9d–18f are concerned, the positive exchange interactions associated with large bond lengths are weak, even though the introduction of Cr atom has the effect of reduction on them. Then the contribution to the exchange interaction from these iron pairs can also be neglected. In the next step, we will focus on the effect of 6c–6c length based on the variation of Curie temperature.

In the above subsection, the analysis of site preference behavior shows that Cr atoms have a pronounced preference for occupying 6c site. On the one hand, the number of 6c–6c (Fe–Fe) dumbbell pairs reduces and weakens the negative exchange interaction with the increase of Cr atom, and is thus beneficial in improving the Curie temperature. On the other hand, bond lengths of 6c–6c pairs decrease with increasing Cr content, and thus enhance the negative exchange interaction and lead to a lower Curie temperature. So there is a competing mechanism arising from Cr content increase, i.e. the variation of Curie temperature is not monotonic with the increment of Cr concentration.

Further, the Fe atoms in 6c site have only one nearest neighbor Fe atom in 6c site. When the Cr atom substitutes Fe atom in 6c site, the mixing enthalpy of Fe–Fe pairs is equal to that of Fe–Cr pairs and the probability of Cr–Cr (6c–6c) pairs increases with Cr content. The relationship between the Curie temperature and Cr concentration is $x - \frac{1}{2}x^2$, and Curie temperature increases at a lower Cr concentration and decreases at a higher Cr concentration. Since the negative exchange interaction of Cr–Cr (6c–6c) is weaker than that of Fe–Fe, it can be neglected.

Givord et al. [22] have shown that the threshold of switching Fe–Fe exchange interaction is 2.45 Å. In other words, if the distance of Fe–Fe is shorter than 2.45 Å, then the exchange interaction would be negative, if it is larger than 2.45 Å, then it would be positive. In the present calculation, the threshold of Fe–Fe exchange interaction is taken as 2.46 Å, instead of 2.45 Å from the experimental analysis. This difference may be caused by a systemic error in derivation of these quasi-ab initio interatomic potentials.

5. Conclusion and discussion

In this work, we provide a series of quasi-ab initio interatomic potentials in the rare-earth intermetallics Nd$_2$(Fe, Cr)$_{17}$. The trick is to drive take the ab initio calculation for some simple virtual structures, together with lattice-inversion technique. These potentials have been applied to a string of examples, only otherwise solved with difficulty, in which there are structural stability, site preference of Nd$_2$(Fe, Cr)$_{17}$ and lattice parameters for this anisotropic system. A more interesting example is the variation behavior in Curie temperature. It is notable that Curie temperature of R–Fe compound is mainly driven by the structure. Like FCC Fe, R$_2$Fe$_{17}$ phase has a strong magnetovolume effect. As far as the distance dependence of the exchange interaction is concerned, there are complex relationships between the ferromagnetic and antiferromagnetic interactions and the dumbbells in 2:17 system. Regarding this, Sabiryanov et al. [9,10,23,24] have carried out excellent experiments...
using ab initio method. Obviously, interatomic potentials could not deal with the magnetic properties, but the structure of the complex system can be studied and then some experimental phenomena related to the structure might be explained by the empirical theory model. This point can also be viewed as one of the potential applications.

All the calculated results in this work are in good agreement with the experimental results. This means the applicability of the present quasi-ab initio interatomic potentials is well proved by demonstration. Surprisingly, the pair-potentials can describe the structures of these extremely anisotropic materials [13–15], especially for these rare-earth intermetallics.

The calculation of the bond lengths of Fe dumbbells together with the analysis of site preference provides the explanation of the variation behavior of Curie temperature, but the temperature effects are neglected in the calculation of the distance between the Fe–Fe atoms. It is notable that the occupancy factors of Cr atom in Nd$_2$Fe$_{17-x}$Cr$_x$ are not studied in detail and the factor in 6c site is taken simply as 1.0, since the energy decreases markedly when Cr occupies 6c site as shown in Fig. 2. Moreover, the threshold of exchange interaction 2.46 Å deviating from experimental value instead of 2.45 Å can be understood, since there are systemic errors in the derivation of these potentials. All these points should be improved in a future study.

As a process of statics, energy minimization requires to search a local minimum value in the phase space from the start point. There are three defects: the effect of initial configuration, local minimum and zero temperature. For the complex system, there are many local minima in phase space, so the energy minimization cannot search the most stable structure with the lowest energy. In order to study this, many samples have to be considered. At zero temperature, the vibration properties and other dynamics properties are neglected. Against the drawbacks, three methods are applied to solve them. The global deformation and atom random shift are used to solve the question of initial structure to some extent. Moreover, simulated annealing molecular dynamics are applied to overcome the initial configuration and the contribution of temperature is considered.

According to our calculations, in a few cases, the ab initio calculation of structural properties for some rare-earth compounds can be performed. However, this kind of “direct” calculation for “real” complex materials has less accuracy than the indirect calculation based on the interatomic potentials extracted from “virtual” materials. In general, it is necessary to calculate the cohesive energy curve to extract the interatomic potentials, but the direct ab initio calculation for a complex rare-earth compound is either difficult or impossible. This forces us to seek some virtual structures which contain all the required interatomic potentials and the calculation of which becomes easy and reliable.

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