The Magnetic, Structural and Electronic Properties of Novel Y\textsubscript{2}FeSi Full-Heusler Alloy

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The paper presents the results of the plane wave density functional theory calculations of newly predicted Y\textsubscript{2}FeSi full-Heusler alloy with optimized lattice constants. We have explored two most likely structures (AlCu\textsubscript{2}Mn and CuHg\textsubscript{2}Ti prototypes) with different atomic arrangements and including collinear magnetic ordering. The results show that the classic L2\textsubscript{1} structure is more favourable in terms of energy than the second one studied in this work. Moreover, for both crystalline structures ferromagnetic arrangement is more favourable, although in the second structure the difference in energy between ferromagnetic and nonmagnetic state is small. Study did not revealed band gap in any spin channel, yet the spin polarization in majority channel at the Fermi level is significant ($\sigma = 83\%$). The calculated structural lattice $a$ parameter for the energetically lowest structure is $a = 13.0996\ a_0$. For the lattice constant range of 12.2 to 13.2 $a_0$ the total magnetic moment varies significantly from 0.86 $\mu_B$ to 1.74 $\mu_B$ per formula unit.

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1. Introduction

Recently, both half-Heusler (HH) and full-Heusler (FH) materials have attracted a lot of attention due to their excellent properties that include half metallic state and magnetic ordering, in the same time. Those properties result in their potential application in spintronic devices such as spin-filters [1] or spin injection devices [2, 3]. However, to adjust parameters of such materials, a basic knowledge at the lowest atomic level is required, especially focused on its electronic properties. The electronic structure of FH and HH materials is inextricably linked to its crystallographic structure, thus changing the atomic site occupations will significantly influence materials various properties, despite the same chemical composition.

The full-Heusler alloys of the X\textsubscript{2}YZ composition (where X and Y are elements from transition metal d-orbital block and Z is sp-orbital element from p block [4]) crystallize in one of two main structures, namely L2\textsubscript{1} (AlCu\textsubscript{2}Mn-type) and CuHg\textsubscript{2}Ti-type. The final structure in which material crystallizes depends on the overall energy which it can obtain by choosing one of them. Here many properties affect this energy like the range of energy which it can obtain by choosing one of them. Here many properties affect this energy like the range of energy which it can obtain by choosing one of them. Therefore, the choice of the structure can significantly influence the magnetic and electronic properties of the material.

In this paper, the magnetic structural and electronic properties of novel Y\textsubscript{2}FeSi full-Heusler alloy are determined using density functional theory (DFT) calculations.

2. Computational details

The calculations were completed using Quantum Espresso software package [9]. Both geometrical optimization and electronic structure calculations were performed using DFT which were carried out by the means of plane wave method. The generalized gradient approximation (GGA) in the Perdew, Burke, and Ernzerhof (PBE) form were used [10]. For all calculations the projector augmented-wave (PAW) pseudopotentials were used. The electronic configurations for Y, Si, and Fe elements were [Kr] 5s\textsuperscript{2} 5p\textsuperscript{6} 4d\textsuperscript{5}, [Ne] 3s\textsuperscript{2} 3p\textsuperscript{2} 3d\textsuperscript{6}, and [Ar] 4s\textsuperscript{2} 3d\textsuperscript{10} 4p\textsuperscript{6}, respectively. When considering L2\textsubscript{1} structure (space group Fm-3m, no. 225) the Y occupies 8c ($\frac{1}{2}$, $\frac{1}{2}$, $\frac{1}{2}$), Si — 4b ($\frac{1}{4}$, $\frac{1}{4}$, $\frac{1}{4}$) and Fe — 4a (0,0,0) prototype AlCu\textsubscript{2}Mn. For the second Heusler structure of CuHg\textsubscript{2}Ti prototype (F-43m, no. 216), Y atom occupies 4a (0,0,0) and 4c ($\frac{1}{2}$, $\frac{1}{2}$, $\frac{1}{2}$), Si — 4d ($\frac{1}{4}$, $\frac{1}{4}$, $\frac{1}{4}$) and Fe — 4b ($\frac{1}{2}$, $\frac{1}{2}$, $\frac{1}{2}$) sites. All computations were done including scalar relativistic effects. Before final calculations to ensure accurate results we conducted proper convergence tests. On that base, we established a 17 $\times$ 17 $\times$ 17 k-points mesh aligned in the Monkhorst–Pack scheme [11] for sampling of the Brillouin zone. The kinetic energy cutoff was set to ecutwfc= 90 Ry (0.1224 × 10\textsuperscript{4} eV) and cutoff for charge density ecutrho= 900 Ry (1.2245 × 10\textsuperscript{4} eV). To properly treat metallic materials and accelerate the convergence Marzari–Vanderbilt [12] cold smearing of the Fermi surface with value of 0.005 Ry (0.068 eV) was used. The electronic convergence criteria was set to 10\textsuperscript{−8} Ry (1.36 × 10\textsuperscript{−7} eV). For the relaxation procedures,

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the total force threshold for ionic minimization was set to $1 \times 10^{-3}$ a.u. and kinetic stress for equilibrated structure below 0.1 kPa. All relaxation calculations were done including proper magnetic ordering when needed, two options were considered: non-magnetic (NM) case and ferromagnetic (FM) spin ordering for both investigated structures.

3. Results and discussion

Figure 1 shows both considered structures of Y$_2$FeSi material. At first stage the structural properties (equilibrium lattice constant) were calculated for all the structures using classic minimization of energy in respect of cubic lattice parameter $a$ and substituting results to the Murnaghan equation of state [13] (see Fig. 2, Eq. (1)). Then we utilized the Broyden–Fletcher–Goldfarb–Shanno (BFGS) algorithm to further improve accuracy and relax the ionic positions inside the cell. The cell parameters data for both structures resulted from last step have been summarized in Table I.

As can be seen, the most energetically stable configuration is ferromagnetic $Fm\bar{3}m$ ($L2_1$). For the second investigated structure, both FM and NM states show similar behaviour. In this case the calculation showed that attempt to enforce a ferromagnetic ordering strives for non-magnetic configuration during SCF (Self-Consistent Field) cycles. Because the $L2_1$ structure is more favourable, the following part of the article will refer to this phase exclusively.

From the fit of energy-volume curve to the Murnaghan equation of state (1) the bulk modulus can be calculated. For the $L2_1$ FM structure we report its value to be $B = 75.9$ GPa.

$$E(V) = E_0 + \frac{K_0 V}{K_C} \left( \frac{V_0}{V} \right)^{K_0} - 1 + 1 - \frac{K_0 V_0}{K_0 - 1},$$

(1)

where $K_0$ — bulk modulus, $K'_0$ — derivative of $K_0$, $E_0$ — equilibrium energy, $V_0$ — equilibrium volume, $V$ — volume.

Figure 3 shows change of total and local magnetic moments for Y$_2$FeSi in $L2_1$ FM configuration in respect of lattice $a$ parameter. The total magnetic moment is defined as:

$$\int (n_{up} - n_{down}) \, d^3r,$$

(2)

Where $n_{up}$ and $n_{down}$ are magnetic moments coming up from spin up and down, respectively. The calculated total magnetic moment for relaxed structure is 1.56 $\mu_B$/cell. As can be seen, total magnetic moment rises with rising $a$ up to 12.8 a.u. and, above this value, increase of $\mu_B$ is smaller but still visible. The main contribution to total magnetization is caused by Fe site with very small

<table>
<thead>
<tr>
<th>Structure</th>
<th>$a$ lattice parameter [a.u.]</th>
<th>Volume [a.u.$^3$]</th>
<th>Density [g/cm$^3$]</th>
<th>Total energy [eV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Fm\bar{3}m$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>FM</td>
<td>13.09958</td>
<td>561.9686</td>
<td>5.218</td>
<td>$-12806.67186$</td>
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<tr>
<td>NM</td>
<td>12.92950</td>
<td>540.3624</td>
<td>5.425</td>
<td>$-12806.36365$</td>
</tr>
<tr>
<td>$F-43m$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>FM</td>
<td>12.57045</td>
<td>496.5837</td>
<td>5.904</td>
<td>$-12805.44116$</td>
</tr>
<tr>
<td>NM</td>
<td>12.56100</td>
<td>495.4646</td>
<td>5.898</td>
<td>$-12805.43805$</td>
</tr>
</tbody>
</table>
addition of silicon moment. At the same time both yttrium sites are polarized anti-parallel to the iron, lowering the total magnetization.

Below 12.8 a.u. as the cell compression increases, a sharp decrease in total magnetization can be seen. This is mainly caused by drop of Fe local moment, which behaves similarly in this range and two yttrium sites aligned anti-parallel. A very slight drop of Si from 0.1253 \( \mu_B \) at \( a = 12.8 \) to 0.1212 \( \mu_B \) at \( a = 12.2 \) a.u. can also be observed. At \( a = 12.2 \) a.u. (which corresponds to 2.38 GPa of hydrostatic pressure) the total magnetic moment drops to 0.86 \( \mu_B \), almost twice smaller when compared to equilibrium lattice constant.

As the results have shown, the most energetically stable configuration from those considered both with non-magnetic and ferromagnetic spin arrangement is the \( L_2_1 \) with ferromagnetic spin ordering. The magnetic properties are strongly related to the crystal structure, causing an increase in the total magnetic moment as the cell size increases beyond equilibrium, and a sharp drop, when cell is compressed. However, despite of the increase in total magnetization the decrease in local moment of Y ions are also observed, therefore an imbalance resulting from opposite magnetic moments exists. The low influence of Si local moment is stable in studied range. For the second investigated structure (\( \text{CuHg}_2\text{Ti} \) prototype) at equilibrium, materials magnetization drops to zero.

The electronic structure calculations (atom-projected density of states) showed that for equilibrium lattice constant of \( a = 13.0996 \) a.u. for \( L_2_1 \) ferromagnetic structure, main contribution to the DOS at the Fermi level and therefore to total magnetic moment (1.56 \( \mu_B \)) comes from iron ions. High spin polarization of \( \sigma = 83\% \) opens up application possibilities for use in spin filter or spin injection devices.

**4. Conclusions**

In this paper we studied structural, magnetic and electronic properties of novel full-Heusler \( \text{Y}_2\text{FeSi} \) material.