Calculation of magnetic moments and lattice parameters Co-based Heusler alloys with determination of their energy favorable structure

Marina V. Mamonova, Irina A. Prudnikova
1Department of Theoretical Physics, Omsk State University, Omsk, Russia
2Omsk State Agrarian University, Omsk, Russia
E-mail: 1mamonovamv@omsu.ru 2ia.prudnikova@omgau.org

Abstract. In this work we present results of ab-initio investigation atomic structures and magnetic properties of Cobalt-based full-Heusler compounds with composition Co2YZ by using VASP software package. The lattice parameters of Co2FeAl(CFA), Co2FeSi(CFS), Co2FeAl0.5Si0.5(CFAS) Heusler alloys depending on the parameters of convergence are calculated. Investigation of CFA and CFS alloys in B2-type and L21-type structures with determination of their energy efficiency of the formation are carried out. The values of the magnetic moment of CFAS atoms in the bulk and in CFAS/Ag(100)/CFAS multilayer structure taking into account the relaxation effects are determined.

1. Introduction
Cobalt-based full-Heusler compounds with composition Co2YZ (where Y is a transition metal and Z is a main group element) are attracting attention due to their predicted half-metallic behaviour, a much desired property for spintronic devices [1-2]. Knowledge of the basic magnetic properties of these materials, especially in the form of thin films, is required both to exploit these promising materials and to understand the properties of magnetic multilayer structures based on them. Prominent are the Co2-based Heusler compounds, which gained a particular interest and became most widely studied in this field due to their high Curie temperatures, a requirement for spin-dependent electron transport devices. Tezuka et al. [3] reported tunnel junctions built using the iso-electronic compound Co2FeAl(CFA), Co2FeSi(CFS), Co2FeAl0.5Si0.5(CFAS). The junctions exhibited tunnel magnetoresistance (TMR) ratios for the L21 structure of 51% and 78% at 300 K and 5 K, respectively. In this work, we use the spin-density functional method for theoretical description and calculation of energy and magnetic characteristics of Co2FeAl(CFA), Co2FeSi(CFS), Co2FeAl0.5Si0.5(CFAS) Heusler alloys.

2. Model and methods
We studied Cobalt-based full-Heusler compounds with the use of VASP (Vienna Ab-Initio Simulation Package) software package [4] by means of the Projector Augmented Wave (PAW) method with PBE [5] version of generalized gradient approximation (GGA) to describe the exchange-correlation interactions.
Figure 1. a) \(L_2\)-type and b) \(B_2\)-type ordered structure of \(Co_2FeAl\)-alloy

Table 1. Result of calculation cell total energy and lattice parameters for \(B_2\) and \(L_2\)-ordered structures of \(Co_2FeAl\)-alloy.

| Type ordering | Energy, eV     | \(a, \text{Å}\) | \(a_{\text{lit}}, \text{Å}\) | \(a_{\text{exp}}, \text{Å}\) [6] |
|---------------|---------------|----------------|-----------------|-----------------|
| \(B_2\)      | -107.1048(64) | 5.6800(400)    | —               | 5.7300          |
| \(L_2\)      | -109.8830(20) | 5.6965(9)      | 5.6900          | 5.7030          |

Heusler compounds with the general formula \(Co_2YZ\) crystallize in the \(L_2\) and the \(B_2\) structures. At \(L_2\) structure (figure 1a) the cubic unit cell consists of four interpenetrating fcc sublattices, two of which are occupied by Co atoms and the other two by the Y and Z atoms, respectively. At \(B_2\) (figure 1b) structure in \((0,0,0)\) and \((1/2,1/2,1/2)\) sites are randomly occupied by Y and Z with an equal probability.

Since the half-metallicity of full-Heusler alloys depends on their atomic ordered crystal structures, determination of the energy efficiency of the formation of various ordered crystal structures are necessary. We obtained the total energy and lattice parameters as the functions of convergence parameters. We confined ourselves to values of the plane waves cut-off energy \(E_{\text{cut}} = 400\text{eV}\) and Monkhorst-Pack grid size 16x16x16.

The results of our calculations have shown that the bulk energy of CFS alloy \(L_2\)-type structure is less than in the case of in \(B_2\)-type structure, this suggests that the \(L_2\)-type structure is energetically more favorable. The values of the lattice constant were found from the condition of the minimum energy of the unit cell. The calculated bulk lattice parameters of \(B_2\) and \(L_2\)-ordered structures of \(Co_2FeAl\) Heusler compound are listed in table 1 and are in good agreement with the experimental values.

Similar calculations total energy and lattice parameters were conducted for another Heusler alloy \(Co_2FeSi\) and are presented in table 2. These results determines that the formation of a \(L_2\)-ordered structure for \(Co_2FeSi\)-alloy is also energetically more favorable as for \(Co_2FeAl\). The agreement of our calculated values with experimental ones demonstrate the reliability of our calculations.

Comparison of the magnetic moments of the atoms in both \(L_2\) and \(B_2\)-type ordered structures \(Co_2FeSi\) alloy, which values are presented in table 3, allows to conclude that as
Table 2. Result of calculation cell total energy and lattice parameters for B2 and L21-type ordered structures of Co2FeSi-alloy.

| Type ordering | Energy, eV  | a, Å    | a_{lit}, Å [7] | a_{exp}, Å [8] |
|---------------|------------|---------|----------------|----------------|
| B2            | -115.0021(88) | 5.5732(139) | 5.5780         | —              |
| L21           | -116.5981(76) | 5.6224(6)   | 5.6310         | 5.6400         |

Table 3. Result of calculation magnetic moment of atoms $\mu$, $\mu_B$ and cell total magnetic moment $\mu_{tot}$, $\mu_B$ for Co2FeSi-alloy.

| Type ordering | $\mu_{Co}$ | $\mu_{Fe}$ | $\mu_{tot}$, $\mu_B$ |
|---------------|-------------|-------------|-----------------------|
| B2            | 0.9200      | 1.6040      | 16.1056               |
| L21           | 1.3130      | 21.6396     |                       |

Table 4. Comparison results of calculation cell total energy, lattice parameters and magnetic moment of atoms for L21-type ordered structure Co-based Heusler alloys.

| Alloy  | Energy, eV   | a, Å   | $\mu_{Co}$, $\mu_B$ | $\mu_{Fe}$, $\mu_B$ | $\mu_{tot}$, $\mu_B$ |
|--------|--------------|--------|----------------------|----------------------|-----------------------|
| CFAS   | -113.41573(965) | 5.579(12)   | 1.305                | 2.812                | 21.700                |
| CFS    | -116.5981(76)  | 5.6224(6)    | 1.3130               | 2.7840               | 21.6396               |
| CFA    | -109.8830(20)  | 5.6965(9)    | 1.187                | 2.751                | 19.940                |

magnetic moments of individual Co and Fe atoms and cell total magnetic moment in L21-type structure is greater than corresponding values in the case of B2-type structure. This is due to the fact that the atoms of iron and cobalt in the case of B2-type structure are in the same layer. $Co_2FeAl_{0.5}Si_{0.5}$ (CFAS) Heusler alloy is more interesting as material for thin film in multilayer
structure, since, for example, values of the tunnel magnetoresistance (TMR) ratio CFAS are larger than those found for pure Co$_2$FeAl or Co$_2$FeSi alloys [3]. Since the previous calculations showed that the $L2_1$-type ordered structure is more energetically favorable for the alloys of the CFA and CFS, it was decided to consider it immediately for the CFAS. Results of calculation cell total energy, lattice parameters and magnetic moment of atoms for $L2_1$-type ordered structure Co-based Heusler alloys (CFA, CFS and CFAS) are combined in table 4. Among these alloys, CFAS has the lowest lattice constant, but the largest magnetic moment.

3. Investigation of magnetic properties CFAS/Ag/CFAS multilayer structure
In the case of CFAS/Ag/CFAS multilayer structure, the Ag lattice cell parameter is too small to accommodate a CFAS compound in the cube-on-cube epitaxial relationship. However, as shown in figure 3, if the CFAS cube edge grows at an angle of 45 with respect to an in-plane Ag(001) direction, a good lattice match is obtained, enabling epitaxial growth.

![Figure 3. Epitaxial relationships between CFAS and Ag(100)](image)

The supercell of CFAS/Ag/CFAS structure is consist of the 13 monatomic layers and vacuum layer with thickness 5Å. The layers of CFAS are placed on each side of the Ag five-layer slab, as shown in figure 4. The magnetic moment of atoms is directed collinearly along the z axis. The dimension of the k-point grid was selected 21x21x1. Calculations of this system consisting of 52 atoms and thickness 28Å were carried out on the basis of the Center for Data of the Far Eastern Branch of the Russian Academy of Sciences in the city of Khabarovsk, since these calculations require a large amount of RAM. Total CPU time at 8 cores OpenPower8 was 26623sec $\approx$ 7hours and maximum memory used was 10137.22 KBytes.

Figures 4 present the arrangement of atoms projected along the z axis and result of the calculation magnetic moment of atoms. Comparison of the values of the magnetic moment of CFAS atoms in the bulk (table 4.) and in the film adsorbed on Ag shows, that the Co atoms most distant from the substrate have greatest magnetic moment. For Fe atoms we obtained that magnetic moment for atoms in the bulk cell are slightly larger then magnetic moment for atoms located on substrate.

This figure 5 represents the comparative arrangement of the atomic layers and the change in the interlayer distance of the system before and after relaxation. An analysis of the figures 5 shows that the interlayer distance of outermost layers are compressed and the remaining especially silver interlayer distance are expanded.
Figure 4. System CFAS/Ag/CFAS (Co - pink, Fe - yellow, Al - blue, Ag - gray)

Figure 5. Atomic layers position for CFAS/Ag/CFAS system without and with according relaxation.

4. Conclusions

In this work we present results of ab-initio investigation atomic structures and magnetic properties of Cobalt-based full-Heusler compounds $\text{Co}_2\text{FeAl}$ (CFA), $\text{Co}_2\text{FeSi}$ (CFS), $\text{Co}_2\text{FeAl}_{0.5}\text{Si}_{0.5}$ (CFAS) by using VASP software package.

The results of our calculations have shown that the bulk energy of CFA and CFS alloys in $\text{B}_2$-type structure is greater than in the case of $\text{L}_2_1$-type structure, this suggests that the $\text{L}_2_1$-type structure is energetically more favorable. Also total magnetic moment in $\text{L}_2_1$-type structure is greater than in the case of $\text{B}_2$-type structure.

Investigation of the alloys lattice parameter dependence on the parameters of convergence made it possible to choose the optimal values of the plane waves cut-off energy $E_{\text{cut}} = 400$ eV and Monkhorst-Pack grid size 16x16x16. The optimized lattice constants for Co-based Heusler alloys $a_{\text{CFA}} = 5.6965(9)$ Å, $a_{\text{CFS}} = 5.6224(6)$Å, $a_{\text{CFAS}} = 5.5790(4)$ Å obtained by us are in good agreement with the experimental values.

Comparison of the values of the magnetic moment of CFAS atoms in the bulk and in the film adsorbed on Ag shows, that the Co atoms most distant from the substrate have greatest magnetic moment. For Fe atoms we obtained that magnetic moment for atoms in the bulk cell are slightly larger then magnetic moment for atoms located on substrate.

These results can be applied in numerical simulation by the Monte Carlo methods of the nonequilibrium behavior of multilayer magnetic superstructures.
Acknowledgments
This research was supported by the grants 17-02-00279, 18-32-00814 of Russian Foundation of Basic Research. The simulations were supported in through computational resources provided by the Shared Facility Center “Data Center of FEB RAS” (Khabarovsk).

References
[1] Bass J and Pratt W P 1999 J. Magn. Magn. Mater. 200 274–89
[2] Prudnikov V V, Prudnikov P V and Romanovskiy D E 2016 J. Phys. D: Appl. Phys. 49 235002
[3] Tezuka N, Ikeda N, Miyazaki A, Sugimoto S, Kikuchi M and Inomata K 2006 Appl. Phys. Lett. 89 112514
[4] Kresse P G and Furthmüller J 1996 Phys. Rev. B 54 11169
[5] Perdew J P, Burke K and Ernzerhof M 1996 Phys. Rev. Lett. 77 3865
[6] Elmers H J, Wurmehl S, Fecher G H et al. 2004 Appl. Phys. A 79 557
[7] Amari S, Dahmane F, Bin Omran S et al. 2016 Journal of the Korean Physical Society 69 1462–68
[8] Wurmehl S, Fecher G H, Kandpal H C, Ksenofontov V 2005 Phys. Rev. B 72 184434