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First principles electronic structure calculations of Co_2CrBi Heusler system

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ABSTRACT

First principles calculation results of a new full Heusler system Co_2CrBi in stoichiometric composition were presented. The calculations are based on the density functional theory (DFT) within the spin-polarized generalized gradient approximation (σ–GGA) and plane wave pseudopotential method. The system shows nearly half-metallic behavior with very low electronic density of states of minority spins at Fermi level yielding high spin polarization ratio R=0.96. The total magnetic moment of the system was calculated as $5.05\mu_B$, which is largely localized at chromium site with $\mu_{Cr} = 3.35\mu_B$. The electronic character of the compound is determined by the 3d electronic states of cobalt and chromium atoms. f-Electronic states of bismuth atom were also included in calculations although it has no distinct effect on electronic structure. The system can be labelled as a new half-metallic material for technological applications.

1. Introduction

Half-metallic ferromagnetism is of great interest due to invaluable technological applications in the field of spintronics. These materials are characterized by high-spin polarization at Fermi level. In the ideal case, the electronic band structure of one spin state shows metallic character with finite density of states at Fermi level while the band structure of the other spin state is a semiconductor yielding a high spin polarization ratio. The perfect half-metallism is an ideal case which can only be achieved at extreme physical conditions like zero temperature, the absence of impurities, and by neglecting spin–orbit interactions [1]. The large magnetic moment and high Curie temperature are also necessities for the stability of ferromagnetic phase and high-technology device applications. The Heusler compounds are a set of materials some of which are shown to be half-metallic from first principles calculations.

Half-metallic ferromagnetism was first introduced by de Groot et al. [2] in half-Heusler compound NiMnSb whose crystal structure conforms to $C1_b$ crystallographic group. Principally, half-metals are able to conduct a fully spin-polarized current which results in a very large magneto-resistance. These types of materials play a prominent role in various spin-dependent electronic applications like spin injection to semiconductors [3–5], spintronics [6,7], and giant magneto-resistance spin valve [8].

The first principles calculations are largely used as an extensive tool in studies of these materials and theoretically, several compounds were predicted to be half-metallic [1,9–13]. The electronic structure and magnetic properties of several half- and full-Heusler compounds were studied extensively in previous works [14,15]. The electronic structure and spin-polarization properties of half-metallic Heusler alloys were also reviewed in a recent comprehensive ab initio study [16]. Co-based full-Heusler compounds (Co_2YZ) in cubic $L2_1$ structure, specially Co_2MnSi, seem to be the most promising material for spintronics applications due to ideal physical properties like high Curie temperature (T_c=985 K) [17], wide band gap in minority spins [13] and being easy to synthesize experimentally. The Curie temperature and total magnetic moment of Co_2FeSi system were determined as 1100 K and $6\mu_B$, respectively [18]. These values make the Co_2FeSi system a good candidate for future technological applications. Co-based Heusler compounds were investigated theoretically in view of density functional calculations and most of them are predicted to be half-metallic [11–14,17,19,20]. Among the several Heusler systems, Co_2MnSi compound was used in production of thin films [21–23] and devices [24,25] by several groups.

The origin of the half-metallicity of full-Heusler compounds is more complex than in the half-Heusler alloys due to the presence of the states located entirely at the Co sites [26,14] and these alloys are considered to be an ideal local moment system [27,28]. The integer magnetic moment is an important characteristic property for half-metallic systems in stoichiometric composition. The total magnetic moment of the system obeys Slater–Pauling [29,30] rule in which the saturation magnetic moment scales with the number of valance electrons [31]. The small deviations from integer magnetic moment lead to significant disruptions in half-metallic character by non-zero electronic density of states for minority spins around Fermi level.

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Up to now, the reported half-metallic ferromagnetic Heusler compounds are largely manganese based. In this study, a chromium based Heusler alloy was studied from first principles. In literature, there is only one study about the Co_2CrBi full Heusler system [32], which was based on the full potential linearized-augmented plane wave method. In this study, the relevant system was studied comprehensively by plane wave pseudopotential method within the generalized gradient approximation for exchange-correlation functional.

2. Computational details

The full-Heusler alloys are ternary intermetallic compounds based on the X_2YZ stoichiometry for the $L2_1$ phase ($Fm\bar{3}m$ space group, #225). X atoms are transition metals which stand on (0 0 0) and ($\frac{1}{2}$ $\frac{1}{2}$ $\frac{1}{2}$) Wyckoff crystallographic positions, while Y and Z are magnetic transition metal and III–V group element occupying the positions ($\frac{1}{4}$ $\frac{1}{4}$ $\frac{1}{4}$) and ($\frac{3}{4}$ $\frac{3}{4}$ $\frac{3}{4}$), respectively. The importance of these materials arise from the ferromagnetic behavior, even though none of the atoms in the composition is ferromagnetic.

All calculations have been performed using the PWscf code, distributed with the Quantum ESPRESSO package [33]. The exchange-correlation potential was approximated by spin polarized GGA (generalized gradient approximation) of the density functional theory [34,35] (DFT) with Perdew–Burke–Ernzerhof parameterization [36]. The utilization of GGA functionals is important, which is more successful in description of the physical properties of Heusler compounds than L(S)DA, so the obtained results are in better agreement with experiments. Ultrasoft pseudopotentials are generated by scalar relativistic calculation for cobalt and chromium atoms with non-linear core correction, while a norm-conserving pseudopotential is used for bismuth atom. The valance states of the atoms considered are as follows: Co: $3d^7\,4s^2$, Cr: $3d^4\,4s^2$, Bi: $6s^2\,6p^3$. Brillouin zone integration was performed with automatically generated $12\times12\times12$ k-point mesh following the convention of Monkhorst and Pack [37] yielding 144 k points in the irreducible wedge of the Brillouin zone centered at $\Gamma-$point. Wave-functions were expanded in plane wave basis sets up to a kinetic energy cut-off value of 1160 eV. This corresponds to about 5350 plane waves. These values are tested and determined to provide convergence in self-consistent calculations. Methfessel–Paxton type smearing was applied on fermionic occupation function with $\sigma=0.25$ eV smearing parameter [38]. The Kohn–Sham equations were solved by iterative Davidson type diagonalization method [39] with 1×10^{-8} Ry energy convergence threshold. Spin–orbit effects are neglected in the calculation procedure.

The static equation of states of the system was constructed using Vinet equation of states [40], which is found to be the most accurate among the several equations of states formulations in a previous comprehensive study [41]. The fits are performed using total energies at 15 different volumes ranging from 0.81 to 1.15 V_0.

3. Results and discussion

The half-metallic character of several half-metals is strongly dependent on the stability of crystal structure. The applied stress or small deviations from equilibrium state can destroy the half-metallicity. Therefore the determination of equilibrium structural parameters has central importance for further analysis in first principles calculations. The static equation of states of Co_2CrBi system was constructed using Vinet equation as shown in Fig. 1. It is worth noting that all parameters used in calculations were tested for convergence (e.g. electron kinetic energy cut-off and k-point mesh). The obtained statistical error in fitting process is less than 0.01% which is a strong indication of accuracy. The calculated equilibrium volume of fcc primitive cell is 403.627 a.u.^{3} yielding 11.7313 a.u. optimized lattice constant. The calculated structural parameters and total and partial magnetic moments of the atoms in composition were given in Table 1 together with other theoretical work [32]. The equilibrium lattice constant was calculated as 11.4499 a.u. in Ref. [32] which is based on the FLAPW method with GGA functionals. The calculated bulk modulus of the system is 132.7 GPa which is relatively low in comparison to other half-metallic Heusler systems (e.g. $B\cong220$ GPa for Co_2MnSi). Since the bulk modulus denotes the mechanical resistivity of the material to a uniform hydrostatic pressure, Co_2CrBi system cannot be regarded as a hard material. Up to now, there is not any experimental study about this system, so there is no chance for comparison of the calculated parameters.

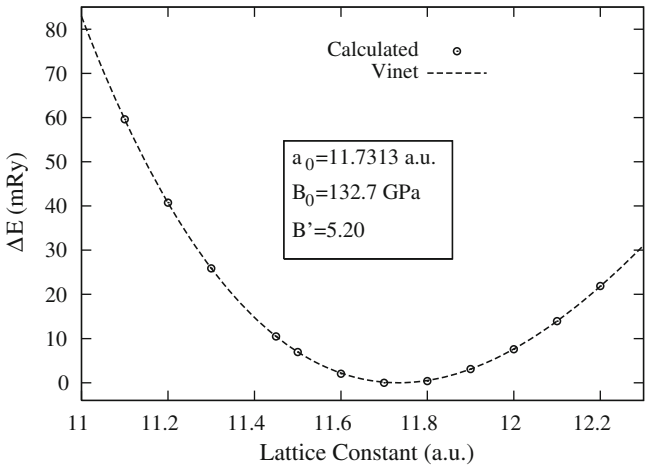

Fig. 1. Static equation of states of Co_2CrBi system according to Vinet formulation. Energy was expressed as relative energy due to equilibrium state. The circles show the computed data, while the line shows the fit curve of Vinet equation.

Table 1
Calculated values of equilibrium structural parameters of Co_2CrBi together with available theoretical work.

	Present work	Ref. [32]
a (a.u.)	11.7313	11.4499
B_0 (GPa)	132.7	–
B'	5.20	–
μ_{tot} (μ_B)	5.05	4.98
μ_{Co} (μ_B)	1.74	1.52
μ_{Cr} (μ_B)	3.35	3.58
μ_{Bi} (μ_B)	−0.02	−0.12

The total magnetic moment of perfect half-metallic Heusler systems obeys Slater–Pauling rule [29,30] in which total magnetic moment of the system is an integer and is expressed as $M_t=Z_t-24$, where Z_t is the total number of valance electrons. Co_2CrBi system has 29 valance electrons in total and the calculated net magnetic moment is $5.05\mu_B$. The partial magnetic moments of each atom in composition are 1.74, 3.35, and $-0.02\mu_B$ for Co, Cr, and Bi atoms, respectively. The magnetic moment of the system is largely localized on chromium site as expected. The slight deviation of net magnetic moment from integer value is a clue for non-perfect half-metallic character and is due to the absence of energy gap in minority spin channel. The absolute magnetization of the system is calculated as $5.27\mu_B$. The difference between net and absolute magnetization can be attributed to non-collinear spin configurations. The appearance of local antiferromagnetic states is also a reason. In this sense, the negative magnetic moment of bismuth