

A density functional theory (DFT) study of Co₂CrGe: LSDA method



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Abstract

The structural, electronic and magnetic properties of Co₂CrGe, a Heusler alloy, have been evaluated by first principles density functional theory and compared with the known experimental and theoretical results. Generalized gradient approximation (GGA) is used for structural study where as Local spin density approximation (LSDA) for electronic calculation. First principles structure optimizations were done through total energy calculations at 0K by the full potential linearized augmented plane wave (FP-LAPW) method as implemented in WIEN2K code.

Keywords: GGA, half-metallicity, DOS and band structure.

Resumen

Las propiedades estructurales, electrónicas y magnéticas del Co₂CrGe, una aleación Heusler, han sido evaluadas por la teoría de densidad funcional de primeros principios y son comparados con los experimentos conocidos y resultados teóricos. La aproximación generalizada de gradiente (GGA) es usada para el estudio estructural con la aproximación de la densidad spin local (LSDA) para el cálculo electrónico. Las optimizaciones de estructura de los primeros principios fueron realizados a través de los cálculos de energía total a 0K por el potencial completo linealizado de onda plana (FP-LAPW) como método implementado en el código WIEN2K.

Palabras clave: GGA, media-metalicidad, DOS y estructura de banda.

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I. INTRODUCTION

In 1983, de Groot discovered half-metallic ferromagnetism in semi-Heusler compound NiMnSb [1] by using first-principle calculation based on density functional theory. Heusler alloys are the ternary intermetallic compounds with composition X₂YZ, where X and Y are transition elements (Ni, Co, Fe, Mn, Cr, Ti, V etc.) and Z is III, IV or V group elements (Al, Ga, Ge, As, Sn, In etc.). One of the promising classes of materials is the half-metallic ferrimagnets, *i.e.*, compounds for which only one spin channel presents a gap at the Fermi level, while the other has a metallic character, leading to 100% carrier spin polarization at E_F [4]. After that, half-metallicity attracted much attention [2], because of its prospective applications in spintronics [3]. The electronic and magnetic properties of Co₂MnAl [5] and Co₂CrSi [6] using LSDA shows the half-metallicity at the ground state. In this present work, we report the result of GGA and LSDA of bulk electronic structure and magnetic properties of Co₂CrGe.

II. COMPUTATIONAL DETAILS AND CRYSTAL STRUCTURE

The FP-LAPW method (WIEN2K) [7] was applied to band structure calculations of Co₂CrGe. GGA [8] and LSDA were used for the exchange correlation potential. The multipole expansion of the crystal potential and the electron density within muffin tin (MT) spheres was cut at $l=10$. Nonspherical contributions to the charge density and potential within the MT spheres were considered up to $l_{max}=6$. The cut-off parameter was $RK_{max}=7$. In the interstitial region the charge density and the potential were expanded as a Fourier series with wave vectors up to $G_{max}=12a.u^{-1}$. The MT sphere radii (R) used were 2.35a.u. for Co, 2.35a.u. for Cr and 2.21a.u. for Ge. The number of k-points used in the irreducible part of the Brillouin zone is 286.

Crystal structure: Heusler alloy [9] with chemical formula X₂YZ (X = Co, Y = Cr and Z = Ge). The full Heusler structure consists of four penetrating fcc sublattices with atoms at X₁(1/4, 1/4, 1/4), X₂(3/4, 3/4, 3/4), Y(1/2,

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 1/2, 1/2) and Z(0, 0, 0) positions which results in L_{21} crystal
 structure having space group Fm-3-m as shown in Fig. 1.

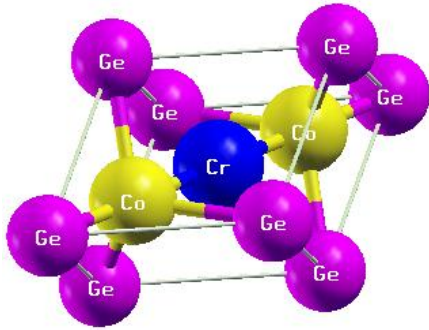


FIGURE 1. Unit Cell Structure of Co₂CrGe.

III. RESULTS AND DISCUSSIONS

The volume optimization was performed using the lattice constant by taking the experimental one. The calculated total energies within GGA as function of the volume were used for determination of theoretical lattice constant and bulk modulus. The bulk modulus was calculated using the Murnaghan's equation of state [10]. The calculated values of lattice constant and bulk modulus are presented in Table I.

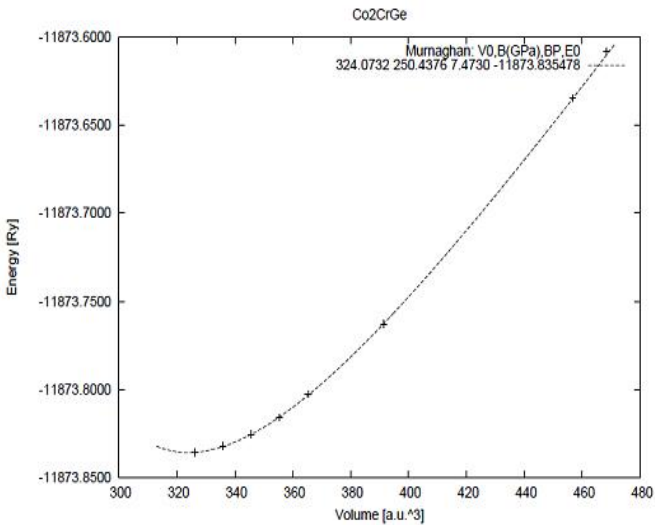


FIGURE 2. Volume optimization of Co₂CrGe.

TABLE I. Lattice constant and Bulk modulus.

Lattice Constants a_0 (Å)			Bulk Modulus B(GPa)	Equilibrium Energy (Ry)
Previous	Calculated	$\Delta(a_0)$		
5.740[11]	5.770	0.03	250.437	-11873.836

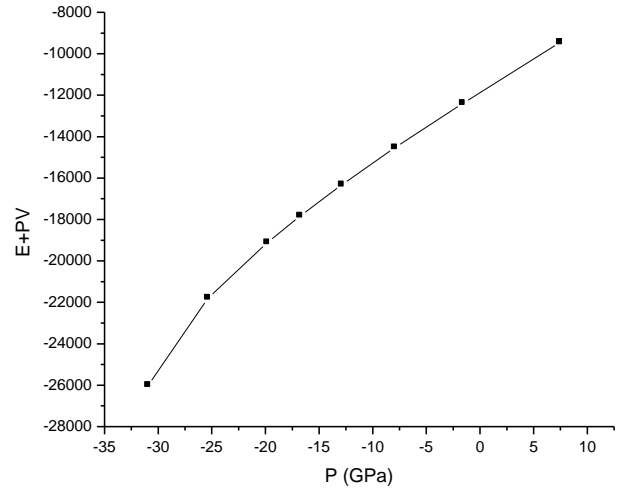


FIGURE 4. Enthalpy versus Pressure derivative.

The calculated bulk modulus is 250.4376GPa and its pressure derivative is found to be 7.4730 for Co₂CrGe. The optimized lattice constants for Co₂CrGe is 5.770Å and the change in the lattice constant of Co₂CrGe with that of experimental one is 0.030. The optimized lattice parameters were slightly higher than the experimental lattice parameters, the change in lattice parameter is given by $\Delta(a_0)$. As shown in above Figs. (2, 3) the volume derivative decreases with the increase in pressure on the other hand the enthalpy (H) is increasing and finally reaches -9452.0364Ry at stable volume 324.0732a.u.⁻³ and pressure derivative of 7.4730. The values of total and local moments are given in Table III in comparison with the earlier results. In order to understand the formation of magnetic properties, it is necessary to consider their density of states (DOS) and band structure.

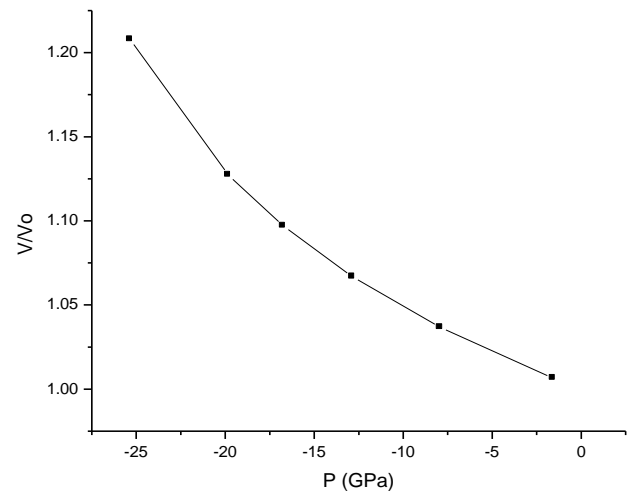


FIGURE 3. Change in the volume versus pressure derivative.

A. Spin Polarization and half-metallic ferromagnets

The electron spin polarization (P) at Fermi energy (E_F) of a material is defined by Eq. (1) [12].

$$P = \frac{\rho \uparrow(E_F) - \rho \downarrow(E_F)}{\rho \uparrow(E_F) + \rho \downarrow(E_F)}. \quad (1)$$

Where $\rho \uparrow(E_F)$ and $\rho \downarrow(E_F)$ are the spin dependent density of states at the E_F . The \uparrow and \downarrow assigns the majority and the minority states respectively. P vanishes for paramagnetic or anti-ferromagnetic materials even below the magnetic transition temperature. It has a finite value in ferromagnetic materials below Curie temperature [13]. The electrons at E_F are fully spin polarized ($P=100\%$) when $\rho \uparrow(E_F)$ or $\rho \downarrow(E_F)$ equals to zero. In present work, we have studied the Co_2CrGe system which shows 100% spin polarization at E_F [TABLE II]. According to our result, the compound Co_2CrGe is interesting as it shows large DOS at the E_F of $\rho \uparrow(E_F)=3.00$ states/eV [TABLE II]. The reason for this large value is that E_F cuts through strongly localized states of Cr-d whereas the contribution of Co-d states to $\rho \uparrow(E_F)$ is very small as illustrated in Fig. 4(b). On the other hand $\rho \downarrow(E_F)=0.00$ states/eV for both Co and Cr atoms according to this Co_2CrGe is a half-metal which gives 100% spin polarization at E_F .

TABLE II. Energy gap and Spin polarization.

Energy gap E_g (eV)			Spin Polarization		
$E_{\max}(\Gamma)$	$E_{\min}(X)$	ΔE	$\rho \uparrow(E_F)$	$\rho \downarrow(E_F)$	P%
0.24	0.00	0.24	3.00	0.00	100

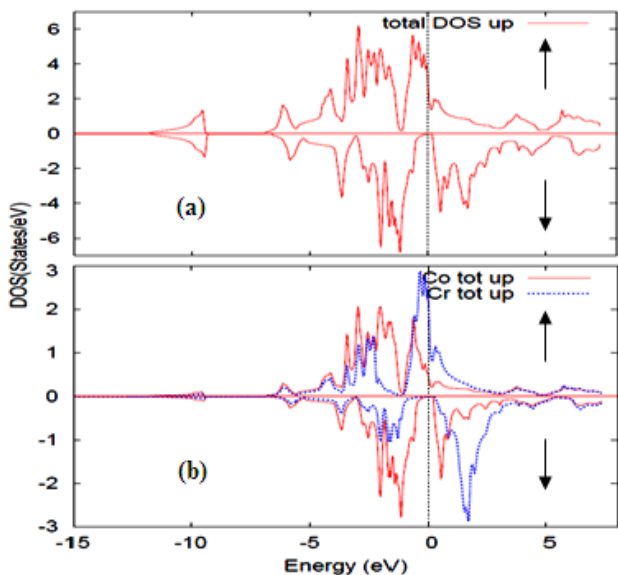


FIGURE 5. Total DOS of Co_2CrGe .

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 Figs. (5, 6) summarizes the results of the DOS which were calculated using LSDA. It is shown in Figs. 6(a, c) the majority contribution of DOS is from the d states of Co and Cr atoms. The Cr-d gives almost an exchange splitting type pattern as shown in Fig. 6(c). The sharp peaks appear at the fermi level in spin up region for Cr-d atoms. The contribution of Co-d atoms are very small at the conduction region. The hybridization between the Co-d and Cr-d atoms are appear between 3.5eV and 5.5eV, which responsible for the creation of magnetic moment. According to Fig. 7 the indirect band gap along the Γ -X symmetry for Co_2CrGe is 0.24eV. For Co_2CrGe the Fermi energy (E_F) lies in the middle of the gap of the minority-spin states, determining the half-metal character [Fig. 5(a)]. The formation of gap for the half-metal compounds was discussed by Galanakis *et al.* [14] for Co_2MnSi , is due to the strong hybridization between Co-d and Mn-d states, combined with large local magnetic moments and a sizeable separation of the d -like band centers.

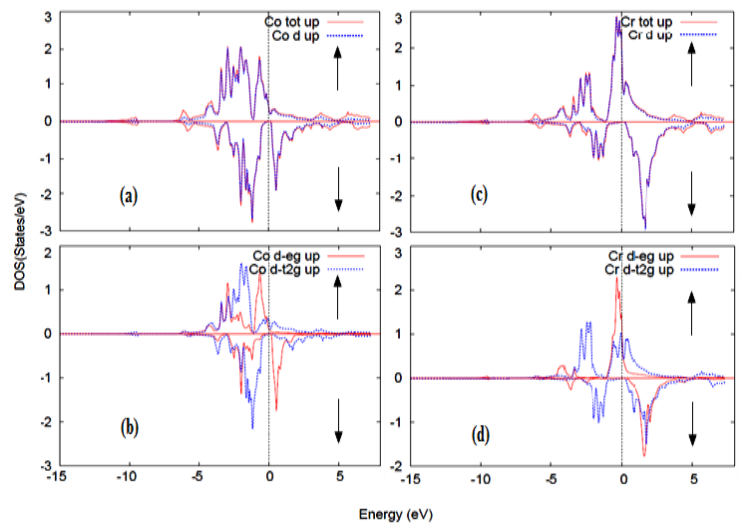


FIGURE 6. Partial DOS of Co and Cr atoms.

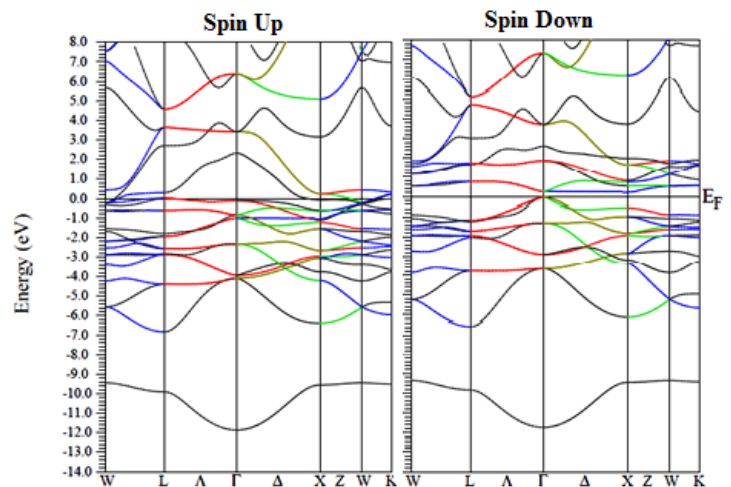


FIGURE 7. Band Structure.

B. Magnetic properties calculated in the LSDA

Starting with the compound under investigation, all the information regarding the partial, total and the previously calculated magnetic moments are summarized in Table III. It is shown in Table III the calculated total magnetic moment is almost an integer value in case of Co_2CrGe as expected for the half-metallic systems. In most cases the calculated magnetic moments are in good agreement with the previous results. We have found that the Co sites contribute much less compared with the Cr sites this is because Cr-d states shows exchange splitting. The same observation was also reported and explained by Kandpal *et al.* [15] because of the indirect connection between the specific magnetic moment at Co and the hybridization arising from the interaction between the electrons at the Co sites with the neighboring electrons in the Co t_{2g} states. As shown in Table III the Ge atom carry a negligible magnetic moment, which does not contribute much to the overall moment. We have also noticed that the partial moment of Ge atoms aligned anti-parallel to Co and Cr moments of the systems. It emerges from the hybridization with the transition metals and is caused by the overlap of the electron wave functions. The small moments found at the Ge sites are mainly due to polarization of these atoms by the surroundings, magnetically active atoms as reported by Kandpal *et al.* [15].

TABLE III. Total and partial magnetic moments.

Magnetic Moment μ_B of Co_2CrGe				
Previous	Calculated			
	Co	Cr	Ge	Total
4.00[11]	0.932	2.122	-0.029	3.999

IV. CONCLUSIONS

We have performed the total-energy calculations to find the stable magnetic configuration and the optimized lattice constant. The DOS, magnetic moments and band structure of Co_2CrGe were calculated using FP-LAPW method. The calculated results were in good agreement with the previously calculated results. For Ferromagnetic compounds the partial moment of Ge being very small and the contribution is very less in the total magnetic moment. We have investigated the possibility of appearance of half-metallicity in the case of the full Heusler compound Co_2CrGe which shows 100% spin polarization at E_F . The existence of energy gap in minority spin (DOS and band structure) of Co_2CrGe is an indication of being a potential HMF. This is also evident from the calculated magnetic moment for Co_2CrGe is 3.9999 μ_B . The calculated result is in qualitative agreement with the integral value, supporting the HMF.

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