Study of Heusler compounds Co2YSi (Y = Mn, Cr) using a full potential linearized augmented plane wave (FP-LAPW) method

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Abstract: We have performed the volume optimization followed by the calculation of electronic structure and magnetic properties on Co_2MnSi and Co_2CrSi. The structure optimization was based on generalized gradient approximation (GGA) method. The calculation of electronic structure was based on full potential linear augmented plane wave (FP-LAPW) method and exchange correlation. Results of density of states (DOS) and band structures shows the half-metallicity of Co_2MnSi and Co_2CrSi with an integer value of magnetic moments 5.031 μ_B and 4.006 μ_B respectively which follows the Slater-Pauling rule.

Key words: GGA, LSDA, half-metallicity, DOS and band structure.

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I. Introduction

Half-metallic ferromagnets (HMFs), is a magnetic material where the majority spin band is metallic and the minority–spin band is semiconducting with an energy gap at the Fermi level. In present scientific research of material science, the half-metal ferromagnets have become one of the most studied classes of materials. The existence of a gap in the minority-spin band structure leads to 100% spin polarization of the electron states at the Fermi level which makes the systems applicable for the developing field of spintronics [1]. In half-metals, the creation of a fully spin-polarized current should be possible, that should maximize the efficiency of magnetoelectronics devices [2]. Materials having high spin polarization can be used for tunnel magnetoresistance(TMR) and the giant magnetoresistance (GMR) [3]. The Co-based Heusler alloys Co_2YZ (Y: transition metal, Z: *sp* atom) are the most prospective candidates for the application in spintronics. This is due to a high Curie temperature beyond room temperature and the simple fabrication process such as dc magnetron sputtering in Co_2YZ [4]. Sandeep *et al.* [5, 6] studied the electronic properties of compounds like $NdCrSb_3$, $SmCrSb_3$ and $GdCrSb_3$ by first principles method. Ghimire *et al.* [7, 8] studied the electronic properties of CrO_2 using density functional theory and also studied the electronic and optical properties of $SbTaO_4$. Rai *et al.* [9, 10] investigated the ground state of Co_2MnAl and Co_2CrSi using LDA+U and LSDA method respectively and reported the half-metallicity. Rai and Thapa have also investigated the electronic structure and magnetic properties of X_2YZ (X = Co, Y = Mn, Z = Ge, Sn) type Heusler Compounds by using a first Principle Study and reported HMFs [11]. Rai et al. (2012) also studied the electronic and magnetic properties of Co_2CrAl and Co_2CrGa using both LSDA and LSDA+U and reported the increase in band gap, hybridization of *d-d* orbitals as well as *d-p* orbitals when treated with LSDA+U [12]. In this paper we have studied the full-Heusler compounds Co_2MnSi and Co_2CrSi within FP-LAPW method. Our main aim is to investigate the half-metallic behaviour of these compounds, which are a better prospective for the spintronic devices.

II. Crystal structure and Calculation Details

2.1 Crystal structure: Heusler alloy [13] with chemical formula Co_2YSb (Y = Sc, Ti), has full Heusler structure with four penetrating fcc sublattices with atoms at X1(1/4,1/4,1/4), X2(3/4,3/4,3/4), Y(1/2,1/2,1/2) and

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

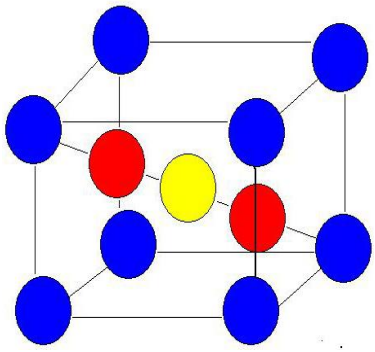

Fig. 1: Unit cell Structure of Co_2YSi: Co (red), Y (yellow) and Si (blue) atoms.

2.2 Calculation Details: The FP-LAPW method (WIEN2K) [14] was applied to band structure calculations of Co_2CrGe. In this method the space is divided into non-overlapping muffin-tin (MT) spheres separated by an interstitial region. The basis functions are expanded into spherical harmonic functions inside the muffin-tin sphere and the Fourier series in the interstitial region. The convergence of basis set was controlled by a cutoff parameter $R_{MT} \times K_{max} = 7$ where R_{MT} is the smallest of the MT sphere radii and K_{max} is the largest reciprocal lattice vector used in the plane wave expansion and made the expansion up to $l_{max} = 6$ in the muffin tins, where l_{max} is the maximum value of angular momentum. The magnitude of the largest vector in charge density Fourier expansion (G_{max}) was 12 $a.u^{-1}$. The cutoff energy which defines the separation of valence and core states was chosen as -6.0 Ry. For k-point sampling, a 21x21x21 k-point mesh in the first Brillouin zone was used. LSDA [15] was used and the convergence criterion for self–consistence calculations was set up to charge convergence equal to 10^{-4}. In the interstitial region the charge density and the potential were expands as a Fourier series with wave vectors up to G_{max}=12 $a.u^{-1}$. The MT sphere radii(R) used were 2.35 a. u. for Co, 2.35 a.u. for Cr and 2.21 a. u. for Si and in case of Co_2MnSi, 2.34 a. u. for Co, 2.34 a.u. for Mn and 2.20 a. u. for Si. The number of k-points used in the irreducible part of the brillouin zone is 286.

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 [16]. The calculated values of lattice constant and bulk modulus are presented in Table 1.

Table 1: The previous lattice constant, calculated lattice constant and bulk modulus

Compound	Lattice constant a_o (Å)		Bulk modulus (GPa)
	Previous	Our Calculation	
Co_2MnSi	5.645[18]	5.665	866.499
Co_2CrSi	5.647[17]	5.699	405.556

Fig-2 shows the DOS of Co_2MnSi, at -1.4 eV below E_F in spin down and spin up regions are contributed by Mn-d atoms but Mn atoms has less contribution in the spin down region. Similarly at -1.4eV, Co-*d* atoms also contributed in both spin up and spin down regions. At 3.2eV and 4.2eV in spin down region both Mn-d and Co-*d* atoms contributed to the total DOS. But negligible contribution from Co and Mn atoms in the spin up channel above E_F. It is shown in Fig-3 that the hybridization takes place between the Co-*d*-*eg* states and Mn-*d*-*eg* states below E_F in the spin up channel, which is responsible for contribution of magnetic moments. The exchange splitting occurs between the Mn-*d* electrons at -1.8 eV and 1.8 eV is also responsible for the creation of gap in the spin down channel shown in Fig-2(b). From Fig-4(b), in Co_2CrSi peaks due to Co atoms are found in the valence region for both the spins. In the conduction region sharp peaks were observed at 0.70 eV and 1.8 eV which were contributed by Co-*d states*. An exchange splitting also occurs between Co-d up and down with a splitting energy of 1.4 eV. In spin down channel, Cr-*d* contributes both in the valence and conduction regions. We have observed two sharp

peaks -1.4 eV and -2.0 eV in valence region due to Cr-*d* states. An exchange splitting of 2.4 eV is observed between spin down and spin up channel due to Cr-*d* states. This exchane splitting is responsible for the creation of energy gap as well as magnetic moment. The total magnetic moments and energy gaps of Co_2MnSi and Co_2CrSi are given in Table 2.

Table 2: The magnetic moments and energy gap are given below.

Compounds	**Magnetic Moment μ_B**		**Energy gap E_g (eV)**	
	Previuos	**Our Calculation**	**Previous**	**Our Calculation**
Co_2MnSi	4.900[17]	5.031	0.798[18]	0.760
	5.000[18]			
Co_2CrSi	4.000[17]	4.006	0.878[17]	0.910

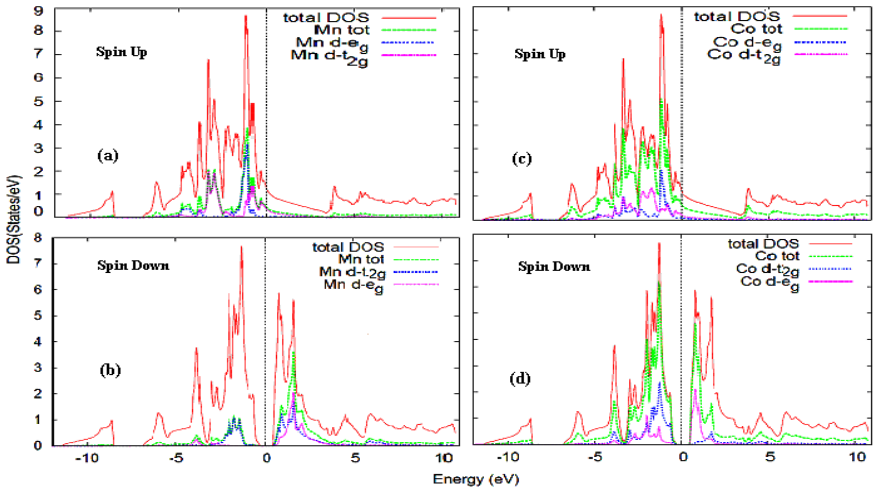

Fig. 2: (a) DOS plot for Mn atoms in spin up (b) DOS plot for Mn atoms in spin down (c) DOS plot for Co atoms in spin up and (d) DOS plot for Co atoms in spin down.

The partial magnetic moments of the atoms Co, Mn and Si are 1.029 μ_B, 3.058 μ_B and -0.055 μ_B respectively. Thus the total magnetic moment is 5.031 μ_B which is approximately an integer value 5.00 μ_B [18]. There are 9 electrons in the valence shell of Co atoms, 7 electrons for Mn atom, 6 electrons for Cr atom and 4 electrons for Si atom. According to Slater-Pauling rule [19] the magnetic moment of Co_2MnSi is 2x9+7+4-24= 5 μ_B, similarly 4 μ_B for Co_2CrSi which are consistent with our results. The total and partial magnetic moments of Co_2MnSi and Co_2CrSi are tabulated in Table 3.

Table 3: The partial and total magnetic moments are tabulated and compared with the previous results.

Compounds	**Previous Magnetic Moment μ_B**			**Calculated Magnetic Moment μ_B**			
	Co	**Y**	**Total**	**Co**	**Y**	**Si**	**Total**
Co_2MnSi	1.06	2.99	5.00[18]	1.03	3.06	-0.06	5.031
Co_2CrSi 0.98	2.08		4.00[17]	0.98	2.10	-0.505	4.006
	1.00	2.03	4.00[18]				

3.1. Band Structure

For overall band structure calculation of Co_2MnSi compounds there exist an energy gap E_g in the spin down region. The width of the energy gap E_g is the difference of the energies of the highest occupied band in the valence region at the Γ-point and the lowest unoccupied band in the conduction region at the X-point, thus it is an indirect gap. With the help of the DOS, it is clear that the energy region lower than -3eV consists mainly of *s* and *p* electrons of the Si atoms which is called the core and the energy region between -3eV and 2eV consists mainly of the *d-electrons* of Co and Mn atoms. Based on the analysis of the band-structure calculation it was shown that the *3d* orbitals of Co atoms from two different sub-lattices, $Co^1(0, 0, 0)$ and $Co^2(1/2, 1/2, 1/2)$, couple and form bonding hybrids $Co^1(t_{2g}/e_g)$ -$Co^2(t_{2g}/e_g)$. In other words, the t_{2g}/e_g orbitals of one of the Co atoms can couple only with the t_{2g}/e_g orbitals the other Co atom. Furthermore, the Co-Co hybrid bonding orbitals hybridize with the Mn(d)-t_{2g},e_g manifold, while the Co-Co hybrid antibonding orbitals remain uncoupled owing to their symmetry. The Co-Co hybrid antibonding t_{2g} is situated below the Fermi energy E_F and the Co-Co hybrid antibonding e_g is unoccupied and lies just above the Fermi level.

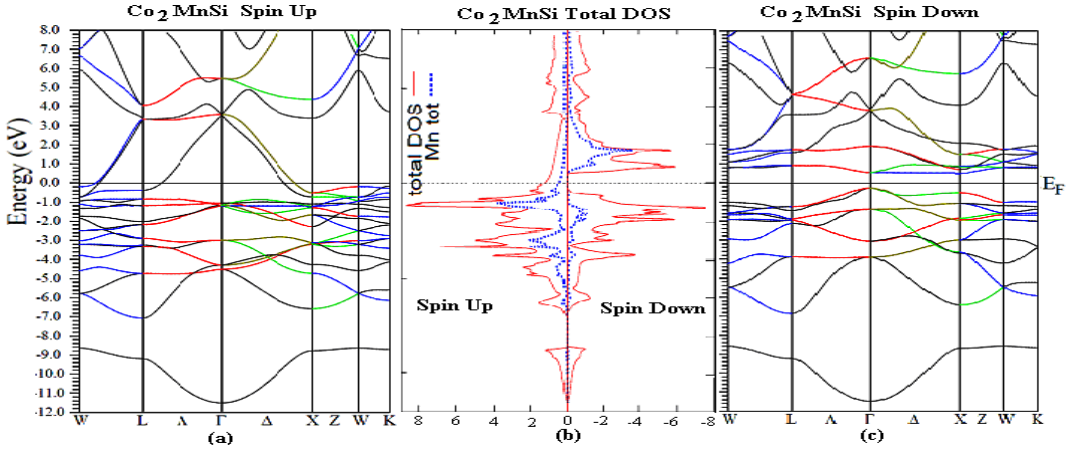

Fig. 3: (a) Energy band of Co_2MnSi for spin up (b) Total DOS of Co_2MnSi and (c) Energy band of Co_2MnSi for spin down.

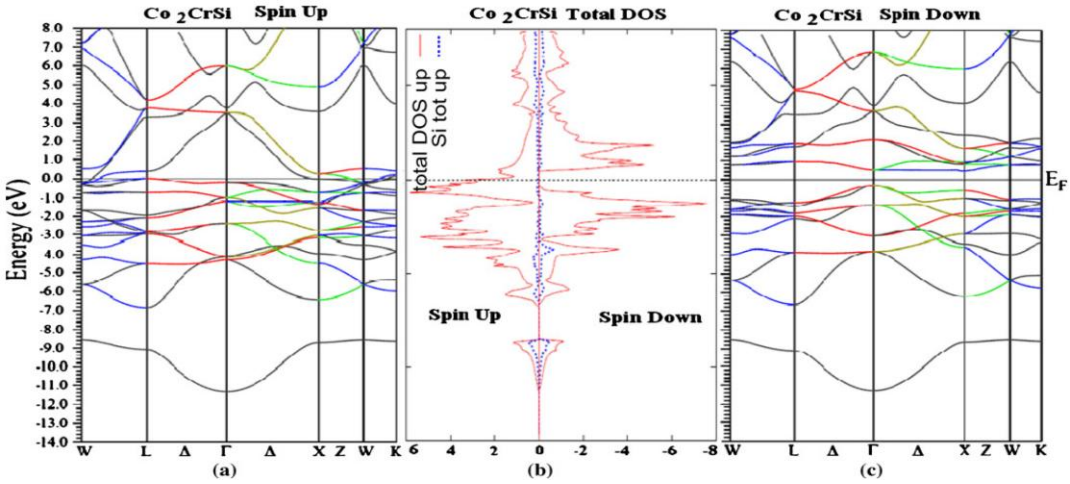

Fig. 4: (a) Energy band of Co_2CrSi for spin up (b) Total DOS of Co_2CrSi and (c) Energy band of Co_2CrSi for spin down.

Thus due to the missing Mn*(d)*-t_{2g},e_g and Co-Co hybrid antibonding hybridization, the Fermi energy is situated within the minority gap formed by the triply degenerate Co-Co antibonding t_{2g} and the doubly degenerate Co-Co antibonding e_g. From Figs-3 (a-c) it is clearly shown that the E_F lies exactly at the middle of the gap between the valence and the conduction bands in spin down region where as there is no such gap occur in the spin up region. Thus there exist a half metallicity in Co_2MnSi. The value of energy gap between Γ and X is 0.760 eV which is an indirect band gap. Almost similar results of 0.798 eV had been reported by Kandpal *et al.* (2006) [18]. From Figs-4(a-c) for Co_2CrSi, we have observed an indirect band gap in spin down channel. The calculated energy gap along Γ-X symmetry is 0.91 eV which is almost close to 0.878 eV [17].

IV. Conclusions

It can be said that due to the existence of gap in DOS for the minority spins, Co_2MnSi and Co_2CrSi is a potential half-metallic ferromagnet. This is also evident from the energy band results as discussed. The calculated magnetic moment for Co_2MnSi is 5.031 μ_B and 4.006 μ_B for Co_2CrSi which is equal to an integer value and follows the Slater-Pauling rule. The integral value of magnetic moment is also one of the evidences for the half metallicity. Due to these characteristics like integer value of magnetic moment, 100% spin polarization at E_F and the energy gap at the Fermi level in spin down channel makes application of half-metallic ferromagnets very important. The Co-based Heusler alloys Co_2YZ (Y is transition elements and Z is the sp elements) are the most prospective candidates for the application in spintronics. This is due to a high Curie temperature beyond room temperature and the simple fabrication process such as dc-magnetron sputtering in Co_2YZ.

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