Magnetic and Half-Metallic Properties of Full-Heusler Alloys Co_2TiZ (Z= Ga, Si, Sn) and Co_2MnZ (Z= Ga, Si)

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The electronic structure and magnetic properties of the full-Heusler alloys Co2TiZ and Co2MnZ (Z element from groups III and IV) were studied by first principle calculations. In this paper it is shown that Co2TiSi, Co2TiSn, and Co2MnSi are half metallic ferromagnets. The magnitude of the total magnetic moment shows a trend consistent with Slater Pauling behavior. The total magnetic moment also depends on the kind of Z atoms although they do not directly contribute to it.

1. Introduction:

Full Heusler alloys are ternary intermetallic with a 2: 1: 1 stiochiometry and chemical formula X2YZ, where X and Y are transition elements and Z is a main group element. They crystallize in the L21 structure which consists of four fcc sub-lattices. They first attracted the interest of the magnetim community when Heusler et al [1] showed that Cu2MnAl was ferromagnetic even though none of its constituents is ferromagnetic by itself. The main interest during the first deaceds after their discovery was concentrated on Cu and Mn containing alloys. Then Kubler et al [2] recognized that minority spin densities at the Fermi level (EF) nearly vanish for Co2MnAl and Co2MnSn. They concluded that this should lead to peculiar transport properties in these Heusler compounds because only the majority density contributes to the states at EF. At the same time de Groot et al [3] predicted that some half-Heusler alloys such as NiMnSb and PtMnSb exhibit half-metallic ferromagnetism, in which electrons of one spin have metallic bands electrons of the opposite spin are insulating. This lead to 100% spin polarization at the Fermi level [4-6] and thus electric conduction takes place with electrons of one spin exclusively. This prediction has been verified also by other authors [7] and the half-metallic ferromagnetic character has been also well established experimentally both by using positron annihilation experiments [8] or inverse photoemission [9]. A half metallic ferromagent act as a spin filters which provides current with high degree of spin polarization. Therefore intensive efforts are made to find half metallic compounds for possible applications in spintronics and spin injection.

Ferromagnetic Co-based Heusler compounds have attracted increasing interest because of their peculiar electronic structure. Further more they cover a wide range of magnetic moments and Curie temperatures (for example Co2MnSi has a Tc = 985K, [10]). In this paper we focus our attention on Co2YZ, Y = Ti and Mn, and Z is an element from group III-IV. We examine the effects of changing the Y and Z elements on the electronic and magnetic structure and of these alloys.

2. Cmputational Details:

In this work the electronic structure calculations are carried out using the EIEN2K package [11]. The theoretical lattice constants were obtained by minimizing the total energy as a function of volume. The atomic sphere radii were taken as 2.00 a.u for Si, 2.20 a.u for Co, Ti, and Mn, and 2.30 a.u for Ga and Sn. The Perdew-Burke-Ernzerof generalized gradient approximation (GGA) potential [12] was used for exchange correlation potential and density of states was calculated using the modified tetrahedron method [13]. The total energy calculations were carried out as spin polarized calculations without spin orbit coupling. 72 k points in the irreducible Brillouin Zone were used.

3. Results and Discussion:

Figure (1) shows majority and minority total DOS of Co_2TiGa. In majority spins metallic behavior is observed. In minority spins the valence bands decrease rapidly below E_F but it does not reach zero DOS, above the E_F the conduction bands starts with very low vanishing DOS producing a small gap of width 0.030 eV the range (+0.109 to +0.136 eV) then it increases rapidly giving a clear peak. The high valence peaks are mainly Co and the small peaks at the bottom of he valence bands are mainly Ga, above E_F Co and Ti equally share forming the conduction bands however the lowest conduction peak is mostly because of the Co, this goes in both spins.

Figure (3) shows the majority and minority energy bands of Co_2 TiSia and Co_2 TiSn along the high symmetry lines. The energy bands of the three alloys exct metallic behavior in majority spins. In the minority spins Co2TiGa has a gap above E_F and Co_2TiSi and Co_2 TiSn have gaps around E_F in the ranges (-0.535 to +0.117eV) and (-0.184 to 0.197eV) of width 0.953 eV and 0.327eV, respectively. Figures (3b. and c) exhibit high degree of similarity between the energy bands of Co_2Ti Si and Co_2TiSn and they are fairly similar to the bands of Co_2TiGa, Fig. (3a) this similarity indicates the small rule of the Z atom in the formation of the electronic structure of these alloys. The main differences are observed at the position of the lowest band which is mostly because of the s-state of the Z element and at the top of the conduction band which is quite

controlled by the p-state. But the main effect of the Z atom is observed in the change of the position and width of the minority gap.

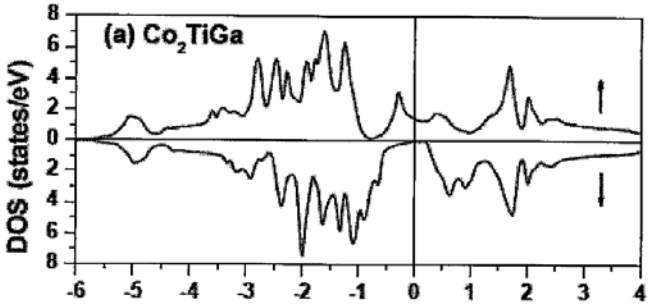

Figure 1 Majority (↑) and minority (↓) total DOS of Co_2TiGa

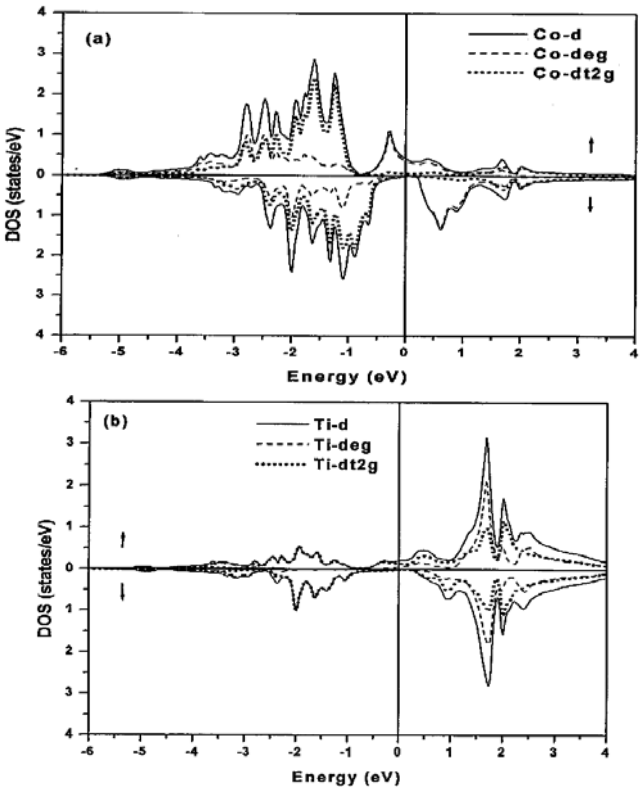

Fig. (2): Majority (↑) and minority (↓) Partial DOS of (a) Co-d states and (b) Ti-d states in Co_2TiGa. The contributions from de_g and dt_{2g} states are shown by the broken and dotted curves.

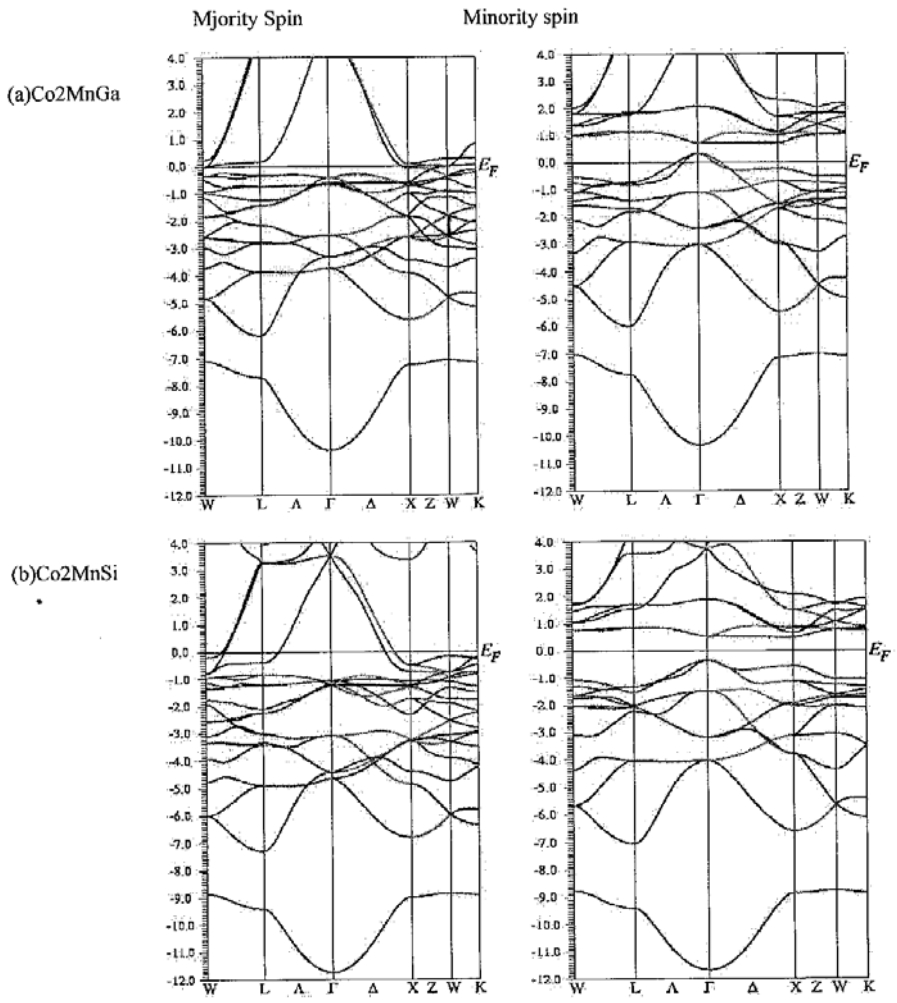

Fig. (3): Majority and minority Energy band structure of (a) Co_2TiGa, (b)Co_2TiSi, and (c) Co_2TiSn.

By studying the partial DOS of Co-d and Ti-d states, as in Fig. (2), it is observed that in Co_2TiZ the d bands of Co are mainly occupied for both spin states and are characterized by the peak near the Fermi level which comes from Co-dt_{2g} states. This is in agreement with Ishida et al – 1982, who studied the electronic structure of Co_2TiZ, (Z= Al and Sn) using the Spherical Augmented Plane wave method (SAPW). Most of Ti-d states are not occupied and that Ti atoms have small magnetic moments in the three alloys. The peaks of the partial DOS show the hybridization between the Co-d and Ti-d states.

Our DOS calculation of the gap width and position show that Co_2TiGa is nearly half-metal and Co_2TiSi and Co_2TiSn are half metals This is in agreement with the previous calculation of Kandpal et al – 2007, and the claulation performed by Lee et al – 2005.

The valence-electron density and spin density maps in the [110] plane are constructed below for Co_2TiGa, Co_2TiSi and Co_2TiSn in Fig. (7). The spin density maps show that only Co atoms have significant magnetic moment. The calculated magnetic moments are presented in table 3-4, they support the spin maps plots and they are in fair agreement with the experimental moments for Co_2TiGa and Co_2TiSn.

CoMnZ Compounds:

Figure (4) shows majority and minority total DOS of Co_2TiGa. In majority spins; metallic behavior is clear. In minority spins; there is a gap of width 0.245eV above E_F in the range (+0.372 to +0.618 eV). In the majority spin, Both Co and Mn share forming Co_2TiGa the valence bands and the small conduction bands. In the minority spin Co states are dominant in the valence bands and Mn states are dominat in the conduction bands.

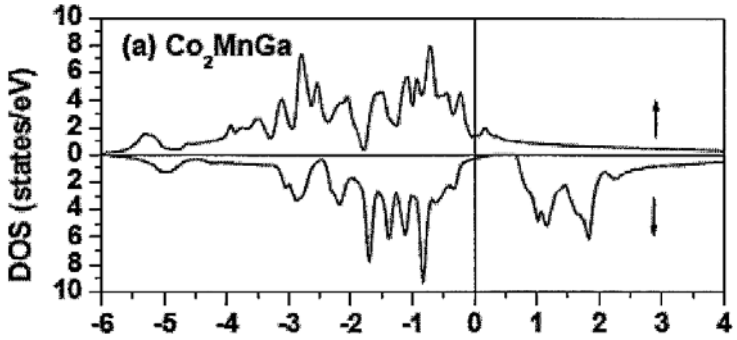

Figure 4 Majority (↑) and minority (↓) total DOS of Co_2MnGa.

Figure (6) shows majority and minority energy bands of Co_2MnGa and Co_2MnSi at the symmetry points of the Brillion zone, in the range – 12 eV +4 eV. In majority spins, thick bands of metallic character are observed below and slightly above. E_F. Co_2MnGa has a gap in the minority spins surrounding E_F at all the symmetry points except at F, where the gap is the narrowest and bands extend above E_F, the bands that cross E_F are mainly Codt_{2g} and Mn dt_{2g}.Co_2MnSi has a gap in the minority spin of width 0.681 eV in the range (-0.230 to + 0.381 eV). The presence of E_F away from the edges of the gap grantees the stability of the half-metallicity of this compound. The band

structures of these two alloys are very similar except at the position of the lowest band which has an s character.

By studying the partial DOS of Co-d and Mn-d states of these two alloys, for example as in Figure (5), it is shown that in majority spins Co-de_g and $codt_{2g}$ equally share forming the DOS of the d-states, which means they almost have the same electron occupancy. In minority spins; The DOS refer to more electron occupancy in the Co-dt_{2g} states and more magnetic moment on Co-de_g Most of Mn of Mn bands, in the majority spins are occupied. In minority spins; Mn has a small DOS in the valence bands and high DOS in the conduction bands. Comparing majority and minority DOS indicaties a high positive magnetic moment on Mn. The DOS of Co-d and Mn-d refer to the strong hybridization between them which is very clear in minority spins, in agreement with Kulkova et al 2004.

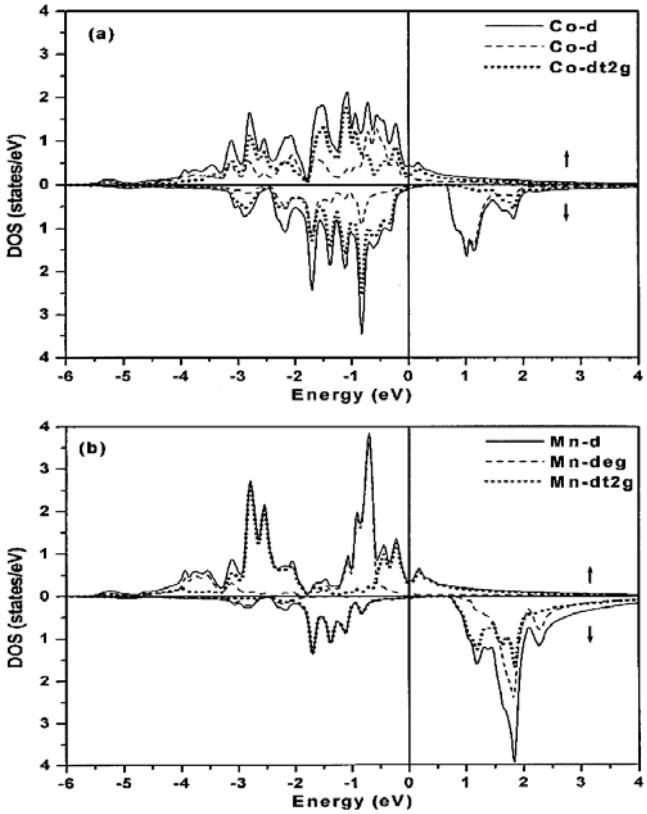

Fig. (5): Majority (↑) and minority (↓) Partial DOS of (a) Co-d states and (b) Mn-d states in Co_2MnGa. The contributions from de_g and dt_{2g} states are shown by the broken and dotted curves.

Co_2MnGa is a nearly half metal that the Fermi level falls within a region of very small spin down DOS. This does not agree with Ishida et al – 1982 who recorded this alloy as a half metallic ferromagnet. Co_2MnSi is a half metal. Our results for these two compounds agree with Galanakis et al, - 2006 and Kandpal et al, - 2006.

The valence electron density and spin density maps in the [110] plane are constructed below for Co_2MnGa and Co_2MnSi, in Fig. (8). They show that Mn atoms have higher magnetic moment than Co atoms. The calculated magnetic moments are presented in Table (1), they support the spin maps plots and they are in good agreement with the experimental measured moments.

Table (1): Experimental and calculated lattice constants, local moments on the atoms and total magnetic moments as calculated with the WI2N 2K, the experimental magnetic moment s and the with of the gap in Co_2YZ Heusler alloya.

Alloys	A_{exp}(Å)	$A_{calc.}$(Å)	Co-μ_B	Y-μ_B	Z-μ_B	Total-μ_B	Exp-μ_B	Gap Width(eV)
Co_2TiGa	5.848[a]	5.837	0.639	-0.139	-0.012	1.001	0.75 a,f	
Co_2TiSi	5.743[b]	5.762	1.032	-0.010	0.019	1.99997	1.09b	0.653
Co_2TiSn	6.077[c]	6.094	1.079	-0.058	0.006	2.000	1.93 [g,]	0.327
Co_2MnGa	5.767[b]	5.718	0.754	2.750	-0.072	4.091	4.05 [h,c]	
Co_2MnSi	5.645[e]	5.641	1.061	2.961	-0.040	5.000	4.90 [h,k]	0.680

[a] Reference 14 [b] Reference 15 [c] Reference 16
[d] Reference 17 [e] Reference 18 [f] Reference 19
[g] Reference 20 [h] Reference 21 [j] Reference 22
[k] Reference 23

Effect of Changing Y atom:

Looking though the band structure graphs of Co_2YZ, Fig. (3 and 6), shows the essential rule of Y atoms in the formation of the electronic structure of Heusler alloys. Comparing the graphs of the partial DOS of Co and Y atoms indicates that strong hybridization take place between the Co and Y atoms that produces new arrangements for electrons occupancy in the energy bands. In case of Y=Mn large exchange splitting occurs is the Y site causing the large magnetic moment on Mn atoms.

For all of the studied compounds it is observed that:

- Replacement of Ga (3^{rd} group element) by Si or Sn (4^{th} group elements) result in; increasing the calculated total magnetic moment and the magnetic moment of the Co atoms.
- The calculated total magnetic moments with The Wien2K, table 1, agree with the magnetic moments which are obtained in case of using the Slater Pauling behavior [25].

$$M_t = Z_t - 24 \qquad (1)$$

M_t : total magnetic moment, Z_t : number of valence electrons.
This is in agreement with Kandpal et al – 2006. Total and partial moments of Co2TiZ are in a good agreement with the moments calculated with Lee et al – 2005.

- The total Calculated magnetic moments are almost integers which are a good indicator for the high percentage of spin polarization in these alloys, and this is a great quality for materials to be used in spintronics applications.

4. Conclusion:

Using the Full potential linearized augmented plane wave plus local orbitals, which is implemented in the WIEN2K computer package, we studied the electronic structure of Co_2YZ (Y= Ti and Mn) and (Z=Ga, Si, and Sn) full Heusler alloys. We have shown using the relativistic spin polarized calculations that of these alloys are ferromagnetic, in good agreement with the experimental measurements, and some of them are half metallic ferromagnets (Co_2TiSi, Co_2TiSn, and Co_2MnSi). These compounds show Slater Pauling behavior and the total magnetic moment per unit cell (Mt) scales with the total number of valence electrons (Z_t). Changing the Y site causes drastic effect on the electronic structure of these alloys. However, the effect of the Z site appears clearly in the width and position of the minority gap without having an essential rule in the electronic structure especially in the middle of the valence and bottom of the conduction bands.

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