



Determination of half metallicity behavior of quaternary heusler alloy crcoScga

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Abstract

In this work, the theoretical study through the calculations of the structural, electronic and magnetic properties of quaternary heusler compound CrCoScGa, using wien2k code, is reported. We adopted the generalized gradient approximation (GGA) to estimate the exchange correlation potential. The negative value of the formation energy shows that CrCoScGa compound have a high structural stability in the type III structure ferromagnetic phase, so it can be experimentally synthesized. According to our electronic calculations, CrCoScGa is a half-metallic ferromagnet (HMF) with a half-metallic gap of 0.22 eV. The total magnetic moment of 3 μ B, mainly originated from Cr and Co atoms, makes this alloy as a spintronic material.

Keywords: Half-metallicity, magnetic properties, band gap, spintronic, heusler compound.

1. Introduction

The discovery of new materials and alloys from the periodic table, based on natural law, which states that combining two different materials does not result in a combination of their properties, but rather gives rise to new characteristics specific to the alloy, is extremely crucial to the advancement of technology and industry [1]. Heusler alloys have recently sparked intense experimental and theoretical interest owing to three distinct properties: half-metallic behavior, magnetic shape memory effect, and inverse magneto-caloric effect. They were firstly discovered in 1903 by the German physicist Friedrich Heusler, who reported that the addition of sp elements (Al, In, Sn, Sb, or Bi) transforms a Cu-Mn alloy into a ferromagnetic material despite the alloy containing no ferromagnetic elements [2]. After a deep understanding of Heusler alloys's structure, it was found that Heusler alloys can be classified into: First, half-Heusler alloys with the general formula XYZ where X and Y are transition metal and Z is an sp-element. Using the Gaussian process regression model, Yun Zhang et al. investigate nearly 140 half-Heusler samples containing alloying elements of Cr, Mn, Fe, Co, Ni, Rh, Ti, V, Al, Ga, In, Si, Ge, Sn, P, As, and Sb to find statistical correlations between lattice constants a_0 of half-Heusler compounds, ionic radii, and Pauling electronegativity of their alloying [3]. Second, full-Heusler alloys with the formula X₂YZ, as an example a combinatorial search of 810 full Heusler alloys is conducted using first-

principles (GGA), total-energy calculations, pseudopotentials, and plane waves to predict their lattice parameters and magnetic moments. According to Gilleben et al., approximately 60% of the investigated intermetallics are thermochemically stable with respect to the constituent elements[4].

When one of the two X atoms in full heusler is replaced by another X' atom, a new type of heusler called quaternary heusler compounds is formed. Such compounds drew a lot of attention in spintronic application after Groot and his colleagues predicted half-metallicity in 1983, while studying the band structure of a half-Heusler alloy NiMnSb [5]. Many studies on half metallic materials have been conducted since that time. In 2020, Muhammed et al [13] studied, The structural stability, band structure, density of states (DOS), magnetic and mechanical properties of Co-based equiatomic quaternary Heusler alloys (EQHAs) CoYCrZ (Z = Si, Ge, Ga, Al), by employing 1st principle calculations Using the FP-LAPW Method. They found that these compounds are good candidate for magnetic tunnel junctions as well as spin valves. While in 2021, Ab Quayoom et al [14] showed that Quaternary Heusler alloys are a future perspective for revolutionizing conventional semiconductor technology. Recently, me and my team predicted the Ferromagnetic Half-Metallicity of the New Quaternary Heusler Alloy CoCrScIn [6]. Based on this work, we will see if the half metallicity remains active if we replace In atom by Ga atom. the purpose of our work is to study the structural, elastic, electronic and magnetic properties of new quaternary heusler compound so it can be later used in , spintronic application

2. Materials and methods

In this paper, we used the full potential and linearized augmented plane wave (FP-LAPW) method to perform first principles calculations[7], with the generalized gradient approximation (GGA) in the Perdew-Burke-Ernzerhof (PBE) scheme implemented in the WIEN2k code[8]. Wave functions, electron densities and potential are developed in combination of spherical harmonics around atomic sites in the Muffin-tin spheres and Fourier series in the interstitial region with a radius of cut-off $R.K_{max}$. To fully describe the studied system, the convergence of $R.K_{max}$ and K_{points} have been tested. The obtained value of the number of K_{points} and $R.K_{max}$, ensuring the convergence of the total energy of the system, are found to be 3000 and 8, respectively. During the self-coherence cycles, the cut-off energy, which defines the separation of the valence and cores states, was chosen to be -6 Ry, and the charge convergence was chosen to be 0.00001.

3. Results and discussions:

3.1. Structural properties:

The most crucial step in ab-initio calculation is the determination of the structural properties of a given system on its ground state, which will enable us later to determine other important physical properties. This structural type has three possible nonequivalent superstructures based on the different positions of the Cr, Co, Sc and Ga atoms, as shown Table 1. The total energy as a function of volume in the structural (type I, type II, type III) and magnetic (Nonmagnetic NM, Ferromagnetic FM, *Antiferromagnetic* AFM) configurations. $E(V)$ is illustrated in Figure 1. Table 2 shows the theoretical results of this work, such as the equilibrium lattice parameter a_0 , the compressibility modulus B and its derivative. The total energy of ground state, formation energy and Debye

temperature of CrCoScGa compound are listed in table 3. These parameters are obtained by adjusting the Murnaghan equation of state [9]:

$$E(V) = E_0 + \frac{9V_0B_0}{16} \left\{ \left[\left(\frac{V_0}{V} \right)^{\frac{2}{3}} - 1 \right]^3 B_0 + \left[\left(\frac{V_0}{V} \right)^{\frac{2}{3}} - 1 \right]^2 \left[6 - 4 \left(\frac{V_0}{V} \right)^{\frac{2}{3}} \right] \right\} \quad (1)$$

Where $E(V)$ represents the energy of the ground state with respect to volume V . V_0 is the unit cell volume at zero pressure, corresponding to the equilibrium lattice parameter a_0 , while B_0 and B'_0 are the compressibility modulus and its derivative, respectively.

From figure 1, we can clearly see that the most fundamental state is the Type III in its FM phase. The formation energy E_f determines whether or not a compound can be experimentally synthesized. E_f denotes the change in energy that occurs when a material is formed from its constituent elements in their respective states of volume. It can be calculated for the CrCoScGa compound as follows:

$$E_{for} = E_{CrCoScGa}^{tot} - (E_{Cr}^{bulk} + E_{Co}^{bulk} - E_{Sc}^{bulk} + E_{Al}^{Bulk}) \quad (2)$$

Where $E_{CrCoScAl}^{tot}$ is the total energy of CrCoScGa compound, and E_{Cr}^{bulk} , E_{Co}^{bulk} , E_{Sc}^{bulk} , E_{Ga}^{Bulk} are the bulk energies of Cr, Co, Sc and Ga atoms, respectively.

The formation energy of CrCoScGa in Table 3 is negative, indicating that this compound is stable and can be experimentally synthesized.

Table 1: Different types of structure for CrCoScGa compound.

Type	Cr	Co	Sc	Ga
Type I	(0,0,0)	(1/4,1/4,1/4)	(3/4, 1/4,11/4)	(1/2, 0,0)
Type II	(0,0,0)	(3/4, 1/4,11/4)	(1/2, 0,0)	(1/4,1/4,1/4)
Type III	(0,0,0)	(1/2, 0,0)	(1/4,1/4,1/4)	(3/4, 1/4,11/4)

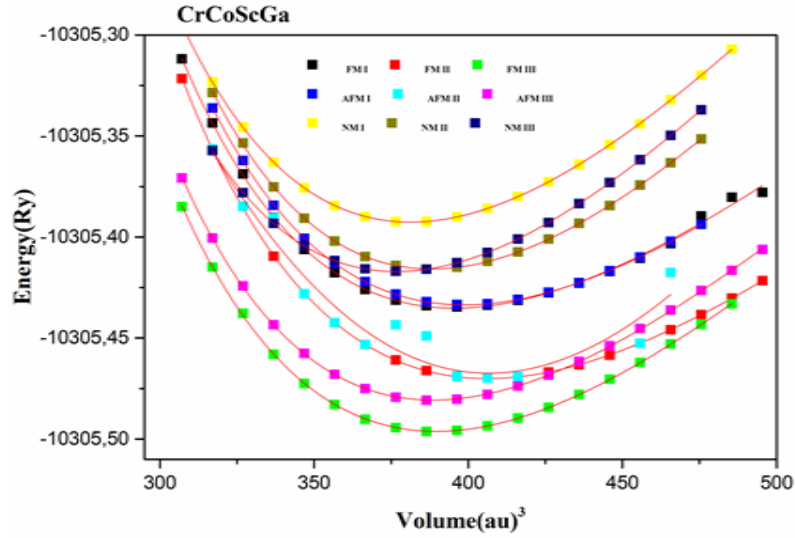


Figure 1. Optimization of total energy as a function of volume using the GGA approximation.

Table 2. The calculated values of the lattice parameter (\AA), bulk modulus (GPa), and its pressure derivative of CrCoScGa compound.

Compound	Phase	B		E_{\min}	
		$a_0(\text{\AA})$	(GPa)		B'
CrCoScGa	FM	6.134	119.728	4.716	-10305.49618
	AFM	6,129	120.466	4.833	-10305,48068
	NM	6.066	137.284	4.332	-10305.41710

Table 3. Total energy of ground state, formation energy (in Ry) and Debye temperature of CrCoScGa

Compound	E_{Total}	$E_{\text{Formation}}$
CrCoScGa	-	θ_D
	10305.49634	-0.068992
		470.288

3.2. Elastic properties:

We studied the elastic properties of our material to confirm its stability, which provide us later with information about its rigidity, anisotropy, thermal expansion, and Debye temperature. The Thomas Charpin method, included in the WIEN2k code was recently developed for calculating the elastic constants of compounds [10]. The number of elastic constants in a cubic crystal can be reduced to three independent constants: C_{11} , C_{12} , and C_{44} . It is clear from the table above that the elastic constants are positive, confirming the mechanical stability criteria of cubic structure:

$$C_{11} > 0, C_{44} > 0, C_{11} > C_{12}, (C_{11} + 2C_{12}) > 0 \text{ et } C_{12} < B < C_{11} \quad (3)$$

We can clearly see from Table 4 that the coefficient A is greater than the unit, indicating that our compound is anisotropic. We can also see that the B/G ratio is less than the critical value of 1.75, which distinguishes between ductile and brittle behaviors (brittle <1.75< ductile), allowing us to classify our compound as brittle.

Table 4. Elastic constants C_{11} , C_{12} , C_{44} , bulk modulus B, Young's modulus, Shear modulus G, Poisson's coefficient ν , and B / G ratio, Anisotropy parameter A.

Compnd	C_{11}	C_{12}	C_{44}	B	G	E	ν	B/G	A
CrCoScGa	189.1	87.4	104.3	121.4	78.2	193.1	4.2	1,6	2,1

3.3. Electronic propertie

Band structure:

In physics of solid-state, band theory is a model of the energy values that the electrons of a solid can take. The Fermi level E_F is defined at 0 eV. From figure 2, we can see that the majority spins of our compound is metallic whereas, for the minority spins, is semiconductor, with the existence of an indirect band gap, where the valence band maximum (VBM) and the conduction band minimum (CBM) are located at Γ and L high symmetry points, respectively. Thus, our material can be classified as half metallic ferromagnet.

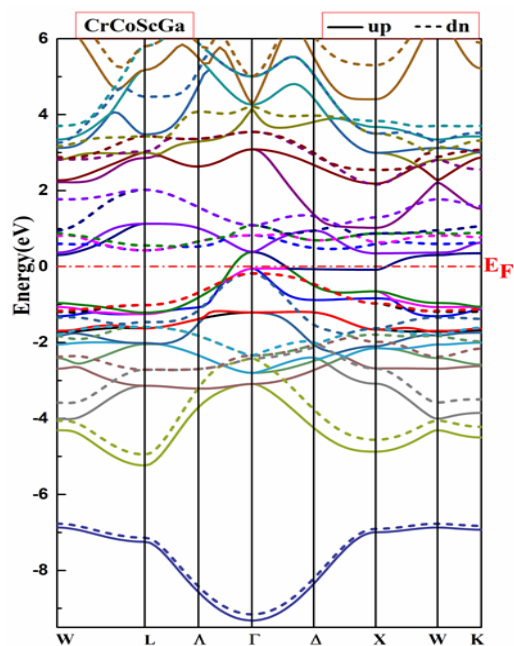


Figure 2: Spin-up and spin-down band structure for CrCoScAl compound by GGA approximation.

Density of states:

In order to analyze and explain the magnetic properties of our compound, we have mapped the total density of states of spins (spin-TDOS) and the partial density of states (spin-PDOS), shown in the figures 3. These figures

confirm that the d states of the transition elements (Cr, Co) contribute the most to the magnetic moment, while Sc and Ga atoms exhibit negligible contributions. We can notice also that the majority-spin (spin-up) has a metallic character, whereas the appearance of the gap at the Fermi level in the case of minority spins, confirming the half-metallicity behavior.

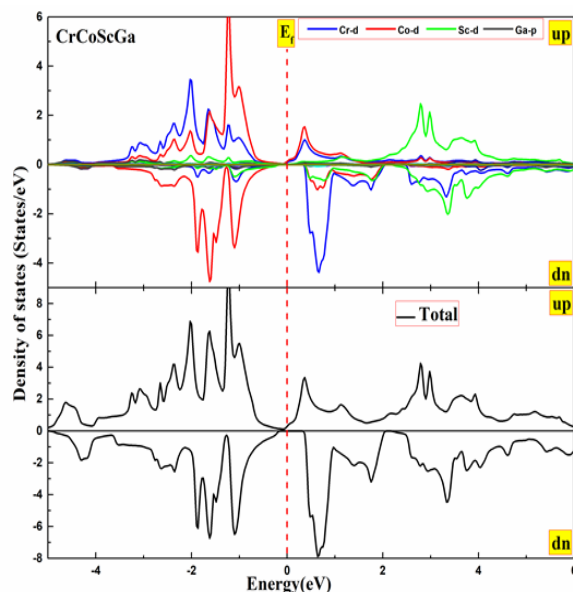


Figure 3. Calculated total and partial densities of states for CrCoScAl by GGA approximation.

3.5. Magnetic properties:

For the half-metallic ferromagnetic compound, the total magnetic moment must be an integer value, according to the Slater-Pauling rule. The integer total magnetic moment of $3 \mu_B$ (shown in Table 4), indicates that the ferromagnetic Heusler obeys the Slater-Pauling rule [11, 12]. By applying the formula decreed by Slater-Pauling, we have:

$$M_t = Z_t - 18 \quad (4)$$

$$Z_t = 20 \quad \text{so} \quad M_t = 20 - 18 = 2\mu_B \quad (5)$$

where M_t is the total magnetic moment per unit of formula and Z_t is the total number of valence electrons. We can also observe that the Cr atom contributes more to the total magnetic moment than the Co atom, while Sc and Ga present negligible contribution.

Table 5. Local and total magnetic moments of CrCoScGa.

Compounds	M_{tot}	M_{Cr}	M_{Co}	M_{Sc}	M_{Ga}	M_{int}
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CrCoScGa	3.000	2.817	-0.193	0.043	-0.039	0.372
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4. Conclusion

In summary, the first principles FP-LAPW method based on DFT in GGA was used to study the structural, electronic and magnetic properties of Heusler quaternary alloy CrCoScGa. for the three types of structure, the most energy stable one is type III in its FM phase. The negative energy of formation indicates the thermodynamic stability of our compound. We can classify it as an anisotropic and brittle material, based on its obtained elastic properties. The total magnetic moment M_t in the unit cell is an integer of $3\mu_B$, which follows the Slater-Pauling rule $M_t = (Z_{tot}-18)$. The electronic properties calculations revealed that CrCoScGa compound exhibits a half metallic behavior. This energy gap was created by d-hybridizations of the transition metal atoms Cr, Co.

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