



## Heusler alloys: A Computational Perspective

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### Abstract

Ferromagnetic Heusler alloys have the imminent applications in spintronic devices and smart materials. Ferromagnetic materials such as Fe, Co, Ni has a spin polarization about 35-40% but Heusler alloys have 100% spin polarization. The advantages of using these alloys are, they have half metallic property, 100% spin polarization at Fermi level, high Curie temperature etc and these alloys can be used for sensors, actuators, and memory storage devices. Heusler alloys are among the largest families of intermetallic compounds with face centered cubic structure. These alloys are divided into Full, half, inverse, quaternary Heusler alloys depending upon their crystal structure. In the past few decades, first principles *i.e.* density functional theory (DFT) calculations play an important role to study the structure, electronic and magnetic properties of various atomic, molecular and materials systems. Because DFT is an easy, capable and economical tool compared to experimental studies. By using the first principles calculations we can calculate even for materials that have not been experimentally done. In this review article, we would like to provide a systematic review of recent developments in the field of computational design of Heusler-based functional materials.

**Keywords:** density functional theory, Heusler alloys, half metal, Ferromagnetic property, Spin polarization, Spintronics

## 1. Introduction

Heusler alloys play a vital role in the development of smart materials for various potential applications (Figure 1). In particular, these alloys are widely studied for their applications in spintronics. The reason is, these alloys have half-metallic ferromagnetism, half metallic anti-ferromagnetism, low thermal conductivity, topological property, magnetic shape memory effect, high curie temperature( $T_c$ ), 100% spin polarization etc [1–5]. Heusler alloys are the ternary intermetallic compound, which was first discovered by German mining engineer “Friedrich Heusler” in 1903. Heusler found that it is possible to obtain ferromagnetic alloys from non-ferromagnetic elements [6, 7]. For example, Ferromagnetic Heusler alloys were made from copper, manganese, bronze alloyed with elements like tin, antimony, bismuth etc [8]. Ferromagnetic metals such as Fe, Co, Ni are the earliest materials used in spintronics applications. These materials were used to build magnetic tunnel junctions and spin valves, because they are inexpensive and readily available [9]. Most known Heusler alloys are ferromagnetic materials but most Ferromagnetic materials are not Heusler alloys.

The crystal structure of Heusler alloy is face centered cubic and it was reported in the year 1934. Similarly, an unusual striking property *i.e.* half metallic ferromagnetism was first reported by Groot *et al.* in the year 1983 [10]. The materials with half metallic ferromagnetic property are the most promising candidate for spintronics applications [11–13] NiMnSb was the first half metallic ferromagnetic material [14]. The materials with half metallic ferromagnetic and anti-ferromagnetic nature are more suitable for switching devices and memory storage devices [15]. In half metals, majority-spin (spin up) shows a metallic behavior whereas minority-spin (spin down) shows a non-metallic behavior (Figure 2). Further, Heusler alloys has the

ability to behave as semiconductor when all the electrons are spin down and behave as a metal when all the electrons are spin up [16–18].

A large number of Heusler alloys can be prepared by combination of almost all elements of periodic table (Figure 3). In the last few decades, Heusler alloys have been extensively studied using various experimental and theoretical techniques. In general, theoretical studies are used as alternate to high cost experimental techniques. Theoretical methods, in particular, density functional theory (DFT) methods play a significant role in studying various atomic, molecular and materials systems. It provides accurate results in comparison with classical approaches. Although enormous amount of review articles on Heusler compounds are reported in the literature, a systematic review of theoretical studies in Heusler compounds are scarce. For instance, He et al, have discussed various computational methodologies on Heusler alloys, their main focus is on high throughput screening or machine learning computational design [19]. Similar studies were also performed by Jin et al. and Jiang et al. [20, 21] Machine learning requires massive data, high error-susceptibility and requires more time and resources but DFT is more efficient and much faster and reduces the computation time significantly. The machine learning studies are still scarce. Hence, in this review article, we would like to provide a systematic overview of recent developments in the field of computational design of Heusler-based functional materials. In particular, our main focus is on quantum mechanical *i.e.* density functional theory studies in Heusler alloys. In chapter 2 we have discussed about various crystal structures of Heusler alloys. In chapter 3 quantum mechanical methods such as ab initio and density functional theory are discussed elaborately. In chapter 4 computational packages that are frequently used for Heusler alloys are discussed and finally in chapter 5 applications and future directions in the studies of Heusler alloys are discussed.

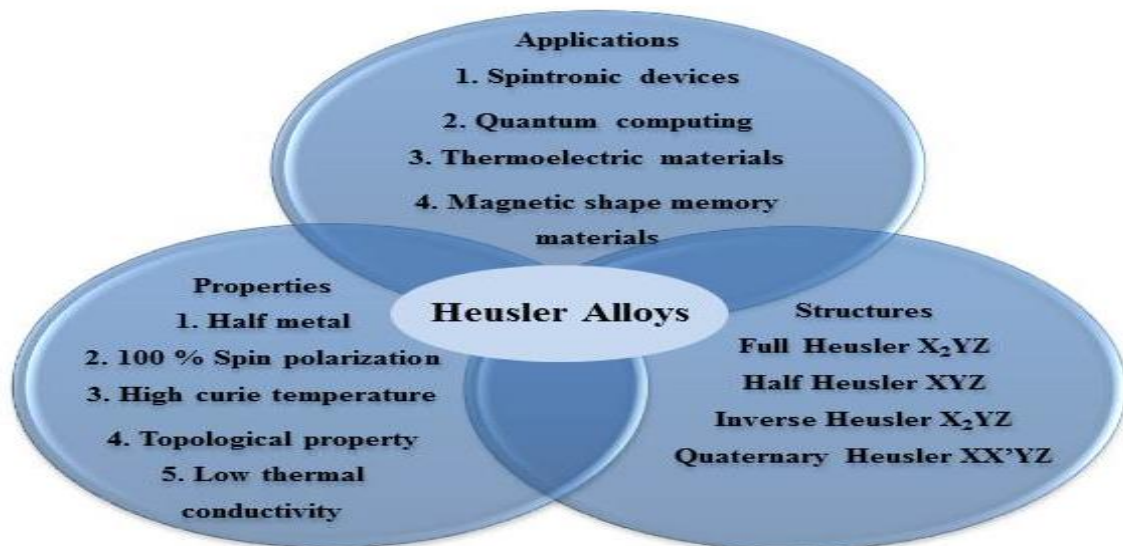


Figure 1 The overall view of Heusler alloys.

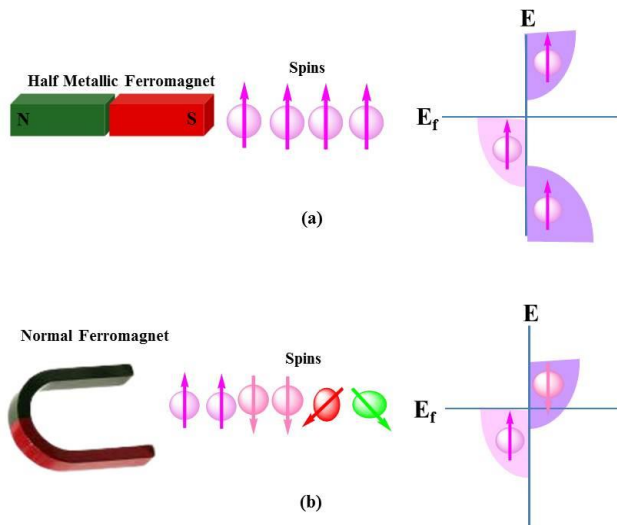


Figure 2 Half metallic Ferromagnet and spins are in same direction with large band gap. b) Normal Ferromagnet and the spins are in different direction with no band gap.

**Heusler Compounds  $X_2YZ$**

Figure 3 Periodic table illustrating the possible combinations of elements suitable in the design of various Heusler alloys.

## 2. Heusler Alloys

### 2.1 Full Heusler alloys

Full Heusler alloys belong to the first family with the general formula  $X_2YZ$ , these are ternary intermetallic compounds where Z is the p-block or main block element, X and Y are transition metal atoms [12]. The full Heusler alloys are in cubic with  $L2_1$  type structure with space group  $Fm\bar{3}m$  [22]. Four interpenetrating FCC sub lattices make up the  $L2_1$  type structure. The unit cell has 4 atoms so that the X atom can occupy at  $(1/4, 1/4, 1/4)$  and  $(3/4, 3/4, 3/4)$  with a center of inversion symmetry and Y, Z atoms are located at positions  $(1/2, 1/2, 1/2)$  and  $(0, 0, 0)$  respectively [23]. The full Heusler alloy sequence is X-Y-X-Z, where X has a larger valence than Y [24]. In 2017, Sanvito et al. have generated 2,36,115 compounds in the Heusler alloy library, by combining various elements in the periodic table. Among them 6778 compounds have a net magnetic moment and finally they concluded that 20 of them have a ferromagnetic ground state [25]. In the same year Balluff et al. predicted 21 anti-ferromagnetic Heusler compounds with a Neel temperature above ambient temperature using DFT [26]. The crystal structure of full

Heusler alloy is illustrated in Figure 4 (a). In this structure, each Y or Z atom has an octahedral symmetry as eight X atom as their first neighbors. The X has the tetrahedral environment with four Y or Z atoms as first neighbor. The symmetry of the crystal is reduced to octahedral one. The X atom contains two sub-lattices that are chemically distinct from one another but whose surroundings are similar because the second is rotated by  $90^\circ$ . [27].  $\text{Co}_2\text{MnSn}$  is the first half metallic Full Heusler [28, 29]. Some popular full Heusler alloys are listed in Table 1.

## **2.2 Semi or half Heusler alloys**

This is the second family with XYZ is the general formula of Heusler alloy. Half Heusler alloys are obtained by keeping a void at the position of one X atom out of two in  $\text{X}_2\text{YZ}$  structure [5, 6, 30]. These alloys crystallize in cubic with  $\text{C1}_b$  structure with F-43m space group [31, 32]. Here X and Y are transition metal atom Z is the main block or P block element with sp valence electrons [8]. Three FCC sub lattices that are interconnected to make up the  $\text{C1}_b$  type structure. The unit cell has three atoms, with the X and Y atoms occupying the positions (0, 0, 0) and (1/4, 1/4, 1/4), respectively, Z atom occupy at (3/4, 3/4, 3/4) and void at (1/2, 1/2, 1/2) [33]. Earlier Ma et al. have obtained 378 half-Heusler compounds with good structural stability and good electrical characteristics using first principles DFT calculations. Furthermore, by examining the electronic and magnetic properties, they found that there are 45 half metals and 34 near half metals. The implication is that the Slater Pauling rule holds true for all half metals. [27]. The main class of half Heusler alloys is semiconducting materials which can give totally optical response. Currently the half-Heusler alloys are much interesting subject for researchers because of their attractive thermoelectric properties. Figure 4 (b) represents half Heusler alloy crystal structure. The earliest known half Heusler alloy is NiMnSb [30]. Table 1 lists a few well-known half-Heusler alloys.

### 2.3 Inverse Heusler alloys

As one of the important subfamily of Heusler compounds and it is the new form of full Heusler alloys. When compared to X element, the Y element has more valence electrons [34]. Here X and Y are transition metal atom and Z is the main block or P block element with sp valence electrons [1]. In the full Heusler alloys the atoms are in X-Y-X-Z sequence but in inverse Heusler alloys atoms are in the sequence of X-X-Y-Z (the position Y and X are interchanged), which crystalizes in X type structure with prototype  $\text{CuHg}_2\text{Ti}$  and  $\text{AgLi}_2\text{Sb}$  [35][36]. This structure has 4 interpenetrating FCC sub lattices. The Y and Z atoms are present at Wyckoff positions 4b (1/2, 1/2, 1/2), 4d (3/4, 3/4, 3/4), 4c (1/4, 1/4, 1/4) and 4a (0, 0, 0) respectively. Inverse Heusler alloys are a potential Heusler alloy subfamily for low magnetic moment systems, and they are utilized in spintronics applications [37]. In order to determine the best materials for spintronics applications, Ma et al. previously conducted a massive first principles computational research of roughly 405 inverse Heusler compounds [38]. They first estimated the strength between cubic phase (full Heusler structure  $L2_1$ ) and inverse phase (inverse Heusler structure XA). Their total energies reveal that 128 XA structures compared to the  $L2_1$  phase, are favored energetically. Finally, they determined that 95 of these 128 inverse Heusler compounds can maintain the cubic structure in the presence of tetragonal distortions, so they decided to take tetragonal distortion into account. The crystal structure of these alloys is illustrated in Figure 4 (c) e.g.:  $\text{Mn}_2\text{CoGa}$  [39].

### 2.4 Quaternary Heusler alloys

Quaternary Heusler alloys are formed by replacing one X atom with 3<sup>rd</sup> TM atom ( $X'$ ) in  $X_2YZ$  structure i.e.  $XX'YZ$ . These alloys are crystallized in the Y-type structure with space group  $F\bar{4}3m$  and has no center of inversion with prototype  $\text{LiMgPdSn}$  [7, 40–42]. Here X and Y atom

are transition metal and Z is the main block or P block element with sp valence electrons [43]. This structure has four interpenetrating FCC sublattices and they occupy at Wyckoff positions. The Z atom is placed at 4a (0, 0, 0), Y atom is placed at 4b (1/2, 1/2, 1/2), X atom is placed at 4c (1/4, 1/4, 1/4), and X' atom is placed at 4d (3/4, 3/4, 3/4) respectively. Earlier He et al. have studied a family of quaternary Heusler compounds that operate steadily in the semiconducting state and are based on the 18 electron rule [44]. They have studied a total of 1320 quaternary Heusler compounds, which also includes rare earth elements with partially filled f orbitals. They have studied the stability of quaternary Heusler alloys in terms of thermodynamic stability and lattice dynamics. The newly identified Heusler semiconductors have the band gap in the range of 0.3-2.5 eV and they also show some different thermoelectric properties. The 18-electron rule is used for exploration of Heusler semiconducting materials. The crystal structure of these alloys is illustrated in Figure 4 (d). Some popular Quaternary Heusler alloys are listed in Table 1. The summary of different types of Heusler alloys, general formula, type, space group, ratio, sub lattices are given in Table 2 and are shown in Figure 5.



Table 1. Some popular Full, Half, Quaternary Heusler alloys.

Full Heusler <sup>a</sup>		Half Heusler <sup>b</sup>		Quaternary Heusler <sup>c</sup>	
Compounds	<i>a</i> (Å)	Compounds	<i>a</i> (Å)	Compounds	<i>a</i> (Å)
Mn <sub>2</sub> PtRh	6.160	AgAuO	5.792	FeCrScSi	5.992
Mn <sub>2</sub> PtCo	6.000	AgAuS	6.219	MnCrTiSi	5.855
Mn <sub>2</sub> PtPd	6.240	AgAuSe	6.407	MnCrVAl	5.897
Mn <sub>2</sub> TaTi	6.036	CuAgO	5.899	MnVTiAs	5.978
Mn <sub>2</sub> CoCr	5.730	CuAgS	5.961	CoVTiAl	5.978
Co <sub>2</sub> MnTi	5.840	CuAgSe	6.081	FeCrTiAl	5.964
Co <sub>2</sub> TiZn	5.802	CuAuS	5.966	CrFeHfGa	6.127
Rh <sub>2</sub> MnTi	6.150	KSrN	6.567	IrVYSn	6.720
Rh <sub>2</sub> MnZr	6.370	KSrP	7.445	CoVYSn	6.620
Rh <sub>2</sub> MnHf	6.321	KSrV	8.184	CoOsTiSb	6.255
Rh <sub>2</sub> MnSc	6.271	KVCd	6.867	RhFeTiSb	6.259
Rh <sub>2</sub> FeZn	6.020	KVZn	6.889	CoRuTiSb	6.228
Co <sub>2</sub> NbZn	5.630	KZnN	5.972	FeVHfSi	6.079
Co <sub>2</sub> TaZn	5.701	KZnP	6.668	MnCrZrSn	6.393
Pd <sub>2</sub> MnAu	5.750	TiAgB	5.523	FeCrYSi	6.236
Pd <sub>2</sub> MnCu	6.130	TiAlAg	6.126	CoFeTaSi	5.856
Pd <sub>2</sub> MnZn	5.840	TiAlAu	6.111	PtVYAl	6.608
Pt <sub>2</sub> MnZn	5.790	TiAlCu	5.827	CoOsCrAl	5.866
Ru <sub>2</sub> MnNb	6.201	ZnCdPb	6.721	NiFeMnAl	5.731
Ru <sub>2</sub> MnTa	6.200	ZnCdSi	6.229	MnCrNbAl	6.077
Ru <sub>2</sub> MnV	6.010	ZnCdSn	6.594	MnCrTaAl	6.053

<sup>a</sup> Ref [25], <sup>b</sup>Ref[112], <sup>c</sup>Ref[113].

Table 2. Comparison of different types of Heusler alloys.

Structures	General Formula	Type	Space Group	Ratio	Sublattices	Example
Full Heusler	X <sub>2</sub> YZ	L2 <sub>1</sub>	Fm3m	2:1:1	Four	Co <sub>2</sub> MnSn
Half Heusler	XYZ	C1 <sub>b</sub>	F-43m	1:1:1	Three	NiMnSb
Inverse Heusler	X <sub>2</sub> YZ	X	Fm3m	2:1:1	Four	Cr <sub>2</sub> CoGa
Quaternary Heusler	XX'YZ	Y	F-43m	1:1:1:1	Four	YFeCrSb

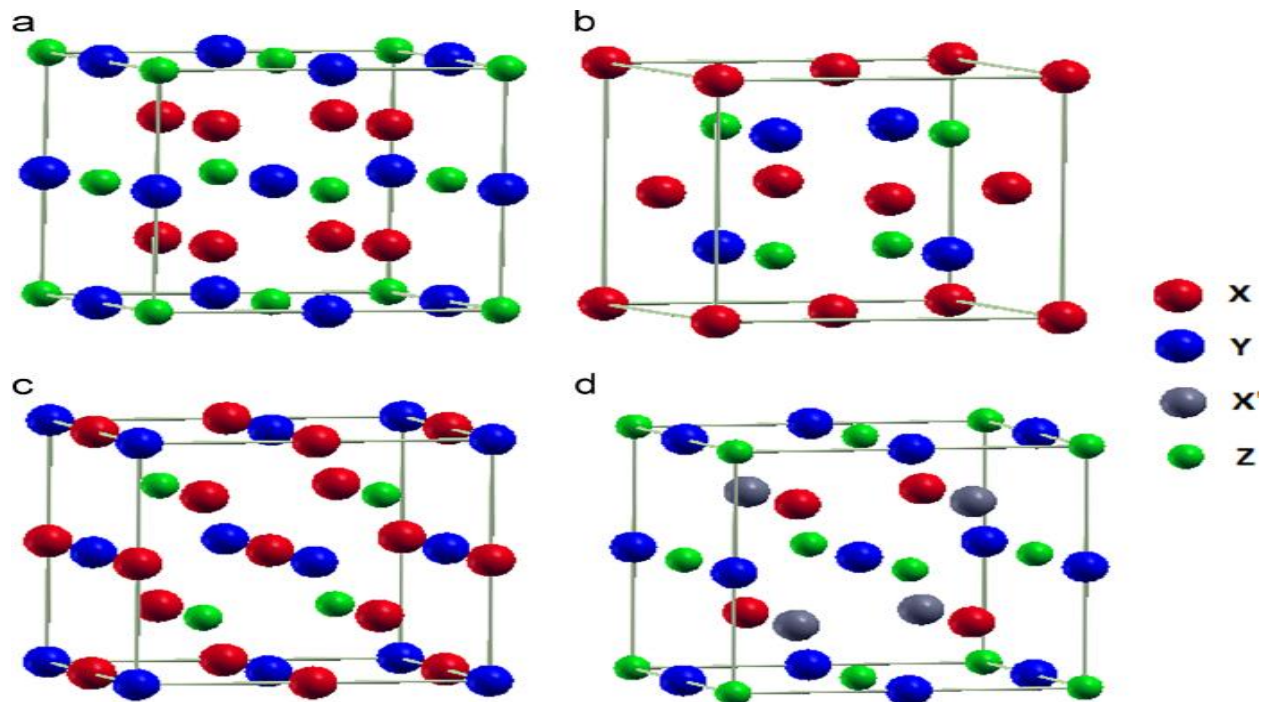


Figure 4 a) Full-Heusler alloys b) Half-Heusler alloys c) Inverse-Heusler alloys d) Quaternary-Heusler alloys.

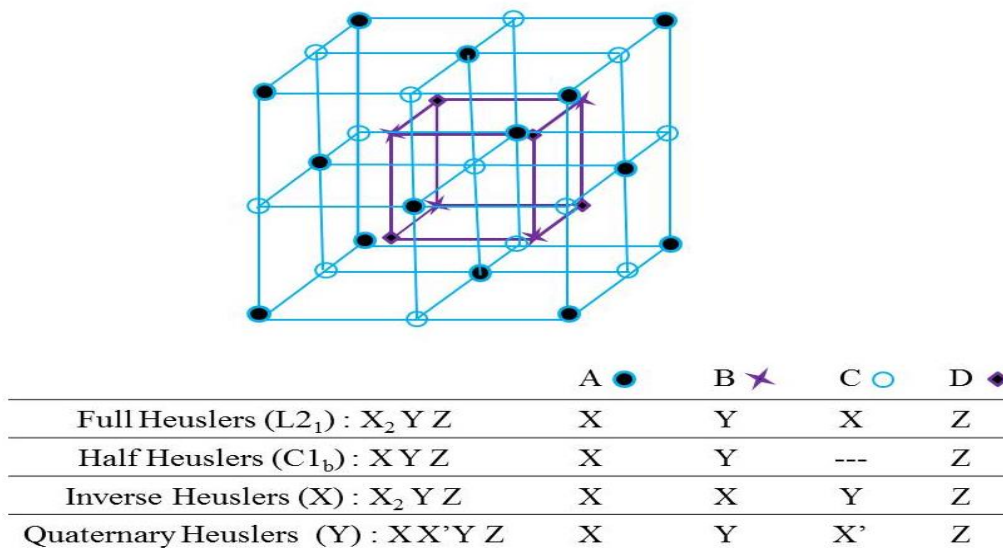


Figure 5 Comparison of four different types of Heusler alloys.

### 3. First-principles calculations

#### 3.1 ab initio method

The Schrödinger equation is unable to solve many body systems exactly because of the inclusion of the electron-electron interaction term. Hence various approximation methods are used to solve these systems [45]. In 1927, Born and Oppenheimer proposed an approximation and it is mostly used in quantum chemistry to compute the properties of larger molecules *i.e.* Born-Oppenheimer approximation [46]. Born-Oppenheimer approximation is defined as the separation between nuclear and electronic motion. The nuclei must move much more slowly than the electrons. The electrostatic repulsion between positively charged nuclei is treated as a constant in the electronic problem thanks to this approximation, which also allows us to eliminate the kinetic energy term of the nuclei. However, the electron-electron interaction term in the Schrödinger equation prevents it from being solved. In 1927, Hartree introduced a method to calculate the approximate wave function and energies for ions and atoms which is called self-consistent field method (SCF) [47]. Hartree made an assumption that the many electron wave functions can be written as a product of a set of single electron wave functions. Then the total wave function is the product of the wave function of the individual electrons. In 1930, Slater and Fock identified that the antisymmetric wave function is not included in Hartree approximation and the exchange interaction between the electrons is also not taken in to account [48]. This drawback is overcome in Hartree-Fock approximation. It is based on a simple approximation to the many-body wave function, which is given by a single Slater determinant [49]. Using this technique, one may determine the  $n$  electron wave function, which is a single Slater determinant that represents the sum of the  $n$  electron wave functions [50]. The most popular ab initio methods are Hartree-Fock

SCF, Moller-Plesset perturbation methods, coupled cluster methods and multireference methods.

The Hartree-Fock equation is,

$$[-\sum_i \frac{\nabla_i^2}{2} - \sum_I \frac{ZI}{|r_i - R_I|} + \frac{1}{2} \sum_{i \neq j} \int \varphi_j^*(r_j) \frac{1}{|r_i - r_j|} \varphi_j(r_j)] \varphi_i(r_i) - \sum_j [\int \varphi_j^*(r_j) \frac{1}{|r_i - r_j|} \varphi_j(r_j)] \varphi_j(r_i) = \varepsilon_i \varphi_i^*(r_i) \quad (1)$$

### 3.2 Density functional theory

The ground state properties and electronic structure of materials can be calculated accurately by first-principles calculation i.e. density functional theory [51–54]. For a given material, the calculated total energy can be used to obtain elastic moduli, lattice parameters and other properties of competing crystal structures [55, 56]. In addition to that phonon dispersion curve, density of states, charge density and electronic band structure can also be analyzed [57–59]. The calculations are usually performed at zero temperature but the results obtain constitute the basis for understanding finite temperature properties. Hohenberg and Kohn introduced the modern DFT in 1964, and it is one of the most frequently used tools for various materials in solid state physics, chemistry, and materials science [60–62]. In DFT the electron density is used as a basic variable in solving Schrödinger equation rather than using the many electron wave functions. DFT mainly depends upon the two fundamental mathematical theorems that was developed and proved by Hohenberg and Kohn [63, 64].

#### 3.2.1 Hohenberg-Kohn theorems

##### 3.2.1.1 THEOREM 1: Proof of Existence

The total energy of a system, which includes the many body effects of electrons such as exchange and correlation in the presence of a static external potential, is a distinct functional of charge density [65]. There is a one-to-one correspondence between the potential and the ground state particle density. The ground state of a many-electron system (under the influence of an external potential) is particularly defined by the ground-state electron density [66].

$$E = E[n(r)] \quad (2)$$

### 3.2.1.2 THEOREM 2: Variational Principle

In simple words, this theorem states that the functional that gives the ground state energy of the system. It gives the minimum energy only if the input density corresponds to the initial ground state density. This theorem helps us to reduce the very difficult problem of finding all the ground state properties of a system and to find the lowest energy of the system with respect to electron density[66].

$$E_0 \leq E[\tilde{\rho}] = T[\tilde{\rho}] + E_{Ne}[\tilde{\rho}] + E_{ee}[\tilde{\rho}] \quad (3)$$

### 3.2.2 Exchange-Correlation Functional

The exchange-correlation functional is unknown to solve the Kohn-sham equations [67]. When attempting to solve the Kohn-Sham equations, this is the main disadvantage. Hence to overcome the problem two approximation methods has been developed to find the unknown exchange-correlation functional. They are

- Local Density Approximation
- Generalized Gradient Approximation

#### 3.2.2.1 Local Density Approximation (LDA)

This is the standard and easiest scheme to approximate the exchange correlation energy and LDA is considered as the mother of all approximations [68]. In DFT calculations, this is one of the first methods to approximate the exchange correlation functional. In that there are two assumptions: 1) the local density exchange correlation energy per particle depends on the local density and 2) the exchange correlation energy is equal to the exchange correlation energy per particle of a homogeneous electron gas, which has the same electron density in a neutralizing

positive background. The LDA approximation gives accurate result for system with slowly varying density [69, 70]. Vibrational energy, bond lengths and other properties are predicted correctly. In the LDA,  $E_{xc}[\rho]$  can be written as

$$E_{xc}[\rho] = \int \rho(r) \varepsilon_{xc}(\rho) dr \quad (4)$$

### 3.2.2.2 Generalized Gradient Approximation (GGA)

The GGA functional is more accurate and reliable particularly for finding hydrogen bonding compare to LDA. The meta-GGA functional gives more improvement over GGA especially for transition metal systems [71, 72]. For a homogeneous gas, the LDA approximation is the same, but the initial electron densities are not uniform throughout the system. The development of the LDA approximation could be to contain the information of the rate of change in the functional. This process can be done by include some gradient terms hence it is called generalized gradient approximation. There are plenty of GGA, hybrid GGA, meta GGA and hybrid meta GGA functionals are available for studying various atomic and molecular properties of many electron systems. In the GGA  $E_{xc}[\rho]$  can be written as

$$E_{xc}[\rho^\alpha, \rho^\beta] = \int f(\rho^\alpha(r), \rho^\beta(r), \nabla \rho^\alpha(r), \nabla \rho^\beta(r)) dr \quad (5)$$

## 3.3 Electronic structure calculations

Electronic structure calculations have become an indispensable tool in many areas of materials science and quantum chemistry [73]. The most widely used methods to find the electronic band structure of metals are, (i) Plane wave Pseudo Potential method (ii) Ultra Soft Pseudo Potential and (iii) Projector Augmented Wave method.

### **3.3.1 Plane Wave Pseudo Potential Method (PWPP)**

In this method no assumption are made about the shape of the wave function and electron density [74]. It is very easy to calculate atomic forces within PWPP formalism because the basis functions are not inserted to the atom. In general, computations are expensive for a system with a many number of electrons. However, pseudo potentials can be used to make the calculations easy [75]. The physical property of solid mainly depends on the valence electrons, thus the computing effort can be decreased. The core electrons are removed in the pseudopotential approximation, and the strong ionic potential is replaced by a weaker pseudo potential that acts on a set of pseudo wave functions rather than the true valence wave functions [76]. The pseudo wave function is built in the core region so that all nodes are removed while the norm is preserved. Since the core state nodes have been eliminated, much fewer plane-wave basis functions are needed to describe this wave function in the core region, which requires less computational work [77]. For this reason it is widely used method for electronic structure calculations.

### **3.3.2 Ultra-Soft Pseudo Potentials (USPP)**

Ultra-soft pseudopotentials were first introduced by Vanderbilt in 1990 [78]. Vanderbilt proposed a new concept on relaxing the norm-conserving constraint to overcome the limitation of the norm-conserving pseudopotentials. All the electron and pseudo wave functions are equal with the norm-conserving pseudopotentials outside the core radius but inside they are allowed to be as soft as possible [79]. Thus the pseudo wave functions are not normalized inside the core radius resulting in charge deficit and to make up this charge deficit, localized atom centered augmentation charges are introduced [80]. Also all information about the real wave function close to the nuclei is lost; making it hard to compute the properties which rely on the core region

and too many parameters need to construct this ultra-soft pseudo potentials limited in its success [81].

### **3.3.3 Projector Augmented-Wave Method (PAW)**

The projector augmented wave method was first introduced by Blochl. It represents an aim to achieve continuously the computing efficiency of the pseudopotential method and the accuracy of FPLAPW (Full-potential linearized augmented plane wave) which is important for DFT calculations on solids [82, 83] The PAW method is a linear transformation between valence and core wave functions. In PAW method all the electron wave functions are constructed from pseudo wave (PS) function [84, 85]. The PS functions in the PAW method are normally smoother compared to norm-conserving pseudopotential methods so that the wave functions can be described with small degrees of freedom. The PAW method comes from augmented wave method and the pseudopotential approach, which merge their traditions into a unified electronic structure method [86]. Softwares implementing the PAW method are VASP (Vienna Ab-Initio Simulation Packages), Quantum espresso, ABINIT, CASTEP (to calculate NMR properties).

## **4. Computational packages**

There are various computational packages available for the investigation of Heusler alloys using DFT. Among them, Quantum espresso, VASP, WEIN2k are widely used. Here, Quantum espresso is open source software. VASP and WEIN2k are commercial packages.

### **4.1 Quantum espresso**

Quantum espresso is abbreviated as opEn-Source-Package-for-Research-in-Electronic-Structure-Simulation and Optimization and distributed under the terms of General Public License [87]. It can be used for super cell or any crystal structure, insulators and metals [88]. The atomic cores



are explained as norm conserving pseudopotential, PAW, USPP sets. Different exchange correlation functionals are used like LDA or GGA and advanced functional like Hubbard U corrections and hybrid functional also used. For instance, Idrissi et al have studied the LuCoVGe equiatomic quaternary Heusler alloys using DFT using quantum espresso. They investigated the electronic and magnetic properties of this Heusler alloys and they found that, this alloys have half metallic ferromagnetic nature [89].

#### **4.2 Vienna Ab-Initio Simulation Package**

The VASP software was developed at the beginning of the 1990s and it is well-known, 3D-periodic, first principles simulation program. PAW framework for pseudopotential, ultrasoft pseudopotentials are implemented in this package. VASP uses a plane wave basis set and periodic boundary conditions [90]. Ions and electrons interaction are explained by PAW, ultrasoft pseudopotential method with different approximations (LDA, GGA, GGA-PBE). VASP can be used to find ground state properties (lattice parameters, cell volume, formation enthalpy) electronic structure such as total and partial state density, their mechanical (elastic constants, bulk modulus, young's modulus and shear modulus) and magnetic properties [91]. For example, Zhang et al have studied  $\text{Ag}_2\text{ScGe}$  Heusler alloys within the first principles calculation performed in the VASP code. They have calculated the mechanical and electrical characteristics, they discovered that, this alloys exhibits ductile and metallic material with elastic anisotropy [92].

#### **4.3 WEIN2k**

WEIN2k is centered on the local orbital technique and linearized augmented plane wave full potential [86, 87]. It consists of several independent F90 programs which are connected together via c-shell scripts. It is based on Full Potential Linearized Augmented Plane Wave method [88].

WEIN2k package is used to calculate the solid state properties. LDA, GGA, Meta-GGA, LDA+U are implemented in this package [121]. Energy bands, electron density phonons, Fermi surface, optical properties can be calculated. For instance, Chibani et al have studied the RuVX Heusler alloys within the first principles calculation i.e. DFT. They have used the WEIN2k code to find the structural, electrical, and transport characteristics. These alloys show indirect band gap, good for transport devices [104]. Benkabou et al have studied the magnetic and electrical characteristics of CoRhMnZ Heusler alloys. They have used full-potential linearized augmented plane wave approach as implemented in the WIEN2k code to calculate their properties. They identified that, this Heusler alloys shows half metallicity and high spin polarization [105].

## **5. Applications and future direction**

The first Heusler alloy was discovered by Friedrich Heusler in 1903 [6]. Since then enormous number of Heusler compounds were reported in the literature to explore their potential applications. Figure 6 shows the bar diagram of experimental and DFT studies in various types of Heusler alloys. From this figure, it can be seen that more attention of experimental and theoretical studies are given in quaternary, inverse, half Heusler alloys. Less attention is given to full Heusler alloys. The reason is full Heusler alloys have potential applications in ferromagnetic shape memory alloys. But other Heusler alloys have received considerable experimental and theoretical attention because of their applications in spintronics, solar cells and thermoelectric generators. It is mostly due to half metallic behavior and making them promising material for spintronic devices.

In general, Heusler compounds have been widely studied for their potential applications in Spintronics, Magnetocaloric effect, Magnetic shape memory alloys [93]. Spintronics is also called spin electronics, it involves the study of active control and manipulation of spin degrees of

freedom in solid-state systems [94]. The basic study of spintronics includes spin dynamics, spin relaxation and spin transport in electronic materials and the spin degrees of freedom of electrons in electronics [95, 96]. In spintronics-based magnetic semiconductor, the ferromagnetism is the most important behavior compared to other magnetic behavior such as iron form a permanent magnet [97]. Heusler alloys are suitable for achieving highly effective spin injection from a ferromagnet into a non-magnetic metal due to their high  $T_c$  and good lattice matching with most major semiconductors. Heusler alloys include not only metallic and semiconducting materials, but also superconducting compounds. Ishikawa et al reported the first superconducting Heusler materials Pd<sub>2</sub>RESn and Pd<sub>2</sub>REPb [98]. The advantage of Spintronic device is to make use of spin rather than the charge of electron, because spintronics has less power, faster, high storage density, low cost, no loss of data if power suddenly off, store data in single chip. The Spintronic device includes magnetic tunnel junction (MTJ), Giant magneto resistance (GMR) and magnetic spin valve (MSV) [99][100]. Nowadays MTJ and GMR can be used for storage of digital information such as magnetic random access memory, hard disk drives etc [101]. Due to high (100%) spin polarization of Heusler compounds these are perfect for spin contacts to semiconductors [102]. The magnetic properties of Heusler alloys mainly depend on lattice matching with substrates and it is major factor for spin injection, spin transport, operation in Spintronic devices [103]. Due to their characteristics, including metamagnetic and shape recovery, magnetic shape memory alloys with Heusler type structures have drawn attention from researchers recently.

Heusler alloys can be used in the field of thermoelectrics in addition to spintronics because they are simple to make and process and contain non-toxic elements [104]. Thermoelectric materials have properties such as semimetal, tunable band gap, tunable charge

carrier concentration, and low thermal conductivity. Advantages of these materials are low cost, industrial process, and available of resources [105]. A new class of so-called topological states has recently emerged. Topological Insulators (TIs), the corresponding materials, have recently undergone experimental study after being predicted theoretically [106]. On the other hand, zero band gaps are present in the majority of reported Heusler topological insulators. The properties of topological insulators are semiconductor with tunable band gap, band gap in bulk, band gap in well structures, and large spin orbit coupling [107]. Band-structure calculations have predicted that many half-Heusler compounds are topological insulators. In addition to that Heusler compounds are used in the field of solar cells, thin films etc. Shape memory alloys are metals that can retain their original shape after being bent or twisted from their original shape when heated to a specific temperature. The properties are high elastic deformation, low cost, low density, low biocompatibility, biodegradability and high ductility. It can be used in the field of aerospace, robotics, sensors [108, 109]. Recent studies on Heusler alloys are devoted to catalysis. Heusler alloys are studied for their potential applications as catalytic materials in various industrial applications [110, 111]

This article provides a generalized overview of computational design of Heusler compounds, their emergent properties, and possible applications based on DFT calculations. Numerous novel Heusler compounds, such as full Heusler, half Heusler, inverse Heusler, quaternary Heusler and numerous new materials have been discovered with applications in optoelectronics, spintronics, thermoelectrics, and piezoelectricity. Highly tunable Heusler alloys have good applications beyond spintronics and thermos-electrics. But still more enough space is available for enhanced properties to explore their applications for ex. Damping constant in STT-MRAM. Currently it is not possible to calculate these properties using first principles DFT

calculations. The upcoming research is based on the computational development techniques like machine learning algorithms which are used to find various atomic and molecular properties of the material. Heusler compounds have been found to have additional intriguing features and uses, such as ideal super elasticity and catalysis. In future, maximum entropy alloys of Heusler will be one of the research directions.

Although, current trend in materials design is shifting towards the machine learning algorithms, we hope that our systematic review will be helpful to the future experimental and theoretical studies on Heusler compounds.

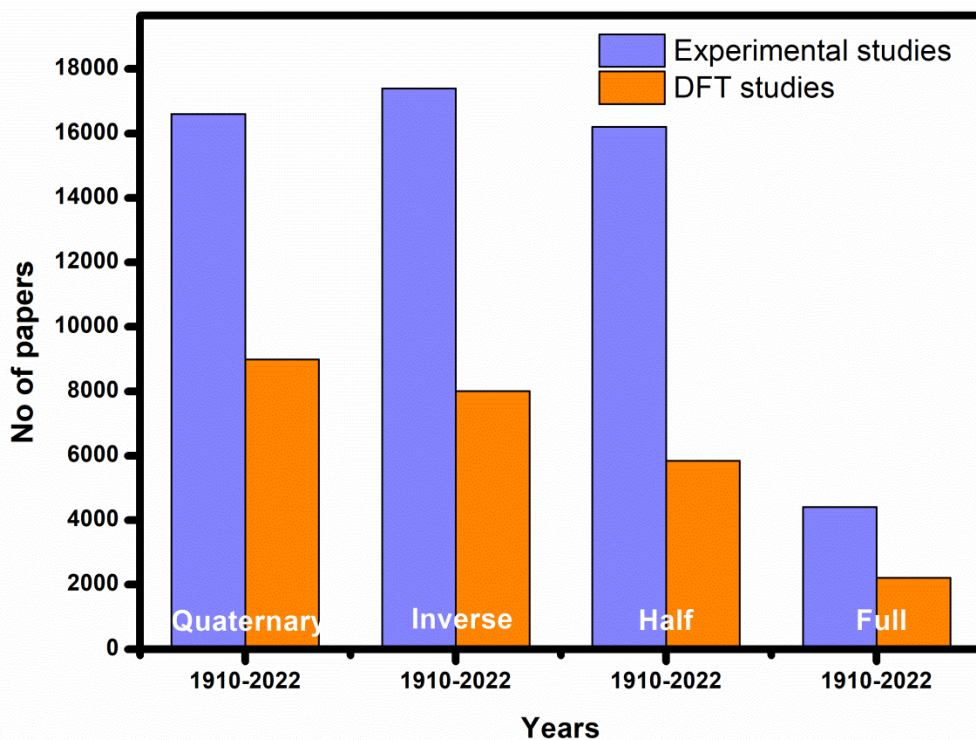


Figure 6 Bar diagram of the DFT and Experimental studies of Heusler alloys were reported during 1910-2022.

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