

Features of The Electronic and Magnetic Properties of Heusler Alloys in The States of A Half-Metallic Ferromagnet and A Spin-Gapless Semiconductor

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The review focuses on Heusler alloys exhibiting a range of remarkable functional properties, such as shape-memory behavior and magnetocaloric effects. Special attention is devoted to Heusler compounds that exhibit half-metallic ferromagnetism and spin-gapless semiconducting characteristics. While these materials possess relatively conventional crystal structures, their unusual electronic configurations and magnetic ordering give rise to distinctive kinetic and magnetic properties. Their magnetic and transport behavior is highly responsive to external stimuli, with variations in chemical composition or external conditions capable of driving transitions between different electronic and magnetic states. Such tunability offers significant prospects for tailoring the electronic and magnetic properties of Heusler alloys, thereby enhancing their potential for technological applications, particularly in spintronic devices and micro- and nanoelectronics.

Keywords: Heusler alloys, half-metallic ferromagnets, spin-gapless semiconductors.

INTRODUCTION

In 1903, the German chemist Friedrich Heusler reported a new class of intermetallic compounds, which subsequently became known as Heusler alloys [1]. Since their discovery, more than 1500 Heusler compounds have been identified, many of which exhibit remarkable and technologically relevant physical properties [2–5]. Among the various members of this family, particular attention has been devoted to materials showing the shape-memory effect (SME), magnetocaloric effect (MCE), half-metallic ferromagnetism (HMF), and spin-gapless semiconductor (SGS) behavior [6, 7].

Research on the magnetocaloric effect gained considerable momentum during the late 1990s and early 2000s following the demonstration of a giant MCE in gadolinium and the $\text{Gd}_5(\text{Si}_2\text{Ge}_2)$ compound [8]. Significant magnetic anisotropy at temperatures close to ambient conditions was subsequently reported in $\text{La}(\text{FeSi})_{13}$ - and MnAs -based materials [9]. Heusler alloys also attracted substantial interest because of their magnetocaloric properties. A representative example is Ni_2MnGa , which exhibits both the shape-memory effect, influenced by temperature and magnetic fields, and the magnetocaloric effect [10]. At present, numerous

research groups worldwide are actively investigating shape-memory and caloric phenomena in Heusler-based materials.

Another important research direction concerns the occurrence of half-metallic ferromagnetic and spin-gapless semiconducting states in Heusler compounds [2, 3, 5]. A half-metallic ferromagnet is characterized by a spin-dependent electronic structure in which an energy gap exists around the Fermi level for one spin channel, whereas the opposite spin channel possesses a finite and generally high density of electronic states. The HMF state was theoretically predicted in NiMnSb [32] and PtMnSb [33, 34] by de Groot and co-workers, while Kübler and colleagues predicted similar behavior in Co₂MnSn [35]. Since these pioneering predictions, HMF materials have been extensively investigated using both theoretical and experimental approaches [5, 6, 36].

The spin-gapless semiconductor state represents a closely related but distinct electronic configuration. It was first proposed theoretically by Wang [37]. In an SGS system, one spin channel exhibits a zero or vanishing band gap rather than the finite density of states characteristic of the conducting spin channel in a conventional half-metal. This behavior is conceptually related to the gapless semiconductor systems extensively investigated by Tsidilkovski and collaborators [38, 39]. The combination of strong spin polarization and unusual electronic structure makes HMF and SGS materials particularly attractive for spintronic applications. Their high spin polarization can, in principle, be maintained even near room temperature, providing important advantages for spin-dependent electronic devices [5, 6].

SGS materials possess an additional distinctive feature: electrons can be excited from the valence band to the conduction band with extremely small energy requirements. For excitation energies below the band gap of the opposite spin channel, the resulting charge carriers can exhibit complete, or nearly 100%, spin polarization. Notably, the same high degree of spin polarization can also be achieved for holes. Furthermore, the electronic, magnetic, and transport characteristics of these systems are not fixed but can be modified through external stimuli, including hydrostatic pressure, electric and magnetic fields, electromagnetic radiation, and the introduction of impurities or chemical substitutions [37]. Such controllability makes Heusler-based HMF and SGS materials promising candidates for tunable spintronic technologies.

A substantial body of research has been devoted to HMF and SGS Heusler alloys, with most studies concentrating on conventional full-Heusler X₂YZ and half-Heusler XYZ systems. In these compounds, X and Y are generally transition-metal elements, whereas Z is commonly selected from the s- and p-block elements of the periodic table. More recently, research interest has expanded toward less conventional Heusler systems, including quaternary compounds [40–42], all-d-electron systems [43–45], and unusual Z₂XY-type Heusler alloys [46–48]. These developments demonstrate the considerable chemical and structural flexibility of the Heusler family.

Consequently, a systematic assessment of the fundamental concepts underlying HMF and SGS states in Heusler alloys is highly desirable, particularly in view of the growing number of recent studies. Such a review should also address the characteristic electronic transport and magnetic properties associated with these states, examine the emerging classes of unconventional Heusler compounds, and identify promising directions for future research. A deeper understanding of these aspects may facilitate the development of Heusler-based

materials with controllable electronic and magnetic properties for advanced spintronic and electronic applications.

HALF-METALLIC FERROMAGNETS

Magnetic materials can generally be described in terms of two spin-dependent electronic states, conventionally designated as spin-up and spin-down. Accordingly, charge transport may be considered to occur through two independent conduction channels associated with carriers of opposite spin orientations. In half-metallic ferromagnets (HMFs), within a broad temperature interval well below the characteristic “gap temperature,” carriers belonging to the spin-down channel become strongly suppressed. Consequently, electrical conduction is dominated by the spin-up channel, resulting in a spin polarization of the charge carriers that can approach 100%. Under these conditions, conventional electron–magnon scattering is suppressed, and electron scattering involving magnons can occur predominantly through two-magnon processes [49].

The contribution of two-magnon scattering is reflected in the low-temperature electrical resistivity through a characteristic power-law dependence, $\rho \propto T^n$, with the exponent generally satisfying $7/2 < n < 9/2$. Experimental investigations of polycrystalline $\text{Co}_2\text{FeSi}_{1-x}\text{Z}_x$ Heusler alloys, where Z represents Al, Ge, Ga, Sn, or In and $x = 0$ or 0.5, revealed a resistivity contribution proportional to $T^{9/2}$ [50]. In single-crystalline Co_2FeSi , a somewhat different dependence, $\rho \propto T^4$, was reported [51]. More recent measurements on Co_2FeAl , Co_2FeSi , and Co_2FeGe have shown power-law behavior with exponents between approximately 3.5 and 4 in the temperature range of 40–65 K. These experimental observations have been attributed to two-magnon scattering mechanisms [50–52].

The origin of half-metallicity in X_2MnZ and XMnZ -type Heusler compounds can be understood from their characteristic electronic structure [32, 34, 35, 53]. If the hybridization between the atomic states of X and Z is initially neglected, the Mn d orbitals form bonding and antibonding states separated by a pronounced energy gap. In the ferromagnetic configuration, strong intra-atomic exchange interaction associated with Hund’s rule produces a substantial energy separation between the two spin subbands of the Mn atoms. One of these spin-dependent subbands lies close in energy to the p states of the Z element. The resulting p–d hybridization can significantly reduce or even eliminate the corresponding energy gap. In contrast, the other spin subband maintains a well-defined gap. When the Fermi energy lies within this gap, the compound can acquire a half-metallic ferromagnetic ground state.

Heusler HMF systems can exhibit either relatively strong or weak magnetism, depending on the nature of their magnetic ordering. These magnetic states may arise from localized moments, itinerant or collectivized magnetism, or a coexistence of localized and itinerant characteristics. The electronic structures of Heusler XYZ compounds containing X = Fe, Co, Ni, Rh, Ir, Pd, or Pt; Y = Ti, V, Zr, Hf, Nb, or Ta; and Z = In, Sn, or Sb were investigated in [54]. The calculations showed that compounds possessing an electronic gap can stabilize different types of magnetic states. Co- and Ni-containing systems tend to exhibit predominantly itinerant or collectivized magnetism, whereas Fe-, Ti-, and V-based compounds may display localized, collectivized, or mixed magnetic behavior.

The existence of an energy gap in HMF Heusler compounds is also closely associated with the Slater–Pauling rule, which establishes a relationship between the total number of valence

electrons, z , and the total magnetic moment, M [55]. For full-Heusler compounds, this relationship can be expressed as

$$M = z - 24.$$

Thus, a compound containing 24 valence electrons is expected, according to the conventional Slater–Pauling picture, to possess a vanishing total magnetic moment. However, recent studies have identified unusual Heusler compounds that do not strictly follow this conventional relationship. For example, Al_2MnCu , belonging to a recently reported class of Heusler materials containing a substantial fraction of p-block elements, possesses 24 valence electrons but exhibits a comparatively large magnetic moment of approximately $1.8 \mu\text{B}$ per formula unit and a Curie temperature of about 315 K [46]. To account for this apparent violation of the Slater–Pauling rule, the authors proposed a phenomenological explanation based on a molecular-orbital description [46].

Another unconventional category is represented by all-d Heusler alloys, in which the constituent elements are exclusively d-block elements [44]. The electronic and magnetic characteristics of equiatomic quaternary Heusler alloys (EQHAs) of the ZnXX'YZn -type, with $X = \text{Fe, Ni, or Co}$, $X' = \text{V or Cr}$, and $Y = \text{Nb, Ti, Zr, or Hf}$, have been investigated using first-principles density functional theory calculations [44]. The results indicate that complete spin polarization can be achieved in these systems. In particular, NiVTiZn and NiVZrZn were identified as half-metallic ferromagnets, whereas FeVNbZn , CoCrTiZn , CoCrZrZn , and CoCrHfZn were found to exhibit half-metallic ferrimagnetic behavior. The overall magnetic moments of these compounds are largely governed by the contributions from the V and Cr atoms.

Heusler alloys can also combine several desirable functional properties within a single material, making them particularly attractive for multifunctional applications. An illustrative example is provided by the $\text{Co}_2\text{VGa}_{1-x}\text{Sb}_x$ system, in which Ga is progressively substituted by Sb [56]. Detailed structural, electronic, and functional investigations demonstrated that the compositions with $x = 0$ and 0.25 preserve their half-metallic character and exhibit complete spin polarization. In particular, $\text{Co}_2\text{VGa}_{0.75}\text{Sb}_{0.25}$ emerges as a promising candidate for spintronic applications. At a higher Sb concentration, $x = 0.5$, the alloy undergoes a martensitic transformation from the cubic $L2_1$ structure to the tetragonal $D0_{22}$ structure at approximately 347 K. The occurrence of this structural transformation enhances its potential for shape-memory applications. Interestingly, $\text{Co}_2\text{VGa}_{0.5}\text{Sb}_{0.5}$ maintains its half-metallic character even though its lattice parameter increases by approximately 2%, demonstrating the considerable robustness of its electronic and magnetic properties against structural modification.

SPIN-GAPLESS SEMICONDUCTORS

Spin-gapless semiconductors have the following features: (1) a small amount of energy is required to excite charge carriers; (2) fully spin-polarized charge carriers (electrons and/or holes) can be generated by light excitation or thermal activation; (3) charge carriers are fully spin-polarized; (4) under the influence of a magnetic field, polarized carriers can move to the edge of thin samples due to the Hall effect [57]. Not only electrons, but also holes can be 100% spin-polarized in spin-gapless semiconductors. These features lead to unique transport properties, i.e., the coexistence of high values of electrical resistivity, Curie temperature, magnetic moment, and a small anomalous Hall effect [58].

Full Heusler alloys can form various structures. Ordered structures can be found in the regular Heusler alloy L21 ($Fm\bar{3}m$) and the inverse Heusler alloy XA ($F\bar{4}3m$), in which the X, Y, and Z atoms are located in distinct sublattices [3, 36]. Disordered structures can also form the Heusler alloys. Examples include B2($Pm\bar{3}m$), where the Y and Z atoms are evenly distributed, A2 ($Im\bar{3}m$), where all the X, Y, and Z atoms are evenly distributed, and other structures [3]. Among Heusler alloys, the Mn_2CoAl compound, which is formed in the inverse Heusler alloy structure (XA structure), is of particular interest because the SGS state was experimentally and theoretically observed in it for the first time [58]. This compound exhibits high magnetic moments ($2 \mu_B$), a high Curie temperature (720 K), a high resistivity (residual resistivity $\sim 440 \mu\Omega \text{ cm}$), a low anomalous Hall coefficient ($\sim 22 \text{ S/cm}$ at 2 K), concentrations of charge carriers ($\sim 10^{20} \text{ cm}^{-3}$), and a Seebeck coefficient ($\sim 2 \mu\text{V/K}$) [58]. If the L21 structure of a regular Heusler alloy forms in the Mn_2CoAl and Mn_2CoGa compounds, the magnetic moment remains at $2 \mu_B$ and the spin-gapless semiconductor state transforms into a half-metallic ferromagnetic state [59]. However, if the B2 or β -Mn structure is observed, the SGS and HMF states do not form in Mn_2CoAl , Mn_2FeAl , and Mn_2NiAl , but rather a metallic density of electronic states is observed [60, 61]. In this case, the magnetic moment in the β -Mn structure of Mn_2CoAl and Mn_2NiAl decreases to 0.56 and $0.32 \mu_B/\text{f.u.}$, respectively, and an antiferromagnetic state with a zero magnetic moment is formed in the B2 structure. The residual electrical resistivity in Mn_2CoAl remains high but is half that observed in XA ($\sim 242 \mu\Omega \text{ cm}$) [58, 60]. Interestingly, the SGS state of the compound is preserved when deviating from the stoichiometry ($Mn_{1.8}Co_{1.2}Al$) [62]. Nowadays, Heusler quaternary alloys are studied intensively [63–65]. The electronic structure and properties of $IrCoTiZ$ Heusler alloys ($Z = \text{Si, Sn, or Pb}$) have been investigated theoretically [65]. These compounds were found to be spin-gapless semiconductors with an energetically stable ferromagnetic phase, a total magnetic moment of $2 \mu_B$, and a Curie temperature above 1000 K. The $IrCoTiSi$ compound exhibits a strong optical response over a wide range of energies, making it an excellent candidate for optoelectronic applications. From the thermal electromotive force (EMF) data, the authors [65] determined that $IrCoTiPb$ and $IrCoTiSn$ exhibit p-type behaviour for both spin projections, whereas $IrCoTiSi$ exhibits mixed behavior (i.e., n-type for spin down and p-type for spin up) [65]. The authors of [64] experimentally studied a quaternary $CoFeMnSn$ alloy that was formed within the $LiMgPdSn$ -type ($F\bar{4}3m$) Heusler structure. The magnetic moment value of $4.04 \mu_B/\text{f.u.}$ corresponds to the Slater-Pauling rule. The Curie temperature is 660 K, the residual resistivity is $\sim 328 \mu\Omega \text{ cm}$, the anomalous Hall coefficient at 2 K is 83.5 S/cm , and the Seebeck coefficient is $5.7 \mu\text{V/K}$ at room temperature. Analysis of the Hall effect and thermal EMF data revealed that holes are the dominant charge carriers. Although the SGS state is most commonly observed in inverse Heusler alloys, the authors of [63] theoretically identified a nearly SGS state in $FeVTaAl$ (type II SGS) and $FeCrZrAl$ (type I SGS) alloys with a regular L21 Heusler structure and an integral magnetic moment at an equilibrium lattice constant [63]. Currently, various 6 materials exhibiting spin-gapless semiconductor properties are being actively investigated, thus making it possible to select a compound with the necessary properties for practical use.

CONCLUSIONS

Thus, we examined two important classes of Heusler alloys: half-metallic ferromagnets (HMFs) and spin-gapless semiconductors (SGSs). HMFs are characterized by the presence of

a gap at the Fermi level for spin-down electronic states and the absence of a gap for spin-up charge carriers. The behaviour of electronic transport in these systems can be modelled with two conduction channels. Spingapless semiconductors are a new class of quantum material with a unique spin polarized band structure. They have a finite band gap only for carriers with one spin direction, and a zero or near-zero gap for carriers with the opposite direction, which makes them promising materials for spintronics devices. nce again, we will outline the latest developments in Heusler alloy research. These trends include searching for and studying new quaternary and double half Heusler alloys (such as XX'YZ and XX'YZZ'), alloys consisting entirely of d metals (known as all-d-metal Heusler compounds), new compounds (such as Al₂MnCu), synthesizing single crystals of Heusler alloys, and studying various effects such as rapid quenching from a melt, thermobaric high-pressure treatment, and high-energy particle irradiation and others. New electronic and magnetic effects include the occurrence of large thermal EMF, the observation of giant Nernst Ettingshausen and anomalous Hall effects, metamagnetic transitions in extremely strong magnetic fields, etc. Thus, new materials based on Heusler alloys and the novel arising phenomena can be used in spintronics and micro- and nanoelectronic devices.

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