



Half-Metallic Heusler Alloys for Spintronic Applications: Materials Design, Properties, and Device Perspectives

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Abstract

Spintronics exploits the spin degree of freedom of electrons in addition to their charge, enabling next-generation electronic devices with enhanced speed, reduced power consumption, and non-volatile functionality. A central challenge in spintronics is the realization of materials with high spin polarization at the Fermi level that remain stable under device operating conditions. Half-metallic Heusler alloys have emerged as leading candidates in this regard due to their unique electronic structures, high Curie temperatures, and structural versatility. This review provides a comprehensive and critical overview of half-metallic Heusler alloys for spintronic applications. We discuss crystallographic families, electronic and magnetic properties, theoretical foundations of half-metallicity, synthesis routes, and the impact of disorder and defects. Special emphasis is placed on their integration into spintronic devices such as magnetic tunnel junctions, giant magnetoresistance structures, and spin-transfer-torque memory. Current challenges and future research directions are outlined, highlighting pathways toward robust room-temperature spin polarization and large-scale device integration.

Keywords - Heusler alloys; half-metallicity; spintronics; magnetic tunnel junctions; spin polarization; magnetoresistance.

1. Introduction

Spintronics exploits the spin degree of freedom of electrons in addition to their charge, enabling electronic devices with enhanced functionality, reduced power consumption, and non-volatility compared with conventional charge-based electronics [4,16]. Key spintronic technologies, including magnetic tunnel junctions (MTJs), giant magnetoresistance (GMR) devices, and spin-transfer-torque magnetic random-access memory (STT-MRAM), rely critically on materials capable of generating highly spin-polarized currents at room temperature [5,16].

Half-metallic ferromagnets constitute an ideal class of materials for spintronic applications because they exhibit metallic behavior for one spin channel while displaying a band gap at the Fermi level for the opposite spin channel, resulting in 100% spin polarization in the ideal case [1]. Since the theoretical prediction of half-metallicity in NiMnSb by de Groot *et al.* [1], extensive efforts have been devoted to identifying materials that combine robust half-metallicity with high Curie temperatures and compatibility with thin-film growth.

Among the various half-metallic systems, Heusler alloys have emerged as particularly attractive candidates due to their versatile chemical compositions, tunable electronic and magnetic properties, and well-defined crystallographic structures [2,3]. Both full-Heusler (X_2YZ) and half-Heusler (XYZ) compounds have been

theoretically predicted and experimentally demonstrated to exhibit half-metallicity, following the Slater–Pauling rule that links their magnetic moments to valence electron count [6,14]. Co-based full-Heusler alloys, such as Co_2MnSi and Co_2FeAl , have shown high Curie temperatures and large spin polarization, making them suitable for room-temperature device operation [10,15].

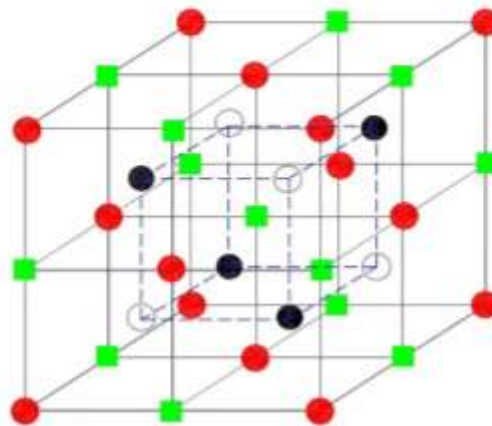
Despite their promising intrinsic properties, the realization of ideal half-metallicity in practical devices remains challenging. Structural disorder, deviations from stoichiometry, and interfacial imperfections can introduce minority-spin states at the Fermi level, significantly reducing spin polarization [8,11,12]. Nevertheless, advances in materials synthesis, epitaxial thin-film growth, and interface engineering have enabled the successful integration of Heusler alloys into MTJs and other spintronic devices, yielding large magnetoresistance ratios and improved device performance [5,10,13].

This review provides a comprehensive overview of half-metallic Heusler alloys for spintronic applications, covering their fundamental electronic structure, materials synthesis strategies, and device implementations. Current challenges and emerging directions, including ferrimagnetic and topological Heusler compounds, are also discussed to outline future pathways toward high-performance spintronic technologies [7,17,18].

2. Materials and Properties

2.1. Classification and Crystal Structures of Heusler Alloys

Heusler alloys are a broad class of intermetallic compounds crystallizing in highly ordered cubic structures and exhibiting diverse electronic and magnetic properties, including half-metallicity, ferrimagnetism, and topological behavior [2,3]. Based on composition and atomic site occupancy, they are commonly classified into full-Heusler, half-Heusler, inverse Heusler, and quaternary Heusler alloys.



Different Heuslers	General Formula	Space Group	Site Occupancy			
			● A	○ B	■ C	● D
Half Heuslers	XYZ	$F\bar{4}3m$ (216)	X	Y		Z
Full Heuslers	X_2YZ	$Fm\bar{3}m$ (225)	X	Y	X	Z
Quaternary Heuslers	$\text{XX}'\text{YZ}$	$F\bar{4}3m$ (216)	X	Y	X'	Z
Inverse Heuslers	X_2YZ	$F\bar{4}3m$ (216)	X	X	Y	Z

Fig.1: Different Heusler structures depending on the site occupancy of the constituent elements X, X', Y and Z at four different Wyckoff sites A, B, C and D.

Full-Heusler alloys (X_2YZ) crystallize in the $L2_1$ structure (space group $Fm\bar{3}m$), consisting of four interpenetrating fcc sublattices [2,6]. Co-based compounds such as Co_2MnSi and Co_2FeAl are prototypical examples, exhibiting high Curie temperatures and half-metallic or near-half-metallic electronic structures that follow the Slater–Pauling rule [6,14,15]. High atomic ordering is essential, as B2 or DO_3 disorder can significantly reduce spin polarization [8,11].

Half-Heusler alloys (XYZ) adopt the $C1_{\beta}$ structure (space group $F\bar{4}3m$), which can be regarded as a full-Heusler structure with one vacant sublattice [2]. The half-metallic character of this class was first predicted in NiMnSb [1]. Although half-Heusler alloys exhibit tunable electronic properties, their lower Curie temperatures and sensitivity to disorder often limit room-temperature spintronic applications [12].

Inverse Heusler alloys (X_2YZ , XA-type) arise from altered site occupancies driven by electronegativity differences and crystallize in the $F\bar{4}3m$ structure [8]. Many Mn_2 -based inverse Heusler compounds exhibit ferrimagnetism with low net magnetic moments, making them attractive for spintronic devices requiring reduced stray fields [17,18].

Quaternary Heusler alloys ($XX'YZ$ or $X_2Y_{1-x}Y'_xZ$) extend compositional flexibility by partial elemental substitution, enabling fine tuning of lattice parameters, magnetic moments, and electronic structure while preserving cubic symmetry [2,3,14].

Overall, the emergence of half-metallicity in Heusler alloys is strongly governed by crystal structure and atomic ordering. Hybridization between transition-metal d states and main-group sp states leads to a minority-spin band gap in well-ordered structures, whereas disorder and antisite defects disrupt this mechanism and degrade half-metallic behavior [6,8,11,12].

2.2. Electronic and Magnetic Properties

2.2.1. Half-Metallicity and Spin Polarization

The half-metallic character arises when the density of states (DOS) shows a band gap in one spin channel and metallic DOS in the other at the Fermi level. This results in near-complete spin polarization—a key requirement for efficient spintronic performance. The nature of half-metallicity in Heusler alloys is sensitive to crystal structure, stoichiometry, and chemical disorder.

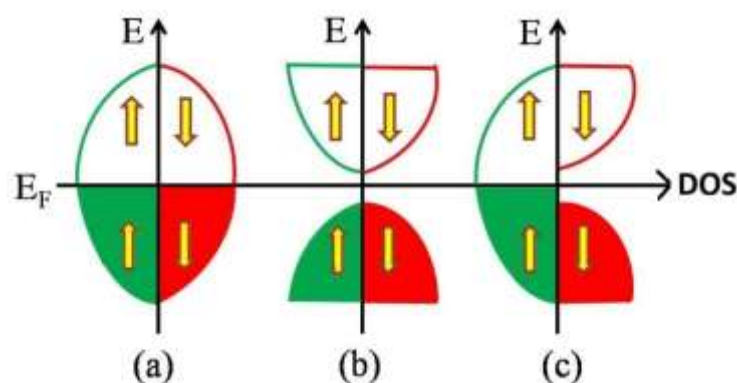


Fig.2: Schematic of density of states (DOS) for a typical (a) metal, (b) Semiconductor and (c) Half-Metals

2.2.2 Curie Temperature and Magnetic Stability

High Curie temperatures (often well above room temperature) are observed in many Co-based Heusler alloys, which is essential for practical device operation. Many Co-based full-Heusler alloys exhibit Curie temperatures exceeding 700 K, making them particularly attractive for device applications. For example, Co_2MnAl exhibits robust ferromagnetism and maintains high spin polarization, making it suitable for MTJs and memory devices.

2.2.3 Slater-Pauling Rule

Magnetic moments in Heusler compounds often follow the Slater-Pauling rule, which relates the magnetic moment to the number of valence electrons. This empirical trend helps guide the prediction of half-metallic behaviour in new compositions.

Although ideal half-metallicity is predicted at zero temperature, thermal excitations such as magnons and electron-phonon interactions reduce spin polarization. Understanding and mitigating these effects remain key

challenges. Magnetocrystalline anisotropy plays a crucial role in device stability and switching behaviour. Heusler alloys offer tunable anisotropy through compositional control and strain engineering.

Density functional theory (DFT) has played a central role in predicting half-metallicity, screening candidate materials, and analysing the effects of strain, disorder, and interfaces. Advanced methods, including hybrid functionals and GW corrections, have further refined predictions of band gaps and spin polarization.

3. Synthesis and Material Optimization

3.1. Bulk Synthesis Methods

Bulk synthesis of Heusler alloys provides the foundational understanding of phase stability, magnetic ordering, and intrinsic electronic properties. The most employed bulk preparation techniques include arc melting, induction melting, and solid-state reaction methods [2,12]. High-purity elemental constituents are typically melted under inert argon atmosphere to minimize oxidation and compositional loss, followed by repeated remelting to improve chemical homogeneity.

Post-synthesis annealing treatments play a critical role in achieving the desired long-range atomic order. Annealing at elevated temperatures (typically 800–1000 °C) promotes diffusion-driven ordering into the ideal $L2_1$ or $C1_\beta$ structures required for half-metallicity [6,14]. Insufficient annealing often results in B2 or DO_3 disorder, which is detrimental to spin polarization and magnetic performance.

Bulk synthesis studies have been particularly important in validating theoretical predictions of half-metallicity and Slater–Pauling behavior in Co-based and Mn-based Heusler alloys [6,14,15].

3.2. Thin-Film Deposition Techniques

For spintronic applications, Heusler alloys must be integrated into thin-film heterostructures, making thin-film synthesis a central research focus. The most widely used deposition techniques include Magnetron sputtering, Molecular beam epitaxy (MBE), Pulsed laser deposition (PLD).

Among these, magnetron sputtering is the most technologically relevant due to its scalability and compatibility with industrial fabrication processes [10,12]. However, sputtered films often require post-deposition annealing to improve crystallinity and atomic ordering.

MBE growth offers superior control over stoichiometry and layer-by-layer growth, enabling the fabrication of epitaxial Heusler films with high structural perfection. MBE-grown Co_2MnSi and Co_2FeAl films have demonstrated enhanced spin polarization and improved interface quality compared to polycrystalline films [9,10].

PLD has also been explored for Heusler alloy growth, although compositional control can be more challenging due to differential ablation rates of constituent elements [12].

3.3. Epitaxial Growth and Substrate Selection

Epitaxial growth is crucial for preserving half-metallicity in thin films by minimizing grain boundaries, strain inhomogeneity, and structural disorder. Common substrates for epitaxial Heusler growth include MgO, which provides good lattice matching and promotes coherent tunnelling in MTJs, GaAs and Ge, for spin injection into semiconductors and Si, for CMOS compatibility.

Among these, MgO has proven particularly effective in achieving high tunnelling magnetoresistance when combined with Co-based Heusler alloys [10,13]. The lattice mismatch between Heusler alloys and substrates can be partially accommodated through strain, which in some cases enhances half-metallicity but may also induce defect formation if not carefully controlled [8].

4. Spintronic Device Applications

4.1. Role of Half-Metallic Materials in Spintronics

The primary objective of spintronic devices is to exploit spin-dependent transport to achieve enhanced functionality compared to conventional electronics. Central to this objective is the availability of materials that can act as efficient spin sources and spin filters, providing highly spin-polarized currents. Half-metallic Heusler alloys are ideally suited for this role due to their theoretically complete spin polarization at the Fermi level and their compatibility with metallic multilayer architectures [1,3,4].

In practical devices, the effectiveness of a spintronic material is not solely determined by its bulk properties but also by its behaviour in thin-film form and at interfaces. Consequently, Heusler alloys have been extensively investigated as active layers in various spintronic device configurations, including magnetic tunnel junctions, giant magnetoresistance structures, and spin-transfer-torque-based memories [5,10,16].

4.2. Magnetic Tunnel Junctions (MTJs)

Magnetic tunnel junctions consist of two ferromagnetic electrodes separated by an ultrathin insulating barrier, typically MgO. The tunnelling current depends on the relative orientation of the magnetizations of the two electrodes, giving rise to tunnelling magnetoresistance (TMR). According to the Jullière model, the TMR ratio is directly related to the spin polarization of the electrodes, making half-metallic Heusler alloys ideal candidates for MTJ applications [5].

Co-based full-Heusler alloys such as Co_2MnSi , Co_2FeAl , and Co_2FeSi have demonstrated particularly high TMR ratios when used as electrodes in MgO-based MTJs [10,13]. The high degree of spin polarization in these materials, combined with coherent tunnelling through crystalline MgO barriers, results in enhanced spin filtering and large TMR values at room temperature.

Experimental studies have shown that achieving high TMR requires not only half-metallic bulk properties but also high atomic ordering and clean interfaces. Structural disorder and interfacial intermixing can introduce minority-spin states at the Fermi level, significantly reducing TMR performance [11,12]. Consequently, MTJ optimization relies heavily on precise materials synthesis and interface engineering.

4.3. Giant Magnetoresistance (GMR) Devices

Giant magnetoresistance is observed in multilayer structures consisting of alternating ferromagnetic and non-magnetic metallic layers, where the electrical resistance depends on the relative magnetization orientation of adjacent ferromagnetic layers. GMR-based devices are widely used in magnetic field sensors and read heads for hard disk drives.

Half-metallic Heusler alloys have been explored as ferromagnetic layers in GMR stacks to enhance spin-dependent scattering and maximize magnetoresistance ratios [4,12]. Compared to conventional ferromagnets such as CoFe, Heusler alloys offer higher intrinsic spin polarization, which translates into improved GMR performance.

Although GMR ratios achieved with Heusler alloys are generally lower than the theoretical limits predicted for ideal half-metals, systematic improvements have been realized through enhanced atomic ordering and optimized multilayer design [2,6]. These results highlight the potential of Heusler alloys in next-generation magnetoresistive sensors.

4.4. Spin-Transfer-Torque and MRAM Applications

Spin-transfer torque (STT) is a phenomenon in which a spin-polarized current exerts a torque on the magnetization of a ferromagnetic layer, enabling current-induced magnetization switching. STT forms the operational basis of STT-MRAM, a leading candidate for universal non-volatile memory.

The efficiency of STT switching depends strongly on the spin polarization of the current. Half-metallic Heusler alloys are therefore attractive for reducing the critical current density required for magnetization reversal [5,16].

Experimental studies have demonstrated that incorporating Co-based Heusler alloys into STT-MRAM structures can significantly improve switching efficiency and thermal stability [10,13].

In addition, Mn-based ferrimagnetic Heusler alloys have attracted attention for STT applications due to their low net magnetization, which reduces stray fields and switching currents while maintaining high spin polarization [17,18].

4.5. Emerging and Advanced Spintronic Applications

Beyond conventional ferromagnetic spintronics, Heusler alloys are increasingly being explored in emerging device concepts such as ultrafast and low-power spintronic devices [17,18], topological Heusler compounds, which host non-trivial band topology and strong spin-orbit coupling, offering new opportunities for dissipation-less spin transport [7], spin caloritronics, where spin currents are generated by thermal gradients, leveraging the unique electronic structure of Heusler alloys [3,16]. These emerging directions significantly broaden the scope of Heusler alloys and position them as multifunctional materials for future spin-based technologies.

4.6. Device-Level Challenges and Perspectives

Despite their promise, several challenges hinder the widespread deployment of Heusler alloys in spintronic devices. These include degradation of spin polarization at interfaces, sensitivity to disorder and off-stoichiometry and thermal stability under high-current operation. Addressing these challenges requires close integration of materials science, device engineering, and theoretical modelling. Continued improvements in thin-film growth, interface control, and materials design are expected to further enhance device performance and reliability [2,16].

5. Conclusion

Half-metallic Heusler alloys represent one of the most promising material classes for spintronic applications due to their unique electronic structure, which enables highly spin-polarized charge transport, high Curie temperatures, and broad compositional tunability [1–3]. Significant progress has been achieved in understanding the fundamental origin of half-metallicity, the role of atomic ordering and defects, and the relationship between materials synthesis and device performance [6,8,11,14]. Advances in thin-film growth and interface engineering have enabled the successful integration of Co-based Heusler alloys into spintronic devices such as magnetic tunnel junctions, giant magnetoresistance structures, and spin-transfer-torque memory elements, demonstrating large magnetoresistive effects and improved energy efficiency [5,10,13,16].

Despite these achievements, the realization of ideal half-metallicity in practical devices remains challenged by thermal effects, chemical disorder, and interfacial degradation, which reduce spin polarization under operating conditions [11,12]. Emerging research directions—including ferrimagnetic and topological Heusler compounds, strain and band-structure engineering, and computationally guided materials discovery—offer promising pathways to overcome these limitations and extend the functionality of Heusler-based spintronic systems [7,17,18]. Continued interdisciplinary efforts spanning materials design, synthesis, characterization, and device engineering will be essential for translating the intrinsic potential of half-metallic Heusler alloys into reliable, scalable technologies for next-generation spintronic applications.

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7. References

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