

**Magnetotransport and Spin Dynamics Investigations
on the Weyl semi-metallic Heusler alloy Co_2MnGa
and its Heterostructures**

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by

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Submitted

In the fulfilment of the requirement of the degree of
the Doctor of Philosophy to the



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Dedicated to my Family

Certificate

This is to certify that the thesis entitled “**Magneto transport and Spin Dynamics Investigations on the Weyl semi-metallic Heusler alloy Co_2MnGa and its Hetero structures**”, which is being submitted by **Ms. Nikita Sharma** to the **Indian Institute of Technology Delhi**, New Delhi, for the award of the degree of **Doctor of Philosophy in Physics**, is a record of bonafide research work carried out by her. She has worked under my supervision and guidance and has fulfilled the requirements for the submission of this thesis, which, in my opinion, has reached the requisite standard.

The results contained in this thesis have not been submitted, in part or full, to any other University or Institute for the award of any degree/diploma.

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Abstract

The topological quantum materials, particularly those exhibiting strong spin-orbit coupling (SOC), have emerged as promising candidates for next-generation spintronic devices owing to their unique surface states, high spin-charge conversion efficiency, and robust magnetic and electronic properties. Among these, the Heusler alloy Co_2MnGa (CMG), a topological Weyl semimetal, offers significant advantages due to its tunable crystallographic ordering, high spin polarization, and potential for integration with other topological or heavy-metal systems.

This thesis focuses on a comprehensive experimental exploration of growth optimization, magnetic transport, and spintronic functionalities in CMG thin films and heterostructures, fabricated using an industry-compatible co-sputtering technique. The thesis is structured into five interconnected studies, each addressing a key scientific challenge related to the integration of topological ferromagnets (FM) in next-generation spintronic devices.

The first part involves optimizing the growth parameters to fabricate both polycrystalline and epitaxial CMG thin films. It is demonstrated that substrate choice and post-deposition thermal treatment critically influence the crystallographic ordering. While films grown on Si(100) predominantly show disordered $A2$ phase, optimized in-situ annealing at 400 °C results in $B2$ ordering. In contrast, epitaxial growth on GaAs(100) at 500 °C with in-situ annealing leads to ordered $B2$ -phase CMG films due to superior lattice matching and thermal compatibility. These results establish a reliable protocol for tuning the atomic order and orientation in CMG films.

Building on the optimized fabrication conditions, the second study explored the magnetic and magnetotransport properties of polycrystalline CMG films with thicknesses ranging from 10–40 nm. The films exhibit soft ferromagnetic, magnetically isotropic behavior with thickness-dependent saturation magnetization and Gilbert damping. The lowest damping constant, $\alpha_{eff} = 6.1(\pm 0.2) \times 10^{-3}$, is achieved in the 40 nm film. The detailed analysis of the anomalous Hall conductivity (AHC) reveals a dominant intrinsic contribution, especially in thicker films, while extrinsic skew scattering becomes more prominent as thickness decreases. The 40 nm film achieves an AHC of 1920 S/cm at 2 K, with nearly 73% attributed to intrinsic Berry curvature effects. The systematic scaling analyses reveal a thickness-dependent competition between intrinsic and extrinsic mechanisms in the anomalous Hall effect (AHE), highlighting the influence of microstructural evolution on spin-dependent transport.

The third study expands this understanding to heterostructures by investigating spin-to-charge conversion via both inverse spin Hall effect (ISHE) and spin Seebeck effect (SSE) in CMG-

based heterostructures with Bi_2Te_3 , Pt, and $\alpha\text{-W}$. Among the systems under study, CMG/ Bi_2Te_3 exhibits the highest spin Hall angle (~ 1.5) despite its higher resistivity, attributed to strong SOC in the topological insulator (TI). Conversely, CMG/W bilayers demonstrate the highest spin mixing conductance due to a clean interface. The thermospin measurements confirm consistency with ISHE results. These findings establish the efficiency of spin-charge conversion in FM/TI and FM/Heavy Metal systems, positioning CMG/ Bi_2Te_3 as a promising candidate for spintronic applications.

In continuation, the fourth chapter investigates the emergence of perpendicular magnetic anisotropy (PMA) and topological transport in CMG/ Bi_2Te_3 heterostructures with ultrathin CMG layers (3–11 nm). PMA is found to be preserved upto 3 nm CMG thickness, with interfacial anisotropy persisting at higher thicknesses. The distinct humps and dips in AHE loops in CMG(7 nm)/ Bi_2Te_3 (10 nm) samples point to the interplay between intrinsic Berry curvature and the strong SOC of Bi_2Te_3 . The sign reversal of the AHE and the appearance of topological Hall signals ($\sim 189 \text{ n}\Omega\cdot\text{cm}$ at 5 K) confirm the presence of non-trivial spin textures, robust even at RT and zero field. Such behavior is absent in CMG/W systems, affirming the essential role of strong SOC and van der Waals coupling in CMG/ Bi_2Te_3 heterostructures.

In the final study, the unconventional magneto-transport behavior of epitaxial CMG (40 nm) films grown on GaAs is explored. The high structural quality is confirmed via ϕ -scan and rocking curve analysis. The X-Ray Diffraction and EXAFS data reveal compressive strain-induced tetragonal distortion, resulting in pronounced out of plane magnetic anisotropy. These epitaxial films exhibit a record high negative longitudinal magnetoresistance ($\sim 2.13\%$ at 5 K) at 5T. The AHE measurements reveal kinks correlated with topological Hall signatures. The temperature dependence of these features and their distinction in origin between magnetoresistance and AHE suggest coexisting real and momentum-space topological phenomena, highlighting the dual topological character of CMG arising from both spin textures and Berry curvature.

Together, these five studies provide a coherent narrative starting from thin-film synthesis and magneto-transport analysis, progressing through spin charge conversion in heterostructures, and culminating in the discovery of topological transport phenomena in epitaxial films. Collectively, this thesis demonstrates that sputtered Co_2MnGa is a versatile and tunable material platform capable of hosting both intrinsic and interfacial topological spin phenomena. This study provides a foundational framework for future application of Heusler-based Weyl semimetals in high-efficiency, room-temperature spintronic devices.

सार

टोपोलॉजिकल क्वांटम मटेरियल्स, विशेष रूप से वे जिनमें मज़बूत स्पिन-ऑर्बिट कपलिंग (SOC) होती है, अपनी अनूठी सतही अवस्थाओं, उच्च स्पिन-चार्ज रूपांतरण दक्षता, और मज़बूत चुंबकीय तथा इलेक्ट्रॉनिक गुणों के कारण अगली पीढ़ी के स्पिन्ट्रॉनिक उपकरणों के लिए अत्यधिक संभावनाशील विकल्प के रूप में उभरे हैं। इनमें से, हीउस्लर एलॉय Co_2MnGa (CMG), जो एक टोपोलॉजिकल वेइल सेमीमेटल है, अपने समायोज्य क्रिस्टलोग्राफिक ऑर्डरिंग, उच्च स्पिन पोलराइज़ेशन, और अन्य टोपोलॉजिकल या हेवी मेटल प्रणालियों के साथ एकीकरण की संभावनाओं के चलते महत्वपूर्ण लाभ प्रदान करता है।

यह शोध प्रबंध CMG थिन फिल्मस और हेटरोस्ट्रक्चर्स में उत्पादन अनुकूलन, चुंबकीय परिवहन, और स्पिन्ट्रॉनिक कार्यात्मकताओं की व्यापक प्रयोगात्मक जांच पर केंद्रित है, जिन्हें एक उद्योग-अनुकूल को-स्पटरिंग तकनीक का उपयोग करके तैयार किया गया है। यह शोध कार्य पाँच आपस में जुड़े अध्ययनों में विभाजित है, जिनमें से प्रत्येक अगली पीढ़ी के स्पिन्ट्रॉनिक उपकरणों में टोपोलॉजिकल फेरोमैग्नेट्स (FM) को एकीकृत करने से संबंधित एक प्रमुख वैज्ञानिक चुनौती को संबोधित करता है।

पहले भाग में, पॉलीक्रिस्टलाइन और एपिटैक्सियल CMG थिन फिल्मस को तैयार करने के लिए ग्रोथ पैरामीटर्स के अनुकूलन पर ध्यान केंद्रित किया गया है। यह दर्शाया गया है कि सब्सट्रेट का चयन और जमावोत्तर तापीय उपचार क्रिस्टलोग्राफिक ऑर्डरिंग को गंभीर रूप से प्रभावित करते हैं। जहाँ Si(100) पर तैयार फिल्में मुख्यतः अव्यवस्थित A2 फेज दर्शाती हैं, वहीं 400 °C पर इन-सिचू ऐनीलिंग से B2 ऑर्डरिंग प्राप्त होती है। इसके विपरीत, GaAs(100) पर 500 °C पर इन-सिचू ऐनीलिंग के साथ एपिटैक्सियल ग्रोथ से अत्यधिक व्यवस्थित B2-फेज वाली CMG फिल्में प्राप्त होती हैं, जो बेहतर लैटिस मेल और तापीय अनुकूलता के कारण होती हैं। ये परिणाम CMG फिल्मस में परमाणु क्रम और उन्मुखता को नियंत्रित करने के लिए एक भरोसेमंद प्रक्रिया स्थापित करते हैं।

अनुकूलित निर्माण स्थितियों पर आधारित दूसरे अध्ययन में, 10–40 nm मोटाई वाली पॉलीक्रिस्टलाइन CMG फिल्मों के चुंबकीय और मैग्नेटोड्रांसपोर्ट गुणों की जांच की गई है। ये फिल्में मोटाई पर निर्भर संतृप्त चुंबकीयत्व और गिल्बर्ट डैम्पिंग के साथ सॉफ्ट फेरोमैग्नेटिक और चुंबकीय रूप से समदिशी व्यवहार दर्शाती हैं। सबसे कम डैम्पिंग कॉन्स्टेंट, $\alpha_{eff} = 6.1(\pm 0.2) \times 10^{-3}$, 40 nm मोटी फिल्म में प्राप्त होती है। एनामलस हॉल कंडक्टिविटी (AHC) का विश्लेषण मोटी फिल्मों में आंतरिक (इन्ट्रिंसिक) योगदान को प्रमुख रूप से दर्शाता है, जबकि पतली फिल्मों में बाहरी (एक्सट्रिंसिक) स्क्यू स्कैटरिंग हावी

होती है। 40 nm फिल्म में 2 K तापमान पर 1920 S/cm की AHC प्राप्त होती है, जिसमें लगभग 73% योगदान आंतरिक बेरी वक्रता से होता है। AHE में इन्ट्रिंसिक और एक्सट्रिंसिक तंत्रों के बीच मोटाई-निर्भर प्रतिस्पर्धा को व्यवस्थित रूप से स्केलिंग विश्लेषण से उजागर किया गया है।

तीसरे अध्ययन में, Bi_2Te_3 , Pt और $\alpha\text{-W}$ के साथ CMG-आधारित हेटरोस्ट्रक्चर्स में इन्वर्स स्पिन हॉल इफेक्ट (ISHE) और स्पिन सीबेक इफेक्ट (SSE) के माध्यम से स्पिन-टू-चार्ज रूपांतरण की जांच की गई है। इन प्रणालियों में, CMG/ Bi_2Te_3 सबसे अधिक स्पिन हॉल एंगल (~ 1.5) दिखाता है, जो टोपोलॉजिकल इंसुलेटर (TI) में मजबूत SOC के कारण है। वहीं, CMG/W द्विपरतें साफ इंटरफेस के कारण उच्चतम स्पिन मिक्सिंग कंडक्टेंस प्रदर्शित करती हैं। थर्मोस्पिन माप ISHE परिणामों के साथ सुसंगत पाए गए हैं। ये निष्कर्ष FM/TI और FM/हेवी मेटल प्रणालियों में स्पिन-चार्ज रूपांतरण की दक्षता को स्थापित करते हैं और CMG/ Bi_2Te_3 को स्पिंट्रॉनिक अनुप्रयोगों के लिए एक प्रबल उम्मीदवार बनाते हैं।

चौथे अध्याय में 3–11 nm मोटाई वाली अल्ट्राथिन CMG परतों के साथ CMG/ Bi_2Te_3 हेटरोस्ट्रक्चर्स में परपेंडिकुलर मैग्नेटिक एनीसोट्रॉपी (PMA) और टोपोलॉजिकल परिवहन की उत्पत्ति की जांच की गई है। पाया गया कि PMA को 3 nm मोटाई तक बनाए रखा जा सकता है, और इंटरफेशियल एनीसोट्रॉपी अधिक मोटाई पर भी बनी रहती है। AHE लूप्स में देखे गए उतार-चढ़ाव और संकेत, विशेषकर CMG(7 nm)/ Bi_2Te_3 (10 nm) सैंपल में, बेरी वक्रता और Bi_2Te_3 की मजबूत SOC के बीच परस्पर क्रिया को दर्शाते हैं। AHE के चिन्ह के उलटने और टोपोलॉजिकल हॉल संकेतों ($\sim 189 \text{ n}\Omega\text{cm}$ @5 K) की उपस्थिति से यह पुष्टि होती है कि गैर-तुच्छ स्पिन बनावट मौजूद हैं, जो कमरे के तापमान और शून्य क्षेत्र पर भी स्थिर रहती हैं। ऐसा व्यवहार CMG/W प्रणालियों में अनुपस्थित है, जिससे यह स्पष्ट होता है कि CMG/ Bi_2Te_3 में मजबूत SOC और वेन डर वॉल्स युग्मन की प्रमुख भूमिका है।

अंतिम अध्ययन में, GaAs पर उगाई गई 40 nm मोटी एपिटैक्सियल CMG फिल्मों में असामान्य मैग्नेटोट्रांसपोर्ट व्यवहार की जांच की गई है। उच्च संरचनात्मक गुणवत्ता को ϕ -स्कैन और रॉकिंग कर्व विश्लेषण द्वारा पुष्टि की गई है। XRD और EXAFS आंकड़े संपीडित तनाव-प्रेरित टेट्रागोनल विकृति को दर्शाते हैं, जिसके परिणामस्वरूप प्रमुख आउट-ऑफ-प्लेन चुंबकीय एनीसोट्रॉपी उत्पन्न होती है। इन एपिटैक्सियल फिल्मों में रिकॉर्ड उच्च नकारात्मक अनुदैर्ध्य मैग्नेटोरेसिस्टेंस ($\sim 2.13\%$ (5 K) at 5T) देखा गया है। AHE माप में टोपोलॉजिकल हॉल संकेतों से संबंधित "किंक्स" पाए गए हैं। तापमान पर इन विशेषताओं की निर्भरता और मैग्नेटोरेसिस्टेंस तथा AHE में उनके अलग-अलग स्रोत स्पेस और

मोमेंटम स्पेस दोनों में टोपोलॉजिकल घटनाओं की सह-अस्तित्व को दर्शाते हैं, जिससे यह स्पष्ट होता है कि CMG में डुअल टोपोलॉजिकल कैरेक्टर विद्यमान है।

इन पाँच अध्ययनों के माध्यम से यह शोध कार्य एक समेकित दृष्टिकोण प्रस्तुत करता है जो थिन फिल्म संश्लेषण और मैग्नेटोट्रांसपोर्ट विश्लेषण से शुरू होकर, हेटरोस्ट्रक्चर्स में स्पिन-चार्ज रूपांतरण और अंततः एपिटैक्सियल फिल्मों में टोपोलॉजिकल परिवहन की खोज तक पहुँचता है। यह शोध यह प्रमाणित करता है कि स्पटर की गई Co_2MnGa एक लचीली और अनुकूलनीय सामग्री प्लेटफॉर्म है, जो आंतरिक और इंटरफेशियल टोपोलॉजिकल स्पिन घटनाओं की मेज़बानी करने में सक्षम है। पॉलीक्रिस्टलाइन और एपिटैक्सियल प्रणालियों में संरचना, मैग्नेटोट्रांसपोर्ट और स्पिन-चार्ज रूपांतरण के बीच प्रणालीबद्ध सहसंबंध हीउस्लर-आधारित वेइल सेमीमेटल्स को उच्च दक्षता वाले कमरे के तापमान पर कार्यरत स्पिन्ट्रॉनिक उपकरणों में प्रयोग के लिए एक मजबूत आधार प्रदान करता है।

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