SPUTTERED GROWTH OF TOPOLOGICAL INSULATORS (Sb2Te3 & Bi2Te3), LIGHT METAL Zr and 2D VAN DER WAALS FERROMAGNET CrTe2 BASED HETEROSTRUCTURES FOR SPIN TRANSPORT STUDIES

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Sputtered Growth of Topological Insulators (Sb_2Te_3 & Bi_2Te_3), Light Metal Zr and 2D van der Waal Ferromagnet $CrTe_2$ based Heterostructures for Spin Transport Studies

by

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submitted

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

INDIAN INSTITUTE OF TECHNOLOGY DELHI

APRIL 2026

Dedicated to my Family

Certificate

This is to certify that the thesis entitled **"Sputtered Growth of Topological Insulators (Sb_2Te_3 & Bi_2Te_3), Light Metal Zr and 2D van der Waal Ferromagnet $CrTe_2$ based Heterostructures for Spin Transport Studies"**, submitted by **Mr. Nakul Kumar (2020PHZ8348)** to the **Indian Institute of Technology Delhi, New Delhi India**, in partial fulfillment of the requirements for the award of the degree of **Doctor of Philosophy in Physics**, embodies the results of original and bona fide research carried out by him under my supervision and guidance. He has satisfactorily fulfilled all the prescribed requirements for the submission of this thesis, which, in my considered opinion, meets the standards expected for the award of the doctoral degree.

Furthermore, I certify that the work presented in this thesis, in whole or in part, has not been submitted to any other University or Institute for the award of any degree or diploma.

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Abstract

Topological quantum materials, enriched by strong spin-orbit interactions and emergent Berry-curvature driven phenomena, promise a transformative pathway toward ultrafast, low-power spin-based information technologies. In this thesis, a comprehensive exploration of spin, orbital, and topological transport in engineered thin-film heterostructures is undertaken to elucidate the mechanisms governing spin-to-charge conversion, orbital current generation, and Berry-curvature induced transport anomalies in two dimensions (2D) van der Waals (vdW) Ferromagnetic (FM) materials.

The thesis first examines interface-engineered Al_2O_3(0001)/Sb_2Te_3(20nm)/Ru(0-12nm)/$Co_{20}Fe_{20}B_{20}$(20nm)/Ru(4nm), TI/FM heterostructures grown via DC magnetron co-sputtering, where a remarkably high spin-to-charge conversion (SCC) efficiency is demonstrated at room temperature. A pronounced enhancement in the charge current density emerges specifically at a 4-nm Ru interlayer thickness, beyond which the SCC efficiency decreases with additional Ru thickness. Quantitative analyses using inverse spin Hall effect (ISHE) reveal nearly 5 times enhancement in SCC upon the insertion of a 4-nm Ru layer, yielding an inverse Edelstein length (λ_{IEE}) of 0.19±0.01 nm comparable to or exceeding reported values in prominent TI/FM systems. Longitudinal spin Seebeck effect (LSSE) measurements corroborate these trends and further disentangle the pure TI-driven Seebeck contribution from the anomalous Nernst effect originating in the ferromagnetic $Co_{20}Fe_{20}B_{20}$ (CFB).

Building upon these insights, the thesis next delves into the simultaneous generation and interconversion of spin and orbital currents in Zr(t=2-20nm)/CFB, Zr(t)/Pt/CFB, and Zr(t)/Pt/CFB/Pt stacks, examined via spin-orbital pumping ferromagnetic resonance (SOP-FMR) and longitudinal spin-orbital Seebeck (LSOSE) measurements. The moderate spin-orbit coupling in CFB enables the concurrent injection of spin and orbital currents into adjacent layers, where their transformation into charge currents is governed by the interplay of ISHE, inverse orbital Hall effect (IOHE), and inverse orbital Rashba-Edelstein effect (IOREE). A striking increase in the orbital-dominated charge conversion appears when a Pt interlayer is introduced at the Zr/CFB interface, resulting in an enhancement of the effective Rashba-Edelstein length λ_{IORE}^{Zr-eff} from ~0.29 nm in Zr(4.5)/CFB(20) to ~1.98 nm and ~1.70 nm in Zr(4.5)/Pt(2)/CFB(20) and Zr(4.5)/Pt(2)/CFB(20)/Pt(2), respectively representing an almost 500% increase. These results, consistently validated through spin-orbital Seebeck

measurements, uncover new avenues wherein orbital angular momentum rather than spin alone can be harnessed for highly efficient charge-current generation, offering an extended design space for orbital-spin hybrid devices.

The thesis then shifts focus to the intriguing physics of layered vdW ferromagnets, with $CrTe_2$ (CT) thin films synthesized for the first time via DC magnetron sputtering. CT films of thickness 20-60 nm grown on Si(100) at 350°C and 400°C yield low-crystallinity (LC) and high-crystallinity (HC) samples, respectively. The crystalline quality profoundly influences the magnetic and electronic behavior, with Curie temperatures T_C of ~140 K (LC) and ~220 K (HC), and robust perpendicular magnetic anisotropy persisting across both series. Notably, the HC-CT films exhibit a dramatic sign reversal in anomalous Hall effect (AHE) below 100 K, attributed to a temperature-driven modification of Berry curvature in momentum space. Electron-magnon scattering dominates their low-temperature resistivity, contrasting with the semiconducting behavior of LC-CT films. Futhermore, the emergent topological Hall signatures in $CrTe_2/Bi_2Te_3$ heterostructures, where the interfacial coupling between the 2D ferromagnet and the topological insulator generates a nontrivial real-space Berry curvature environment conducive to chiral spin textures. The synergy between the strong spin-orbit coupling of Bi_2Te_3 and the intrinsic magnetic order of polyscrystalline and highly crystalline $CrTe_2$ stabilizes nanoscale noncollinear spin configurations, leading to a distinct topological Hall contribution superimposed upon the anomalous and ordinary Hall responses. These findings position $CrTe_2/Bi_2Te_3$ interfaces as promising candidates for topological spintronic memory elements and skyrmion-based logic platforms. These sputtered HC-CT films not only recapitulate but in certain aspects rival the magnetic and topological attributes of MBE- and CVD-grown films, establishing sputtering as an industrially scalable pathway for vdW ferromagnet integration.

Collectively, the investigations presented herein construct a unified and experimentally substantiated picture of how spin, orbital, and topological degrees of freedom intertwine in thin film heterostructures. Through a combination of interface engineering, material-quality optimization, and comprehensive spectroscopic and transport measurements, this work establishes foundational principles and practical pathways for designing next-generation spintronic, orbitronic, and topological devices capable of meeting the stringent demands of quantum-information processing and energy-efficient computing.

सार

प्रबल स्पिन-कक्षा अंतःक्रियाओं (spin-orbit interactions) तथा उद्भूत Berry-curvature प्रेरित घटनाओं से समृद्ध टोपोलॉजिकल क्वांटम पदार्थ, अति-तीव्र (ultrafast), निम्न-ऊर्जा (low-power) स्पिन-आधारित सूचना-प्रौद्योगिकियों की दिशा में एक रूपांतरणकारी मार्ग प्रस्तुत करते हैं। इस शोधप्रबंध में अभिकल्पित पतली-परत हेटरोस्ट्रक्चरों में स्पिन, कक्षीय (orbital), तथा टोपोलॉजिकल परिवहन का व्यापक अध्ययन किया गया है, ताकि स्पिन-से-आवेश रूपांतरण, कक्षीय धारा उत्पत्ति, तथा द्वि-आयामी (2D) वान डर वाल्स (vdW) फेरोमैग्नेटिक (FM) पदार्थों में Berry-curvature प्रेरित परिवहन विसंगतियों को नियंत्रित करने वाले तंत्रों को समझा जा सके।

शोधप्रबंध का प्रथम भाग इंटरफ़ेस-अभिकल्पित $Al_2O_3(0001)/Sb_2Te_3(20nm)/Ru(0\text{-}12nm)/Co_{20}Fe_{20}B_{20}(20nm)/Ru(4nm)$ टोपोलॉजिकल इंसुलेटर/फेरोमैग्नेट (TI/FM) हेटरोस्ट्रक्चरों की जांच करता है, जिन्हें DC magnetron co-sputtering द्वारा विकसित किया गया। इन संरचनाओं में कक्ष-ताप (room temperature) पर अत्यंत उच्च spin-to-charge conversion (SCC) दक्षता प्रदर्शित की गई है। विशेष रूप से 4-nm Ru अंतर-परत मोटाई पर आवेश धारा घनत्व में उल्लेखनीय वृद्धि देखी गई, जिसके बाद Ru की मोटाई बढ़ाने पर SCC दक्षता घटती चली जाती है। Inverse spin Hall effect (ISHE) पर आधारित मात्रात्मक विश्लेषण से स्पष्ट हुआ कि 4-nm Ru परत के समावेशन से SCC में लगभग 5 गुना वृद्धि होती है, जिसके परिणामस्वरूप inverse Edelstein length (λIEE) 0.19±0.01 nm प्राप्त होती है, जो प्रमुख TI/FM प्रणालियों में रिपोर्ट किए गए मानों के तुल्य अथवा उनसे अधिक है। Longitudinal spin Seebeck effect (LSSE) माप इन प्रवृत्तियों की पुष्टि करते हैं और साथ ही शुद्ध TI-प्रेरित Seebeck योगदान को फेरोमैग्नेटिक $Co_{20}Fe_{20}B_{20}$ (CFB) में उत्पन्न anomalous Nernst effect से पृथक भी करते हैं।

इन निष्कर्षों के आधार पर, शोधप्रबंध का अगला भाग Zr(t=2-20nm)/CFB, Zr(t)/Pt/CFB, तथा Zr(t)/Pt/CFB/Pt संरचनाओं में स्पिन तथा कक्षीय धाराओं की समकालिक उत्पत्ति और परस्पर रूपांतरण का अध्ययन करता है। इन प्रणालियों की जांच spin-orbital pumping ferromagnetic resonance (SOP-FMR) तथा longitudinal spin-orbital Seebeck (LSOSE) मापों द्वारा की गई। CFB में मध्यम spin-orbit coupling होने के कारण स्पिन और कक्षीय दोनों धाराएँ समीपवर्ती परतों में एक साथ प्रविष्ट हो सकती हैं, जहाँ उनका आवेश धारा में रूपांतरण ISHE, inverse orbital Hall effect (IOHE), तथा inverse orbital Rashba-Edelstein effect (IOREE) की परस्पर क्रिया द्वारा नियंत्रित होता है। विशेष रूप से, जब Zr/CFB इंटरफ़ेस पर Pt अंतर-परत जोड़ी गई, तब orbital-dominated charge conversion में उल्लेखनीय वृद्धि देखी गई। इसके फलस्वरूप प्रभावी Rashba-Edelstein length λ_{IORE}^{Zr-eff} लगभग 0.29 nm [Zr(4.5)/CFB(20)] से बढ़कर लगभग 1.98 nm तथा लगभग 1.70 nm [क्रमशः Zr(4.5)/Pt(2)/CFB(20) और Zr(4.5)/Pt(2)/CFB(20)/Pt(2)] हो गई, जो लगभग 500% वृद्धि का द्योतक है। ये परिणाम, जिन्हें spin-orbital Seebeck मापों द्वारा भी सुसंगत रूप से सत्यापित किया गया, इस तथ्य को उद्घाटित करते हैं कि केवल स्पिन ही नहीं, बल्कि कक्षीय कोणीय संवेग (orbital angular momentum) को भी अत्यधिक दक्ष आवेश-धारा जनन हेतु उपयोग में लाया जा सकता है। इस प्रकार यह कार्य orbital-spin hybrid युक्तियों के लिए एक विस्तृत अभिकल्पना-क्षेत्र प्रस्तुत करता है।

इसके पश्चात शोधप्रबंध का ध्यान स्तरित vdW फेरोमैग्नेटों के रोचक भौतिकी-विज्ञान की ओर केंद्रित होता है, जहाँ $CrTe_2$ (CT) पतली परतों का प्रथम बार DC magnetron sputtering द्वारा संश्लेषण किया गया। Si(100) पर 350°C और 400°C पर विकसित 20-60 nm मोटाई वाली CT परतों ने क्रमशः निम्न-स्फटिकीयता (low-crystallinity, LC) तथा उच्च-स्फटिकीयता (high-crystallinity, HC) नमूने उत्पन्न किए। स्फटिकीय गुणवत्ता ने इनके चुम्बकीय और वैद्युतिक व्यवहार पर गहरा प्रभाव डाला; LC तथा HC नमूनों के लिए Curie तापक्रम (TC) क्रमशः लगभग 140 K और लगभग 220 K प्राप्त हुआ, और दोनों श्रेणियों में प्रबल perpendicular magnetic anisotropy विद्यमान रही। विशेष रूप से HC-CT परतों में 100 K से नीचे anomalous Hall effect (AHE) के चिह्न में नाटकीय उलटाव देखा गया, जिसे momentum space में Berry curvature के ताप-प्रेरित परिवर्तन से संबद्ध किया गया है। निम्न ताप पर इनके प्रतिरोध में electron-magnon scattering प्रमुख भूमिका निभाती है, जो LC-CT परतों के अर्धचालकीय व्यवहार से स्पष्टतः भिन्न है।

इसके अतिरिक्त, $CrTe_2/Bi_2Te_3$ हेटरोस्ट्रक्चरों में उद्भूत topological Hall signatures भी देखे गए, जहाँ 2D फेरोमैग्नेट और टोपोलॉजिकल इंसुलेटर के मध्य अंतःपृष्ठीय युग्मन एक ऐसी nontrivial real-space Berry curvature अवस्था निर्मित करता है जो chiral spin textures के लिए अनुकूल है। Bi_2Te_3 की प्रबल spin-orbit coupling तथा polycrystalline और highly crystalline $CrTe_2$ की अंतर्निहित चुम्बकीय व्यवस्था के परस्पर समन्वय से नैनो-स्तरीय noncollinear spin configurations स्थिर होती हैं, जिसके परिणामस्वरूप anomalous तथा ordinary Hall response के ऊपर आरोपित एक स्पष्ट topological Hall contribution प्राप्त होती है। ये निष्कर्ष $CrTe_2/Bi_2Te_3$ अंतःपृष्ठों को topological spintronic memory तत्वों तथा skyrmion-आधारित logic platforms के लिए अत्यंत आशाजनक प्रत्याशी के रूप में स्थापित करते हैं। ये sputtered HC-CT परतें न केवल MBE और CVD द्वारा विकसित परतों के चुम्बकीय और टोपोलॉजिकल गुणों की पुनरावृत्ति करती हैं, बल्कि कुछ पक्षों में उनसे प्रतिस्पर्धा भी करती हैं। इस प्रकार sputtering, vdW फेरोमैग्नेट एकीकरण के लिए एक औद्योगिक रूप से स्केलेबल मार्ग के रूप में स्थापित होती है।

समग्र रूप से, इस शोधप्रबंध में प्रस्तुत अध्ययन यह स्पष्ट और प्रायोगिक रूप से प्रमाणित चित्र निर्मित करते हैं कि किस प्रकार स्पिन, कक्षीय और टोपोलॉजिकल स्वतंत्रताएँ पतली-परत हेटरोस्ट्रक्चरों में परस्पर गुंथी हुई हैं। इंटरफ़ेस अभिकल्पना, पदार्थ-गुणवत्ता अनुकूलन, तथा व्यापक स्पेक्ट्रोस्कोपिक और परिवहन मापों के संयोजन द्वारा यह कार्य अगली पीढ़ी की spintronic, orbitronic, और topological युक्तियों के अभिकल्पन हेतु मूलभूत सिद्धांतों और व्यावहारिक मार्गों की स्थापना करता है, जो quantum-information processing तथा ऊर्जा-दक्ष कंप्यूटिंग की कठोर आवश्यकताओं को पूरा करने में सक्षम हो सकती हैं।

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