Magnetotransport And Spin Dynamics Investigations On The Te-based Topological materials And Their Heterostructures Fabricated By Sputtering

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by

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Dedicated

to

My Parents

and

My lovely Sisters

Certificate

This is to certify that the thesis entitled **"Magnetotransport and Spin Dynamics Investigations on the Te-based Topological materials and their Heterostructures Fabricated by Sputtering"**, which is being submitted by **Mr. Lalit Pandey** 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 him. He 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

Topological materials (TMs) belong to the class of quantum materials whose physical properties, unlike the usual classical materials, cannot be explained by the theory of spontaneous symmetry breaking. They are classified based on a topological invariant (ν), which is defined by the band structure topology. A material is considered topological if it has a non-zero value of ν, which can be induced by introducing high spin-orbit coupling (SOC) to a previously non-topological material with $\nu = 0$. This can cause an inversion of conduction and valence bands, leading to a quantum phase transition. During this transition, topological surface states (TSSs) emerge, which exhibit unique properties such as Dirac Fermionic nature, helical spin polarization, and forbidden backscattering of electrons. These states are symmetry protected, making them robust against scattering off from the non-magnetic impurities. Literature reviews on spin-orbit torque devices based on the heterostructure of TMs and ferromagnets (FMs) suggest that TMs possess high values of spin-charge conversion efficiency, which can exceed 50%, resulting in much lower values of switching current compared to those in the heavy metals. Therefore, TMs have attracted attention for their exotic properties and high spin-charge conversion efficiencies, making them promising materials for microelectronics and spintronics applications. Heterostructures composed of TMs and other materials, such as FMs and superconductors, exhibit interesting phenomena such as the quantum anomalous Hall effect, topological superconductivity, and topological magnetoelectric effect. These structures can also host quasiparticles associated with Majorana bound states, Weyl Fermions, and magnetic monopoles, among other striking features and phenomena.

Even having these many exotic properties, the TMs face several difficulties, which hinder the integration of TMs in future microelectronic devices. Some of these difficulties are the absence of a growth procedure to synthesize excellent quality and stoichiometric TMs using industrial-friendly sputtering technique and simple investigation protocol to evaluate topological materials using easily accessible magneto-transport measurements. This thesis is aimed to possibly explore the solutions to these difficulties. For this, the Bi_xTe_y (BT) topological system is grown on Si(100) using co-sputtering. Firstly, the control of altering the Te composition over a wide range in a systematic manner is achieved by closely examining the changes in the structural and compositional analyses vis a vis the growth conditions. Then, by using the same recipe employed in synthesising Bi_xTe_y, thin films of varieties of other TMs and 2D FMs such as Sb_2Te_3, Cr_2Te_3, BiSb, $NiTe_2$, $PtTe_2$, $PdTe_2$ and $PdBi_2$ are successfully synthesized using DC-magnetron sputtering technique only.

For the second challenge, highly oriented Bi_2Te_3 thin films are synthesized on the Al_2O_3 (0001) substrate using the same co-sputtering technique. The detailed investigation of the temperature and magnetic field dependent transport properties of these high quality Bi_2Te_3 thin films is undertaken. The different models such as modified Hikami-Larkin-Nagoaka (HLN), Lu-Shen and modified Al'tshuler-Aronov model are used to analyze the temperature dependent, and in-plane and out-of-plane magnetic field dependent transport behaviour. From the analyses of such data, different topological parameters such as coherency factor (α), decoherence factor (p), Berry phase (ϕ_B), mass term (m) are estimated.

After suggesting possible solutions to challenges that hinder the implementation of TMs in future electronic devices, the proposed growth methodology and investigation protocols is utilized in this thesis work to find answer to some fundamental questions. Some of these questions are – what is the influence of topological properties of TMs by tuning the concentration of chalcogenides (Te or Se), and what is the incorporation of magnetic and non-magnetic elements during the growth of topological materials? To answer these queries, first, using the structural properties information on different Te composition Bi_xTe_y binary alloys thin films, the samples are divided into three different Te-concentration phases namely Te-deficient (TD), Te-stoichiometric (TS), and Te-excessive (TE). It is shown using the magneto-transport measurement on these three samples that through the tuning of Te-concentration, one can manipulate the topological properties of TMs. These experimental findings are further supported by first principle theoretical calculations. For the second query, the Bi_2Te_3 films were intercalated with Cr as magnetic element and Au as non-magnetic element. The different topological parameters are estimated by performing and analyzing the magneto-transport data on pristine, Cr-doped and Au-doped Bi_2Te_3 films using the modified HLN and Lu-Shen models. Based on the estimated parameters, it is demonstrated that the topological properties got significantly suppressed in Cr-doped BT films compared to the pristine BT films while slightly enhanced in the Au-doped BT films. This study correlates well to the well-established phenomenon of suppression of topological properties in the presence of magnetic impurities due to the breaking of time reversal symmetry (TRS) in the topological systems.

After answering these fundamental questions, the thesis work is further extended to examine the spin dynamics and spin-charge conversion efficiency in the heterostructure of TMs and FMs fabricated using sputtering techniques. The ferromagnetic resonance (FMR) measurements are performed on these heterostructure samples and the variation of α_{eff} and $\mu_0 M_{eff}$ with the change in thickness of the FM films of $Co_{60}Fe_{20}B_{20}$ (CFB) in them are investigated. The spin dynamic results show that the effective spin mixing conductance is overestimated in the in-plane FMR due to the two-magnon scattering (TMS) contribution in damping. From the analysis of the out-of-plane FMR, it is found that the effective spin mixing conductance at room temperature to be 13.13 nm^{-2} and 4 nm^{-2} with and without TMS contribution, respectively. Additionally, the effective spin-mixing conductance value gradually increases with decreasing temperature, which is in accordance with the surface coherence length of BT. These findings show the influence of spin-polarized topological surface states on the magnetization dynamics of ferromagnets when placed adjacent to topological materials. The excellent quality of the sputter-grown ferromagnet-topological insulator heterostructure and its efficient spin-charge conversion performance demonstrate the topological materials' importance in future magnetic memory devices. The spin dynamics measurements are also performed on the $PdTe_2/Co_{60}Fe_{20}B_{20}$ bilayers. The effective spin mixing conductance and spin current density are found to be $\sim 2 \times 10^{20}\, m^{-2}$ and $2.5\, MAm^{-2}$, respectively in $PdTe_2/Co_{60}Fe_{20}B_{20}$ bilayers which are quite larger than those observed in HM/FM bilayers, and comparable to those reported for MBE-grown TI/FM heterostructures.

Thus, this thesis work proposes a growth methodology to synthesize TMs by an industry-friendly sputtering technique which yielded a very good success on making highly oriented and good quality films of varieties of TMs. Moreover, this work provides a unified and comprehensive account of a detailed interpretation of the magnetotransport data of a topological insulator system using different theoretical models available in literature, which is important both from the fundamental and technological development of this field in the future. Further, this thesis demonstrates an effective way to manipulate quantum properties such as topological characteristics of the materials by tuning chalcogenides compositions and by incorporating magnetic and non-magnetic elements during the growth of the topological system. Finally, this thesis demonstrates the efficient spin-charge conversion performance in sputtered grown TMs/FMs bilayers which paves the way to implement topological materials in future spintronics devices.

सार

टोपोलॉजिकल मैटेरियल्स क्वांटम सामग्रियों के वर्ग से संबंधित हैं, जिनके भौतिक गुण, सामान्य मैटेरियल्स सामग्रियों के विपरीत, स्पोन्तनौउस स्य्म्मेत्र्य ब्रेकिन्ग के सिद्धांत द्वारा नहीं समझाए जा सकते हैं। उन्हें टोपोलॉजिकल इनवेरिएंट (ν) के आधार पर वर्गीकृत किया गया है, जिसे बैंड संरचना टोपोलॉजी द्वारा परिभाषित किया जाता है। एक सामग्री को टोपोलॉजिकल माना जाता है यदि इसमें ν का गैर-शून्य मान है, जिसे $\nu = 0$ के साथ पहले गैर-टोपोलॉजिकल मैटेरियल्स में उच्च स्पिन-ऑर्बिट कपलिंग पेश करके प्रेरित किया जा सकता है। इससे कंडक्शन और वैलेंस बैंड में व्युत्क्रमण हो सकता है, जिससे क्वांटम फेज ट्रांजिशन हो सकता है। इस ट्रांजिशन के दौरान, टोपोलॉजिकल सरफेस स्टेट्स उभरते हैं, जो डायराक फर्मियोनिक प्रकृति, हेलिकल स्पिन ध्रुवीकरण और इलेक्ट्रॉनों के निषिद्ध बैकस्कैटरिंग जैसे अद्वितीय गुणों का प्रदर्शन करते हैं। ये अवस्थाएं समरूपता से संरक्षित हैं, जो उन्हें गैर-चुंबकीय अशुद्धियों से बिखरने के खिलाफ मजबूत बनाती हैं। टोपोलॉजिकल मैटेरियल्स और फेरोमैग्नेट्स के हेटरोस्ट्रक्चर के आधार पर स्पिन-ऑर्बिट टॉर्क उपकरणों पर साहित्यिक समीक्षा से पता चलता है कि टोपोलॉजिकल मैटेरियल्स में स्पिन-चार्ज रूपांतरण दक्षता के उच्च मूल्य होते हैं, जो 50% से अधिक हो सकते हैं, जिसके परिणामस्वरूप स्विचिंग करंट का मान भारी धातुएँ. की तुलना में बहुत कम होता है। इसलिए टोपोलॉजिकल मैटेरियल्स ने अपने अद्भुत गुणों और उच्च स्पिन-चार्ज रूपांतरण क्षमता के लिए ध्यान आकर्षित किया है, जिससे वे माइक्रोइलेक्ट्रॉनिक्स और स्पिंट्रोनिक्स अनुप्रयोगों के लिए आशाजनक सामग्री बन गए हैं। टोपोलॉजिकल मैटेरियल्स और अन्य मैटेरियल्स, जैसे फेरोमैग्नेट्स और सुपरकंडक्टर्स से बने हेटरोस्ट्रक्चर, क्वांटम विसंगतिपूर्ण हॉल प्रभाव, टोपोलॉजिकल सुपरकंडक्टिविटी और टोपोलॉजिकल मैग्नेटोइलेक्ट्रिक प्रभाव जैसी दिलचस्प घटनाएं प्रदर्शित करते हैं। ये संरचनाएं अन्य हड़ताली विशेषताओं और घटनाओं के अलावा, मेजराना बाउंड स्टेट, वेइल फर्मियन और चुंबकीय मोनोपोल से जुड़े क्वासिपार्टिकल्स की भी मेजबानी कर सकती हैं।

इतनी सारी अद्भुत गुणों के होते हुए भी, टोपोलॉजिकल मैटेरियल्स को कई कठिनाइयों का सामना करना पड़ता है, जो भविष्य के माइक्रोइलेक्ट्रॉनिक उपकरणों में टोपोलॉजिकल मैटेरियल्स के एकीकरण में बाधा बनती हैं। इनमें से कुछ कठिनाइयाँ औद्योगिक-अनुकूल स्पटरिंग तकनीक और आसानी से सुलभ मैग्नेटो-ट्रांसपोर्ट माप का उपयोग करके टोपोलॉजिकल सामग्रियों का मूल्यांकन करने के लिए सरल जांच प्रोटोकॉल का उपयोग करके उत्कृष्ट गुणवत्ता और स्टोइकोमेट्रिक टोपोलॉजिकल मैटेरियल्स को संश्लेषित करने के लिए विकास प्रक्रिया की अनुपस्थिति हैं। इस थीसिस का उद्देश्य संभवतः इन कठिनाइयों के समाधान का पता लगाना है। इसके लिए, Bi_xTe_y (BT) टोपोलॉजिकल सिस्टम को सह-स्पटरिंग का उपयोग करके Si(100) पर छलरचना किया गया है। सबसे पहले, एक विस्तृत श्रृंखला में टेल्यूरियम संरचना को व्यवस्थित तरीके से बदलने का नियंत्रण विकास की स्थितियों की तुलना में संरचनात्मक और संरचनागत विश्लेषणों में परिवर्तनों की बारीकी से जांच करके प्राप्त किया गया है। फिर, Bi_xTe_y

को संश्लेषित करने में नियोजित उसी नुस्खा का उपयोग करके, अन्य टोपोलॉजिकल मैटेरियल्स और 2D फेरोमैग्नेट्स जैसे Sb_2Te_3, Cr_2Te_3, BiSb, $NiTe_2$, $PtTe_2$, $PdTe_2$ और $PdBi_2$ की किस्मों की पतली फिल्मों को केवल DC–मैग्नेट्रॉन स्पटरिंग तकनीक का उपयोग करके सफलतापूर्वक संश्लेषित किया गया है।

दूसरी चुनौती के लिए, अत्यधिक उन्मुख Bi_2Te_3 पतली फिल्मों को उसी सह-स्पटरिंग तकनीक का उपयोग करके Al_2O_3 (0001) सब्सट्रेट पर संश्लेषित किया गया है। इन उच्च गुणवत्ता वाली Bi_2Te_3 पतली फिल्मों के तापमान और चुंबकीय क्षेत्र पर निर्भर ट्रांसपोर्ट गुणों की विस्तृत जांच की गई है। संशोधित हिकामी-लार्किन-नागोका (एचएलएन), लू-शेन और संशोधित अल्टशुलर-अरोनोव मॉडल जैसे विभिन्न मॉडलों का उपयोग तापमान पर निर्भर, और सतह के समानांतर और सतह के लंबवत चुंबकीय क्षेत्र पर निर्भर ट्रांसपोर्ट व्यवहार का विश्लेषण करने के लिए किया गया है। ऐसे डेटा के विश्लेषण से, सुसंगतता कारक (α), डीकोहेरेंस कारक (p), बेरी चरण (ϕ_B), द्रव्यमान अवधि (m) जैसे विभिन्न टोपोलॉजिकल मापदंडों का अनुमान लगाया गया है।

भविष्य के इलेक्ट्रॉनिक उपकरणों में टोपोलॉजिकल मैटेरियल्स के कार्यान्वयन में बाधा डालने वाली चुनौतियों के संभावित समाधान सुझाने के बाद, कुछ बुनियादी सवालों के जवाब खोजने के लिए इस थीसिस कार्य में प्रस्तावित विकास पद्धति और जांच प्रोटोकॉल का उपयोग किया गया है। इनमें से कुछ प्रश्न हैं - चॉकोजेनाइड्स (टेल्यूरियम (Te) या सेलेनियम (Se)) की सांद्रता को ट्यून करके टोपोलॉजिकल मैटेरियल्स के टोपोलॉजिकल गुणों का प्रभाव क्या है, और टोपोलॉजिकल सामग्रियों के विकास के दौरान चुंबकीय और गैर-चुंबकीय तत्वों का समावेश क्या है? इन प्रश्नों का उत्तर देने के लिए, सबसे पहले, विभिन्न Te संरचना Bi_xTe_y बाइनरी मिश्र धातु पतली फिल्मों पर संरचनात्मक गुणों की जानकारी का उपयोग करके, नमूनों को तीन अलग-अलग Te–एकाग्रता चरणों में विभाजित किया जाता है, अर्थात् Te-deficient (TD), Te-stoichiometric (TS), और Te-excessive (TE)। इन तीन नमूनों पर मैग्नेटो-ट्रांसपोर्ट माप का उपयोग करके यह दिखाया गया है कि Te-एकाग्रता की ट्यूनिंग के माध्यम से, कोई टोपोलॉजिकल मैटेरियल्स के टोपोलॉजिकल गुणों में हेरफेर कर सकता है। इन प्रयोगात्मक निष्कर्षों को प्रथम सिद्धांत सैद्धांतिक गणना द्वारा आगे समर्थित किया गया है। दूसरी जिज्ञासा के लिए, Bi_2Te_3 फिल्मों को चुंबकीय तत्व के रूप में क्रोमियम (Cr) और गैर-चुंबकीय तत्व के रूप में सोना (Au) के साथ जोड़ा गया था। संशोधित एचएलएन और लू-शेन मॉडल का उपयोग करके बिना डोप्ड, Cr-डोप्ड और Au-डोप्ड Bi_2Te_3 फिल्मों पर मैग्नेटो-ट्रांसपोर्ट डेटा का प्रदर्शन और विश्लेषण करके विभिन्न टोपोलॉजिकल मापदंडों का अनुमान लगाया जाता है। अनुमानित मापदंडों के आधार पर, यह प्रदर्शित किया गया है कि बिना डोप्ड Bi_2Te_3 फिल्मों की तुलना में Cr-डोप्ड Bi_2Te_3 फिल्मों में टोपोलॉजिकल गुणों को काफी हद तक दबा दिया गया है, जबकि Au-डोप्ड Bi_2Te_3 फिल्मों में थोड़ा बढ़ाया गया है। यह अध्ययन टोपोलॉजिकल सिस्टम में टाइम रिवर्सल समरूपता के टूटने के कारण चुंबकीय अशुद्धियों की उपस्थिति में टोपोलॉजिकल गुणों के दमन की अच्छी तरह से स्थापित घटना से संबंधित है।

इन बुनियादी सवालों के जवाब देने के बाद, स्पटरिंग तकनीकों का उपयोग करके निर्मित टोपोलॉजिकल मैटेरियल्स और फेरोमैग्नेट्स के हेटरोस्ट्रक्चर में स्पिन गतिशीलता और स्पिन-चार्ज रूपांतरण दक्षता की जांच करने के लिए थीसिस कार्य को आगे बढ़ाया गया है। इन हेटरोस्ट्रक्चर नमूनों पर फेरोमैग्नेटिक रेज़ोनेंस (एफएमआर) माप किए जाते हैं और उनमें $Co_{60}Fe_{20}B_{20}$ (CFB) की रोमैग्नेटिक फिल्मों की मोटाई में परिवर्तन के साथ α_{eff} और $\mu_0 M_{eff}$ की भिन्नता की जांच की जाती है। स्पिन डायनेमिक परिणाम बताते हैं कि प्रभावी स्पिन मिश्रण चालन को इन-प्लेन एफएमआर में डम्पिंग में दो-मैग्नन स्कैटरिंग (टीएमएस) योगदान का कम करके आंका गया है। आउट-ऑफ़-प्लेन एफएमआर के विश्लेषण से, यह पाया गया है कि कमरे के तापमान पर प्रभावी स्पिन मिश्रण चालन क्रमशः टीएमएस योगदान के साथ और उसके बिना $13.13\ nm^{-2}$ और $4\ nm^{-2}$ है। इसके अतिरिक्त, प्रभावी स्पिन-मिश्रण चालन मूल्य धीरे-धीरे घटते तापमान के साथ बढ़ता है, जो Bi_2Te_3 की सतह सुसंगतता लंबाई के अनुसार है। ये निष्कर्ष टोपोलॉजिकल सामग्रियों के निकट रखे जाने पर फेरोमैग्नेट्स के चुंबकीयकरण गतिशीलता पर स्पिन-ध्रुवीकृत टोपोलॉजिकल सतह स्थितियों के प्रभाव को दर्शाते हैं। स्पटर-विकसित फेरोमैग्नेट-टोपोलॉजिकल इंसुलेटर हेटरोस्ट्रक्चर की उत्कृष्ट गुणवत्ता और इसका कुशल स्पिन-चार्ज रूपांतरण प्रदर्शन भविष्य के चुंबकीय मेमोरी उपकरणों में टोपोलॉजिकल सामग्रियों के महत्व को प्रदर्शित करता है। स्पिन डायनेमिक्स मापन $PdTe_2/Co_{60}Fe_{20}B_{20}$ बाइलेयर्स पर भी किया जाता है। $PdTe_2/Co_{60}Fe_{20}B_{20}$ बाइलेयर में प्रभावी स्पिन मिश्रण चालन और स्पिन धारा घनत्व क्रमशः $\sim 2 \times 10^{20}\, m^{-2}$ और $2.5\, MAm^{-2}$ पाए जाते हैं, जो HM/FM में देखी गई तुलना में काफी बड़े हैं। फेरोमैग्नेट बाइलेयर, और आण्विक किरण एपिटैक्सी तकनीक निर्मित फेरोमैग्नेट- टोपोलॉजिकल मैटेरियल्स हेटरोस्ट्रक्चर के लिए रिपोर्ट किए गए साहित्य के बराबर है।

इस प्रकार, यह थीसिस कार्य एक उद्योग-अनुकूल स्पटरिंग तकनीक द्वारा टोपोलॉजिकल मैटेरियल्स को संश्लेषित करने के लिए एक विकास पद्धति का प्रस्ताव करता है जिससे टोपोलॉजिकल मैटेरियल्स की किस्मों की उच्च उन्मुख और अच्छी गुणवत्ता वाली फिल्में बनाने में बहुत अच्छी सफलता मिली। इसके अलावा, यह कार्य साहित्य में उपलब्ध विभिन्न सैद्धांतिक मॉडलों का उपयोग करके टोपोलॉजिकल इंसुलेटर सिस्टम के मैग्नेटोट्रांसपोर्ट डेटा की विस्तृत व्याख्या का एकीकृत और व्यापक विवरण प्रदान करता है, जो भविष्य में इस क्षेत्र के मौलिक और तकनीकी विकास दोनों के लिए महत्वपूर्ण है। इसके अलावा, यह थीसिस चॉकोजेनाइड्स रचनाओं को ट्यून करके और टोपोलॉजिकल सिस्टम के विकास के दौरान चुंबकीय और गैर-चुंबकीय तत्वों को शामिल करके सामग्री की टोपोलॉजिकल विशेषताओं जैसे क्वांटम गुणों में हेरफेर करने का एक प्रभावी तरीका प्रदर्शित करती है। अंत में, यह थीसिस स्पटर किए गए विकसित फेरोमैग्नेट- टोपोलॉजिकल मैटेरियल्स बाइलेयर्स में कुशल स्पिन-चार्ज रूपांतरण प्रदर्शन को प्रदर्शित करती है जो भविष्य के स्पिंट्रोनिक्स उपकरणों में टोपोलॉजिकल सामग्रियों को लागू करने का मार्ग प्रशस्त करती है।

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