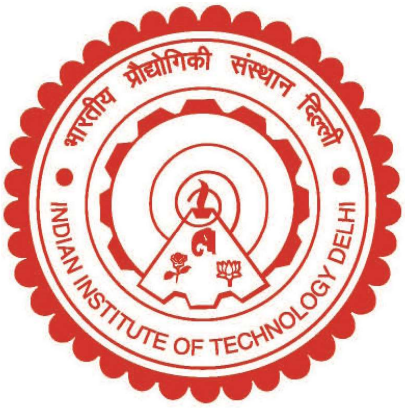
SPIN-TO-CHARGE CONVERSION AND SPIN-ORBIT TORQUE IN HETEROSTRUCTURES UTILIZING VAN DER WAALS MATERIALS AND NON-COLLINEAR ANTIFERROMAGNETS

HIMANSHU



Department of Physics

INDIAN INSTITUTE OF TECHNOLOGY DELHI

May 2024

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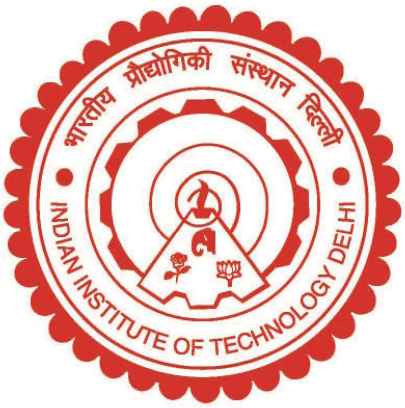
by

HIMANSHU

Department of Physics

Submitted

in fulfillment of the requirements for the degree of Doctor of Philosophy to the



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Certificate

This is to certify that the thesis entitled, **Spin-to-Charge Conversion and Spin-Orbit Torque in Heterostructures utilizing Van der Waals Materials and Non-Collinear Antiferromagnets**, is being submitted by **Mr. Himanshu** to the Department of Physics, Indian Institute of Technology Delhi, for the award of the degree of **Doctor of Philosophy**. This work is a record of bonafide research conducted by him. He has worked under my supervision and guidance and has fulfilled all the requirements for the submission of this thesis. In my opinion, the work has reached the requisite standard.

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

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This thesis is dedicated to my parents.

Himanshu

Abstract

This thesis explores spin-to-charge conversion and spin-orbit torques (SOTs) in two novel classes of quantum materials that are interesting from both fundamental and applied perspectives. The focus is on two significant classes of materials: van der Waals materials and non-collinear antiferromagnets. These quantum materials exhibit unique spin-dependent characteristics, which can be exploited to obtain a substantial tunable spin-charge interconversion efficiency and generate unconventional spin currents. These properties make them highly suitable for applications requiring spin-charge conversion and SOT.

Firstly, large-area van der Waals materials are explored for spin-to-charge conversion and SOT. A systematic study of spin-to-charge conversion at the interface of $Ni_{80}Fe_{20}$ and large area monolayer (ML) transition metal dichalcogenides (TMDs) - MoS_2, $MoSe_2$, WS_2, and WSe_2 - is conducted. Large spin-to-charge conversion is observed in these systems, with efficiencies scaling with the spin-orbit coupling of the ML TMD layer, consistent with the inverse Rashba-Edelstein effect. The efficiency of W-based TMDs exceeds that of Mo-based TMDs. Subsequently, high-quality two-dimensional growth of few-layer SnS on CMOS-compatible Si/SiO_2 substrates using the pulsed laser deposition (PLD) technique is demonstrated. Spin-torque ferromagnetic resonance (STFMR) experiments on SnS(1.3 nm)/$Ni_{80}Fe_{20}$(8 nm) devices reveal evidence of an unconventional SOT. The growth of the α-phase of another van der Waals material, GeTe, on Si(111) substrates is optimized using PLD. Angle-resolved STFMR reveals a substantial in-plane damping-like torque in GeTe/$Ni_{80}Fe_{20}$ devices. The calculated damping-like spin torque conductivity is $(-1.25 \pm 0.11) \times 10^5\ \hbar/2e\ \Omega^{-1}\mathrm{m}^{-1}$, marking one of the highest reported values for van der Waals materials. These results underscore the potential of van der Waals materials for highly efficient spintronics devices.

Subsequently, we focus on the non-collinear antiferromagnet Mn_3Sn, a magnetic Weyl semimetal with great potential for efficient memory devices. We grow high-quality epitaxial $c-$plane Mn_3Sn on Al_2O_3 substrate using Ru as the seed layer. Using spin pumping-induced inverse spin Hall effect measurements on $c-$plane epitaxial $Mn_3Sn/Ni_{80}Fe_{20}$, spin-diffusion length (λ_{Mn_3Sn}), and spin Hall conductivity (σ_{SH}) of Mn_3Sn thin films are measured to be 0.42 ± 0.04 nm and $(-1.40 \pm 0.07) \times 10^5\ \hbar/2e\ \Omega^{-1}\mathrm{m}^{-1}$, respectively. While λ_{Mn_3Sn} is consistent with earlier studies, σ_{SH} is an order of magnitude higher and of the opposite sign, which is explained by the shift in the Fermi level. The growth of $a-$plane and polycrystalline Mn_3Sn is optimized, providing an opportunity to compare spin-charge conversion in different crystalline orientations. The spin Hall angle for $c-$plane and $a-$plane orientations is nearly equal but opposite signs, possibly due to different orientations of

their kagome plane. Additionally, a systematic STFMR study reveals the presence of out-of-plane polarized spin current (p_z) in Mn_3Sn/Py devices. The p_z in $c-$plane Mn_3Sn is anisotropic, while it is isotropic in $a-$plane Mn_3Sn. Unconventional damping-like and field-like torque in $a-$plane and polycrystalline Mn_3Sn are observed for the first time. These results position non-collinear antiferromagnet Mn_3Sn as a promising candidate for next-generation spintronics devices.

सारांश

यह शोध प्रबंध दो नए प्रकार की क्वांटम सामग्रियों में स्पिन-से-चार्ज रूपांतरण और स्पिन-ऑर्बिट टॉर्क्स (SOTs) का अन्वेषण करता है, जो मौलिक और अनुप्रयुक्त दोनों दृष्टिकोणों से दिलचस्प हैं। ध्यान दो महत्वपूर्ण प्रकार की सामग्रियों पर है: वैन डर वाल्स सामग्री और गैर-संरेखित प्रतिचुंबक। इन क्वांटम सामग्रियों में अद्वितीय स्पिन-निर्भर विशेषताएँ होती हैं, जिनका उपयोग उच्चतम समायोज्य स्पिन-चार्ज इंटरकन्वर्ज़न दक्षता प्राप्त करने और असामान्य स्पिन धाराओं को उत्पन्न करने के लिए किया जा सकता है। ये गुण उन्हें स्पिन-चार्ज रूपांतरण और SOT की आवश्यकता वाले अनुप्रयोगों के लिए अत्यधिक उपयुक्त बनाते हैं।

पहले, वैन डर वाल्स सामग्रियों के बड़े क्षेत्र को स्पिन-से-चार्ज रूपांतरण और SOT के लिए अन्वेषित किया जाता है। $Ni_{80}Fe_{20}$ और बड़े क्षेत्र मोनोलायर (ML) ट्रांज़िशन मेटल डायकैल्कोजेनाइड्स (TMDs) - MoS_2, $MoSe_2$, WS_2, और WSe_2 - के इंटरफेस पर स्पिन-से-चार्ज रूपांतरण का एक व्यवस्थित अध्ययन किया जाता है। इन प्रणालियों में बड़े स्पिन-से-चार्ज रूपांतरण देखे जाते हैं, जिनकी दक्षता ML TMD परत की स्पिन-ऑर्बिट कपलिंग के अनुसार स्केल होती है, जो इनवर्स राश्बा-एडलस्टीन प्रभाव के अनुरूप है। W-आधारित TMDs की दक्षता Mo-आधारित TMDs से अधिक होती है। इसके बाद, PLD तकनीक का उपयोग करके CMOS-संगत Si/SiO_2 सबस्ट्रेट पर कुछ-परत SnS की उच्च-गुणवत्ता वाली द्वि-आयामी वृद्धि का प्रदर्शन किया जाता है। SnS(1.3 nm)/ $Ni_{80}Fe_{20}$(8 nm) उपकरणों पर किए गए STFMR प्रयोग असामान्य SOT के सबूत प्रकट करते हैं। PLD का उपयोग करके Si(111) सबस्ट्रेट पर एक अन्य वैन डर वाल्स सामग्री, GeTe, के α-चरण की वृद्धि को अनुकूलित किया गया है। कोणीय-रिज़ॉल्व्ड STFMR GeTe/$Ni_{80}Fe_{20}$ उपकरणों में एक महत्वपूर्ण इन-प्लेन डंपिंग-लाइक टॉर्क को प्रकट करता है। गणना की गई डंपिंग-लाइक स्पिन टॉर्क कंडक्टिविटी (-1.25 ± 0.11) $\times 10^5$ $\hbar/2e\ \Omega^{-1}\mathrm{m}^{-1}$ है, जो वैन डर वाल्स सामग्रियों के लिए रिपोर्ट की गई उच्चतम मूल्यों में से एक है। ये परिणाम वैन डर वाल्स सामग्रियों की अत्यधिक कुशल स्पिन्ट्रॉनिक्स उपकरणों के लिए क्षमता को रेखांकित करते हैं।

इसके बाद, हम गैर-संरेखित प्रतिचुंबक Mn_3Sn पर ध्यान केंद्रित करते हैं, जो एक मैग्नेटिक वाइल सेमीमेटल है और प्रभावी मेमोरी उपकरणों के लिए बड़ी संभावनाएँ रखता है। हम Ru को सीड लेयर के रूप में उपयोग करते हुए Al_2O_3 सबस्ट्रेट पर उच्च-गुणवत्ता वाली एपिटैक्सियल *c*-प्लेन Mn_3Sn की वृद्धि करते हैं। *c*-प्लेन एपिटैक्सियल Mn_3Sn/$Ni_{80}Fe_{20}$ पर स्पिन पंपिंग-प्रेरित इनवर्स स्पिन हॉल प्रभाव माप का उपयोग करके, Mn_3Sn पतली फिल्मों की स्पिन-डिफ्यूजन लंबाई (λ_{Mn3Sn}) और स्पिन हॉल कंडक्टिविटी (σ_{SH}) 0.42 ± 0.04 nm और (-1.40 ± 0.07) $\times 10^5$ $\hbar/2e\ \Omega^{-1}\mathrm{m}^{-1}$ मापी जाती हैं। जबकि λ_{Mn3Sn} पहले के अध्ययन के अनुरूप है, σ_{SH} एक परिमाण के क्रम से अधिक है और विपरीत संकेत का है, जिसे फर्मी स्तर के परिवर्तन द्वारा समझाया गया है। *a*-प्लेन और बहुलकृष्णीय Mn_3Sn की वृद्धि को अनुकूलित किया गया है, जो विभिन्न क्रिस्टलीय उन्मुखियों में स्पिन-चार्ज रूपांतरण की तुलना

करने का अवसर प्रदान करता है। *c*-प्लेन और *a*-प्लेन उन्मुखियों के लिए स्पिन हॉल कोण लगभग समान है लेकिन विपरीत संकेत के हैं, संभवतः उनके कागोमे प्लेन की विभिन्न उन्मुखियों के कारण। इसके अतिरिक्त, एक व्यवस्थित STFMR अध्ययन Mn_3Sn/Py उपकरणों में आउट-ऑफ-प्लेन ध्रुवीकृत स्पिन करंट (p_z) की उपस्थिति को प्रकट करता है। *c*-प्लेन Mn_3Sn में p_z अनिसोट्रोपिक है, जबकि *a*-प्लेन Mn_3Sn में यह समदिश होता है। पहली बार *a*-प्लेन और बहुलकृष्णीय Mn_3Sn में असामान्य डंपिंग-लाइक और फील्ड-लाइक टॉर्क का अवलोकन किया गया है। ये परिणाम गैर-संरेखित प्रतिचुंबक Mn_3Sn को अगली पीढ़ी के स्पिन्ट्रॉनिक्स उपकरणों के लिए एक संभावित उम्मीदवार के रूप में प्रस्तुत करते हैं।

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