Growth and Investigation of Ferromagnetic and Antiferromagnetic Quantum Materials for Spintronic Devices

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Growth and Investigation of Ferromagnetic and Antiferromagnetic Quantum Materials for Spintronic Devices

by

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Department of Physics

Submitted
in fulfilment of the requirements for the degree of the Doctor of Philosophy
to the

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Certificate

This is to certify that the thesis entitled, **Growth and Investigation of Ferromagnetic and Antiferromagnetic Quantum Materials for Spintronic Devices**, being submitted by **Mr. Kacho Imtiyaz Ali Khan** to the Department of Physics, Indian Institute of Technology Delhi, for the award of the degree of **Doctor of Philosophy** is a record of bonafide work carried out by him. He has worked under our supervision and guidance and has fulfilled the requirements for the submission of this thesis, which in our 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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Kacho Imtiyaz Ali Khan

Abstract

Topological quantum materials are extremely promising for next-generation of spintronic devices such as magnetic memories due to their non-trivial band topology. Recently, kagome ferromagnet (FM) Fe_3Sn_2 and antiferromagnet (AFM) FeSn, which belong to the Fe_nSn_m-family ($m : n$ = 1:1, 3:2, 5:3) have emerged as the promising magnetic quantum materials due to their intriguing topological properties, such as the presence of Dirac nodes, Weyl points, flat bands, and topological surface states in their band structure. To date, most of the experimental and theoretical studies on Fe_3Sn_2 and FeSn have been predominantly concentrated on bulk single crystals. However, the experimental realization of the thin film of these materials with stable phase and stoichiometry is challenging. Thin films of these materials and their heterostructures are desirable for spintronics-based device applications. The topological and quantum properties can be further tuned in thin films through reduced dimensionality. In addition, thin films allow the creation of heterostructures with other materials, which may host emergent phenomena. This motivated us to investigate the growth of thin films of Fe_3Sn_2 and FeSn and their heterostructures for spintronics applications. In this thesis, we establish the growth of thin films of ferromagnetic Fe_3Sn_2 and antiferromagnetic FeSn and NiO thin films. Subsequently, we grow heterostructures of these materials with either heavy metal or ferromagnet (in the case of FeSn and NiO) to study spin pumping behavior, which is technologically important for next-generation magnetic recording devices.

In the case of Fe_3Sn_2, we first demonstrate the growth of polycrystalline ferromagnetic Fe_3Sn_2 thin films on Si/SiO_2 substrates by using a Pt seed layer. Our structural and magnetic measurements indicate that a pure ferromagnetic phase of Fe_3Sn_2 thin films with higher saturation magnetization of $M_\mathrm{S} = 464 - 640$ emu/cc is formed for the Pt seed layer, while a mixed-phase (consisting of ferromagnetic Fe_3Sn_2 and antiferromagnetic FeSn) with a lower M_S is formed for the Ta seed layer. The anomalous Hall effect measurements were performed to determine the role of the electronic bands due to the kagome structure of Fe_3Sn_2. We found a non-zero intrinsic anomalous Hall conductivity (σ_{xy}^{int}), a large value anomalous Hall coefficient (R_S), and anomalous Hall angle (θ_AHA) indicating the intrinsic origin of anomalous Hall resistivity in Fe_3Sn_2 thin films. The ferromagnetic resonance study performed in Pt/Fe_3Sn_2 based heterostructure allows us to determine intrinsic damping of Fe_3Sn_2 to be $\alpha_\mathrm{int.} = (3.8 \pm 0.2) \times 10^{-2}$. Furthermore, the spin mixing conductance of Pt/Fe_3Sn_2 is found to be $\mathrm{g}_\mathrm{eff}^{\uparrow\downarrow} = (11.7 \pm 0.6)$ nm^{-2}, which is comparable to other ferromagnet/heavy metal systems.

In the case of FeSn, we demonstrate epitaxial growth using the Pt seed layer on the sapphire substrate. The antiferromagnetic phase was confirmed using magnetization measurements as well as the presence of exchange bias in FeSn/$Ni_{80}Fe_{20}$ bilayers. A large spin mixing conductance of $g_{eff}^{\uparrow\downarrow} = (117 \pm 0.6)$ nm^{-2} is found in FeSn/Py which is nearly two-order of magnitude higher compared to the standard ferromagnet/heavy metal systems. The large value of $g_{eff}^{\uparrow\downarrow}$ is promising for application and indicates the possible role of the kagome lattice of FeSn. We have also investigated the spin pumping in the antiferromagnet NiO/Py system and we showed that the $g_{eff}^{\uparrow\downarrow}$ of an optimized polycrystalline-NiO/Py system can reach the value that for an epitaxial-NiO/Py system.

In the final part of the thesis, we numerically explore the potential application of magnetic quantum material-based heterostructures. Magnetic skyrmions are highly promising for the next generation of racetrack memory devices as they offer ultra-dense storage capacity and low power consumption in comparison to domain-wall based racetrack memory devices. In this thesis, we numerically studied the nucleation process of the FM and AFM skyrmions in racetrack devices. We found an ultrafast nucleation single/multiple AFM skyrmion using in-plane spin-polarized current in an AFM material. In addition, our method also shows low energy consumption for nucleation using in-plane spin-polarized current compared to out-of-plane spin-polarized current. We stabilize FM skyrmion for a range of external magnetic fields in ferromagnet Fe_3Sn_2 without using Dzyaloshinskii–Moriya interaction. The strong uniaxial magnetic anisotropy and dipole-dipole interaction of the kagome lattice in Fe_3Sn_2 facilitate the nucleation of both skyrmions and anti-skyrmions.

सारांश

टोपोलॉजिकल क्वांटम पदार्थ अपने गैर-तुच्छ बैंड टोपोलॉजी के कारण अगली पीढ़ी के स्पिंट्रोनिक उपकरणों जैसे चुंबकीय यादों के लिए बेहद आशाजनक हैं। हाल ही में, कागोम लौहचुंबकीय (FM) Fe_3Sn_2 और प्रतिलौहचुंबकीय (AFM) FeSn, जो Fe_mSn_n-परिवार (m : n :: 1:1, 3:2, 5:3) से संबंधित हैं, अपने पेचीदा टोपोलॉजिकल गुणों जैसे कि डिराक नोड्स, वेइल पॉइंट, फ्लैट बैंड और टोपोलॉजिकल सतह स्थितियों की उपस्थिति के कारण आशाजनक चुंबकीय क्वांटम पदार्थ के रूप में उभरे हैं। आज तक Fe_3Sn_2 और FeSn पर अधिकांश प्रयोगात्मक और सैद्धांतिक अध्ययन मुख्य रूप से बड़े एकल क्रिस्टल पर केंद्रित रहे हैं। हालाँकि, स्थिर चरण और स्टोइकोमेट्री के साथ इन पदार्थो की पतली फिल्म का प्रायोगिक कार्यान्वयन चुनौतीपूर्ण है। इन पदार्थो की पतली फिल्में और उनकी हेटरोस्ट्रक्चर स्पिंट्रोनिक्स-आधारित डिवाइस अनुप्रयोगों के लिए वांछनीय हैं। टोपोलॉजिकल और क्वांटम गुणों को कम आयामीता के माध्यम से पतली फिल्मों में और अधिक ट्यून किया जा सकता है। इसके अलावा, पतली फिल्में अन्य पदार्थो के साथ हेटरोस्ट्रक्चर के निर्माण की अनुमति देती हैं, जो अप्रत्याशित घटनाओं की मेजबानी कर सकती हैं। इसने हमें स्पिंट्रोनिक्स अनुप्रयोगों के लिए Fe_3Sn_2 और FeSn की पतली फिल्मों और उनके हेटरोस्ट्रक्चर बनाने के लिए प्रेरित किया । इस शोध पुस्तिका में, हम लौहचुंबकीय Fe_3Sn_2 और प्रतिलौहचुंबकीय FeSn और NiO पतली फिल्मों का उत्पादन स्थापित करते हैं। इसके बाद, हम स्पिन पंपिंग व्यवहार का अध्ययन करने के लिए भारी धातु या लौहचुंबकीय (FeSn और NiO के मामले में) के साथ इन पदार्थो के हेटरोस्ट्रक्चर विकसित करते हैं, जो अगली पीढ़ी के चुंबकीय रिकॉर्डिंग उपकरणों के लिए तकनीकी रूप से महत्वपूर्ण है।

Fe_3Sn_2 के मामले में, हम सबसे पहले Pt प्रारंभिक परत का उपयोग Si/SiO_2 सब्सट्रेट पर करके पॉलीक्रिस्टलाइन लौहचुंबकीय Fe_3Sn_2 पतली फिल्मों के विकास को प्रदर्शित करते हैं। हमारे संरचनात्मक और चुंबकीय माप से संकेत मिलता है कि Pt प्रारंभिक परत के लिए M_S = 464 - 640 emu/cc के उच्च संतृप्ति चुंबकीयकरण के साथ Fe_3Sn_2 पतली फिल्मों का एक शुद्ध लौहचुंबकीय चरण बनता है, जबकि Ta प्रारंभिक परत के लिए निम्न M_S के साथ एक मिश्रित चरण लौहचुंबकीय Fe_3Sn_2 और प्रतिलौहचुंबकीय FeSn से मिलकर बनता है। Fe_3Sn_2 की कागोम संरचना के कारण इलेक्ट्रॉनिक बैंड की भूमिका निर्धारित करने के लिए अनामोलस हॉल प्रभाव माप किए गए थे। हमें एक अशून्य आंतरिक अनामोलस हॉल चालकता (σ_{int}), एक बड़ा परिमाण अनामोलस हॉल गुणांक (Rs), और अनामोलस हॉल कोण (θ_{AHA}) मिला जो Fe_3Sn_2 पतली फिल्मों में असामान्य हॉल प्रतिरोधकता की आंतरिक उत्पत्ति का संकेत देता है। Pt/ Fe_3Sn_2 आधारित हेटरोस्ट्रक्चर में किया गया लौहचुंबकीय अनुनाद अध्ययन हमें Fe_3Sn_2 की आंतरिक अवमंदन को $\alpha_{int.} = (3.8\pm0.2)\times10^{-2}$ निर्धारित करने की अनुमति देता है। आगे, Pt/Fe_3Sn_2 का स्पिन मिश्रण संचालन $g_{eff}^{\uparrow\downarrow}$= (11.7 ± 0.6) nm^{-2} पाया जाता है, जो अन्य लौहचुम्बक/भारी धातु प्रणालियों से तुलनीय है।

FeSn के मामले में, हम Al_2O_3 सब्सट्रेट पर Pt प्रारंभिक परत का उपयोग करके एपिटैक्सियल फिल्मों के विकास को प्रदर्शित करते हैं। FeSn/ $Ni_{80}Fe_{20}$ बाइलेयर में चुंबकीयकरण माप के साथ-साथ विनिमय पूर्वाग्रह की उपस्थिति का उपयोग करके प्रतिलौहचुंबकीय चरण की पुष्टि की गई थी। FeSn/Py में $g_{eff}^{\uparrow\downarrow} = (117 \pm 0.6)\ nm^{-2}$ का एक बड़ा स्पिन मिश्रण चालन पाया जाता है जो मानक लौहचुंबक/भारी धातु प्रणालियों की तुलना में परिमाण के लगभग दो-क्रम अधिक है। $g_{eff}^{\uparrow\downarrow}$ का बड़ा परिमाण उपयोग के लिए और FeSn के कागोम जाली की संभावित भूमिका को इंगित करने के लिए आशाजनक है। हमने प्रतिलौहचुंबकीय NiO/Py सिस्टम में स्पिन पंपिंग की भी जांच की है और हमने दिखाया है कि $g_{ef}^{\uparrow\downarrow}$ एक अनुकूलित पॉलीक्रिस्टलाइन-NiO/Py प्रणाली उस मूल्य तक पहुंच सकती है जो एक एपिटैक्सियल NiO/Py प्रणाली के लिए है।

शोध पुस्तिका के अंतिम भाग में, हम संख्यात्मक रूप से चुंबकीय क्वांटम सामग्री-आधारित हेटरोस्ट्रक्चर के संभावित अनुप्रयोग का पता लगाते हैं। अगली पीढ़ी के रेसट्रैक मेमोरी उपकरणों के लिए चुंबकीय स्किर्मियन अत्यधिक आशाजनक हैं क्योंकि वे डोमेन-वॉल आधारित रेसट्रैक मेमोरी उपकरणों की तुलना में अल्ट्रा-सघन भंडारण क्षमता और कम बिजली की खपत प्रदान करते हैं। इस शोध पुस्तिका में, हमने रेसट्रैक उपकरणों में लौहचुंबकीय और प्रतिलौहचुंबकीय स्किर्मियन्स की न्यूक्लिएशन प्रक्रिया का संख्यात्मक रूप से अध्ययन किया। हमने प्रतिलौहचुंबकीय सामग्री में इन-प्लेन स्पिन- ध्रुवीकृत धारा का उपयोग करके एक अल्ट्राफास्ट न्यूक्लिएशन सिंगल/मल्टीपल प्रतिलौहचुंबकीय स्किर्मियन पाया। इसके अलावा, हमारी विधि आउट-ऑफ़-प्लेन स्पिन-ध्रुवीकृत धारा की तुलना में इन-प्लेन स्पिन-ध्रुवीकृत धारा का उपयोग करके न्यूक्लिएशन के लिए कम ऊर्जा खपत को भी दर्शाती है। हम Dzyaloshinskii–Moriya इंटरेक्शन का उपयोग किए बिना लौहचुंबकीय Fe_3Sn_2 में बाहरी चुंबकीय क्षेत्रों की एक श्रृंखला के लिए लौहचुंबकीय स्किर्मियन को स्थिर करते हैं। Fe_3Sn_2 में कागोम जाली की मजबूत एकअक्षीय चुंबकीय अनिसोट्रॉपी और द्विध्रुवीय-द्विध्रुवीय अंतःक्रिया स्किर्मियन और एंटी-स्किर्मियन दोनों के न्यूक्लियेशन की सुविधा प्रदान करती है।

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