FABRICATION AND INVESTIGATIONS ON THE SPUTTERD CoFeB BASED HETEROSTRUCTURES COMPRISING OF TRANSITION METAL DICHALCOGENIDES AND HEAVY METALS

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By

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Submitted

in the fulfilment of the requirement of the degree of the Doctor of Philosophy

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Dedicated to my Family

Certificate

This is to certify that the thesis entitled **"Fabrication and Investigations on the sputtered CoFeB based Heterostructures comprising of Transition Metal Dichalcogenides and Heavy Metals"**, which is being submitted by **Mr. Nanhe Kumar Gupta** 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

Conventional electronics-based memory devices that operate only through the charge attribute of the electron are not capable of possessing non-volatile memory feature. Additionally, there is a problem of performance and speed gap between the different memory stages. Spintronics-based memory devices, such as Magnetoresistive random access memories (MRAMs), utilize the spin attribute of the electron along with the charge for storing and processing the information. These MRAMs are very fast and non-volatile in nature, and hence can operate as 'Universal memory'. In these devices, the data is stored in a materials stack in which dominate role is played by ferromagnetic layer (FMs) and hence the required properties of the FMs such as high Curie temperature (T_c), low Gilbert parameter (α), high spin polarization (η), *etc.* become crucial for the fast and energy-efficient operations. There also exit a few promising MRAM structures typically formed by Ferromagnetic/high spin orbit coupled (SOC) nonmagnetic (FM/NM) heterostructures which are used for efficient generation, propagation, and manipulation of spin current for the fast and low power consumption memory operations. In such cases, the quality of the NM and the interface becomes vital.

Hence, in this thesis work, we have grown and studied thin films of Co-based FM amorphous alloy *i.e.*, $Co_{60}Fe_{20}B_{20}$ (CoFeB). The study includes structural, magnetic (static as well as dynamic) and transport properties on these grown films. Along with this, the spin dynamic properties of tw different types of heterostructures of CoFeB/NM are also explored by utilizing the spin pumping measurements. Molybdenum Mo (as NM) with different varying of its two crystalline phases is used. In addition, a Mo-based dichalcogenide (MoS_2) is also explored as potential NM in FM/NM heterostructures. First, we have optimised the growth of high quality CoFeB thin films and then focussed on the tuning in the magnetic properties of CoFeB layer with the growth parameter (specifically the Ar-pressure (P_{Ar}) during the sputtering). The ferromagnetic resonance measurements revealed that P_{Ar} has a direct impact on the inhomogeneous linewidth, anisotropy, and the damping constant (α). The low value of α ~ $(4.16 \pm 0.10) \times 10^{-3}$ is achieved in film grown at P_{Ar} = 1.1 mTorr with negligible inhomogeneity and low anisotropy. At this optimum value of P_{Ar}, the surface roughness of the films is found to be lowest (~4 Å). The study demonstrated that to achieve a desired low value of α, the parameter α can be fine-tuned by controlling the P_{Ar}. After optimizing good quality of CoFeB, we have systematically investigated the influence of the annealing temperature (T_A) on the structural and magnetic properties of the Ta/CoFeB/Ta heterostructures. The dependence of α, saturation magnetization, exchange constant, and coercive field on T_A is

thoroughly studied. Notably, these parameters show abrupt changes with T_A, which are strongly correlated with the changes in the structural properties of CoFeB. Interestingly, the α is initially found to decrease with T_A≤400℃, followed by a sharp enhancement at 415℃, at which the crystallizaiton of CoFeB gets completed. The α variation is found to be robust in the range of thicknesses from 28nm to 5nm of CoFeB. A record low value of $\alpha \sim 3.46(0.06) \times 10^{-3}$ even at high T_A (~400°C) is achieved which is so far not reported in CoFeB related studies. Occurrence of a sharp transition in electrical resistivity at 400°C also validates the onset of significant modification in the microstructure of CoFeB. The crystallite size, surface roughness, and magnetic anisotropy are all found to be highly sensitive to the choice of T_A.

Followed by successful optimization of high quality CoFeB thin films and annealing temperature (*i.e.*, 400°C), we explored the utility of CoFeB/Mo heterostructures (here Mo as an heavy metal HM) in spintronics applications. It is found that while in both the as-deposited and annealed CoFeB/Mo heterostructures, Mo crystallizes in the form of mixed phases *i.e.*, body centered cubic (*bcc*) and face centered cubic (*fcc*), the dominance, however, changes from *fcc* to *bcc* on annealing. The FMR measurements reveal that despite of relatively low spin orbit coupling of Mo, the spin pumping efficiency and magnetic properties in both the as-deposited as well as in the annealed heterostructures was comparable with those reported in similar CoFeB based heterostructures comprising of other heavy nonmagnetic HMs such as W and Ta. The spin efficiency parameters are, however, slightly better when Mo film is predominantly in the *fcc* phase as compared to the case when Mo film dominantly comprises of the *bcc* phase. Importantly, the study shows that despite the weak spin-orbit interaction, the 4*d* transition metal Mo could be a suitable choice as a non-magnetic material for spin generation and generation for the case when the thermal stability of different layers in the CoFeB based spintronic devices is of paramount concern. After exploring the HM Mo as the NM, a 2D transition dichalcogenide material (*i.e,* MoS_2) is also explored as another HM layer for its potential use in CoFeB/NM. Different numbers of layers (1-4 monolayers) of MoS_2 were grown and its effect on spin pumping in MoS_2/CoFeB heterostructures is observed. The as grown heterostructures are annealed at optimised temperature (400°C) followed by spin dynamics investigation by performing the spin pumping measurements. The spin pumping study in these heterostructures revealed 49% (51%) enhancement in damping with monolayer compared to bare CoFeB in the as-deposited (annealed) series.

Thus, this thesis work links the relation between the growth, annealing temperature and spin dynamic properties of ferromagnetic $Co_{60}Fe_{20}B_{20}$ and different $Co_{60}Fe_{20}B_{20}$/NM heterostructures (NM -: Mo & MoS_2), and provides the fundamental understanding about the interfacial effects on the spin transport phenomena in FM/NM interfaces. This will be crucial for the performance optimization in practical spintronic memory applications comprising of CoFeB based FM/NM heterostructures.

सार

पारंपरिक इलेक्ट्रॉनिक्स-आधारित मेमोरी डिवाइस जो केवल इलेक्ट्रॉन की चार्ज विशेषता के माध्यम से संचालित होते हैं, गैर-वाष्पशील मेमोरी सुविधा रखने में सक्षम नहीं हैं। इसके अतिरिक्त, विभिन्न मेमोरी चरणों के बीच प्रदर्शन और गति अंतर की समस्या भी है। स्पिंट्रोनिक्स-आधारित मेमोरी डिवाइस, जैसे मैग्नेटोरसिस्टिव रैंडम एक्सेस मेमोरी (MRAMs), जानकारी को संग्रहीत करने और संसाधित करने के लिए चार्ज के साथ-साथ इलेक्ट्रॉन की स्पिन विशेषता का उपयोग करते हैं। ये एमआरएएम प्रकृति में बहुत तेज़ और गैर-वाष्पशील हैं, और इसलिए 'यूनिवर्सल मेमोरी' के रूप में काम कर सकते हैं। इन उपकरणों में, डेटा को सामग्री के ढेर में संग्रहीत किया जाता है जिसमें प्रमुख भूमिका फेरोमैग्नेटिक परत (FMs) द्वारा निभाई जाती है और इसलिए एफएम के आवश्यक गुण जैसे उच्च क्यूरी तापमान (T_c), कम गिल्बर्ट पैरामीटर (α), उच्च स्पिन तेज़ और ऊर्जा-कुशल संचालन के लिए ध्रुवीकरण (η), आदि महत्वपूर्ण हो जाते हैं। कुछ आशाजनक एमआरएएम संरचनाएं भी हैं जो आमतौर पर फेरोमैग्नेटिक/हाई स्पिन ऑर्बिट कपल्ड (SOC) नॉनमैग्नेटिक (FM/NM) हेटरोस्ट्रक्चर द्वारा बनाई जाती हैं, जिनका उपयोग तेज और कम बिजली खपत वाले मेमोरी संचालन के लिए स्पिन करंट के कुशल उत्पादन, प्रसार और हेरफेर के लिए किया जाता है। ऐसे मामलों में, NM और इंटरफ़ेस की गुणवत्ता महत्वपूर्ण हो जाती है।

इसलिए, इस थीसिस कार्य में, हमने सह-आधारित FM अनाकार मिश्र धातु यानी, $Co_{60}Fe_{20}B_{20}$ (CoFeB) की पतली फिल्मों का विकास और अध्ययन किया है। अध्ययन में इन विकसित फिल्मों पर संरचनात्मक, चुंबकीय (स्थिर और साथ ही गतिशील) और परिवहन गुण शामिल हैं। इसके साथ ही, स्पिन पंपिंग मापों का उपयोग करके CoFeB/NM के दो अलग-अलग प्रकार के हेटरोस्ट्रक्चर के स्पिन गतिशील गुणों का भी पता लगाया जाता है। मोलिब्डेनम Mo (NM के रूप में) का उपयोग इसके दो क्रिस्टलीय चरणों के विभिन्न भिन्नता के साथ किया जाता है। इसके अलावा, FM/NM हेटरोस्ट्रक्चर में संभावित NM के रूप में एक Mo-आधारित डाइक्लोजेनाइड (MoS_2) का भी पता लगाया गया है। सबसे पहले, हमने उच्च गुणवत्ता वाली CoFeB पतली फिल्मों के विकास को अनुकूलित किया है और फिर विकास पैरामीटर (स्पटरिंग के दौरान विशेष रूप से Ar-दबाव (P_{Ar}) के साथ CoFeB परत के चुंबकीय गुणों में ट्यूनिंग पर ध्यान केंद्रित किया है। लौहचुंबकीय अनुनाद माप से पता चला कि P_{Ar} का अमानवीय लिनिविथ, अनिसोट्रॉपी और अवमंदन स्थिरांक (α) पर सीधा प्रभाव पड़ता है। $\alpha{\sim}4.16(0.10)\times10^{-3}$ का निम्न मान नगण्य अमानवीयता और कम अनिसोट्रॉपी के साथ P_{Ar} = 1.1 mTorr पर विकसित फिल्म में प्राप्त किया जाता है। P_{Ar} के इस इष्टतम मूल्य पर, फिल्मों की सतह का खुरदरापन सबसे कम (~4 Å) पाया जाता है। अध्ययन से पता चला कि α का वांछित निम्न मान प्राप्त करने के लिए, पैरामीटर α को P_{Ar} को नियंत्रित

करके ठीक किया जा सकता है। CoFeB की अच्छी गुणवत्ता को अनुकूलित करने के बाद, हमने Ta/CoFeB/Ta हेटरोस्ट्रक्चर के संरचनात्मक और चुंबकीय गुणों पर एनीलिंग तापमान (T_A) के प्रभाव की व्यवस्थित रूप से जांच की है। T_A पर α, संतृप्ति चुंबकत्व, विनिमय स्थिरांक और अवपीड़क क्षेत्र की निर्भरता का गहन अध्ययन किया गया है। विशेष रूप से, ये पैरामीटर T_A के साथ अचानक परिवर्तन दिखाते हैं, जो CoFeB के संरचनात्मक गुणों में परिवर्तन के साथ दृढ़ता से संबंधित हैं। दिलचस्प बात यह है कि शुरुआत में α $T_A \leq 400°C$ के साथ घटता हुआ पाया जाता है, इसके बाद 415°C पर तेज वृद्धि होती है, जिस पर CoFeB का क्रिस्टलीकरण पूरा हो जाता है। CoFeB की 28nm से 5nm तक की मोटाई की सीमा में α भिन्नता मजबूत पाई गई है। उच्च T_A (~400°C) पर भी $\alpha \sim 3.46(0.06) \times 10^{-3}$ का रिकॉर्ड निम्न मान प्राप्त किया जाता है, जो अब तक CoFeB से संबंधित अध्ययनों में रिपोर्ट नहीं किया गया है। 400°C पर विद्युत प्रतिरोधकता में तीव्र परिवर्तन की घटना भी CoFeB की सूक्ष्म संरचना में महत्वपूर्ण संशोधन की शुरुआत को मान्य करती है। क्रिस्टलीय आकार, सतह खुरदरापन और चुंबकीय अनिसोट्रॉपी सभी T_A की पसंद के प्रति अत्यधिक संवेदनशील पाए जाते हैं।

उच्च गुणवत्ता वाली CoFeB पतली फिल्मों और एनीलिंग तापमान (यानी, 400°C) के सफल अनुकूलन के बाद, हमने स्पिंट्रोनिक्स अनुप्रयोगों में CoFeB/Mo हेटरोस्ट्रक्चर (यहां भारी धातु HM के रूप में Mo) की उपयोगिता का पता लगाया। यह पाया गया है कि जमा और एनील्ड CoFeB/Mo हेटरोस्ट्रक्चर दोनों में, Mo मिश्रित चरणों के रूप में क्रिस्टलीकृत होता है, यानी body centered cubic (bcc) और face centered cubic (fcc), प्रभुत्व, हालांकि, fcc से बदलता है एनीलिंग पर bcc बीसीसी करने के लिए। FMR माप से पता चलता है कि Mo के अपेक्षाकृत कम स्पिन कक्षा युग्मन के बावजूद, जमा किए गए और साथ ही एनील्ड हेटरोस्ट्रक्चर दोनों में स्पिन पंपिंग दक्षता और चुंबकीय गुण अन्य भारी गैर-चुंबकीय से युक्त समान CoFeB आधारित हेटरोस्ट्रक्चर में रिपोर्ट किए गए लोगों के साथ तुलनीय थे। HM जैसे W और Ta। हालाँकि, स्पिन दक्षता पैरामीटर थोड़े बेहतर होते हैं जब मो फिल्म मुख्य रूप से fcc चरण में होती है, उस स्थिति की तुलना में जब Mo फिल्म मुख्य रूप से bcc चरण में शामिल होती है। महत्वपूर्ण रूप से, अध्ययन से पता चलता है कि कमजोर स्पिन-ऑर्बिट इंटरैक्शन के बावजूद, 4d संक्रमण धातु Mo स्पिन पीढ़ी और पीढ़ी के लिए गैर-चुंबकीय सामग्री के रूप में एक उपयुक्त विकल्प हो सकता है जब CoFeB आधारित स्पिनट्रॉनिक में विभिन्न परतों की थर्मल स्थिरता होती है। उपकरण सर्वोपरि चिंता का विषय है। NM के रूप में HM Mo की खोज के बाद, एक 2d संक्रमण डाइक्लोजेनाइड सामग्री (यानी, MoS_2) को $Co_{60}Fe_{20}B_{20}$/NM में इसके संभावित उपयोग के लिए एक अन्य HM परत के रूप में भी खोजा गया है। MoS_2 की अलग-अलग संख्या में परतें (1-4 मोनोलेयर) उगाई गईं और MoS_2/CoFeB हेटरोस्ट्रक्चर में स्पिन पंपिंग पर इसका प्रभाव देखा गया। विकसित हेटरोस्ट्रक्चर को अनुकूलित तापमान (400°C) पर

एनील्ड किया जाता है, जिसके बाद स्पिन पंपिंग माप करके स्पिन गतिशीलता जांच की जाती है। इन हेटरोस्ट्रक्चर में स्पिन पंपिंग अध्ययन से जमा (एनील्ड) श्रृंखला में नंगे $Co_{60}Fe_{20}B_{20}$ की तुलना में मोनोलेयर के साथ भिगोने में 49% (51%) वृद्धि का पता चला।

इस प्रकार, यह थीसिस कार्य फेरोमैग्नेटिक $Co_{60}Fe_{20}B_{20}$ और विभिन्न $Co_{60}Fe_{20}B_{20}$/NM हेटरोस्ट्रक्चर (NM -: Mo & MoS_2) के विकास, एनीलिंग तापमान और स्पिन गतिशील गुणों के बीच संबंध को जोड़ता है, और स्पिन परिवहन घटना पर इंटरफेशियल प्रभावों के बारे में मौलिक समझ प्रदान करता है। FM/NM इंटरफेस में। यह CoFeB आधारित FM/NM हेटरोस्ट्रक्चर वाले व्यावहारिक स्पिंट्रोनिक मेमोरी अनुप्रयोगों में प्रदर्शन अनुकूलन के लिए महत्वपूर्ण होगा।

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