GROWTH AND INVESTIGATION OF THE MAGNETIC PROPERTIES OF CO-BASED HEUSLER AND AMORPHOUS ALLOY STACKED WITH HEAVY METALS AND 2D-MATERIAL FOR SPINTRONIC APPLICATIONS

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by

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Submitted

in the fulfilment of the requirements of the degree of the Doctor of Philosophy

to the

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Dedicated to my Family

Certificate

This is to certify that the thesis entitled **"Growth and Investigation of the Magnetic Properties of Co-based Heusler and Amorphous Alloy Stacked with Heavy Metals and 2D Material for Spintronic Applications"**, which is being submitted by **Mr. Soumyarup Hait** 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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Soumyarup Hait

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Abstract

Conventional electronics-based memory devices that operate through the charge attribute of the electron suffer from volatility issues and additional problems of gap in the performance speed in different memory stages. Spintronics-based memory devices, such as Magnetoressitive random access memories (MRAMs), utilizes the spin attribute of the electron along with the charge attribute for information storage and its processing purpose. These MRAMs are non-volatile and fast, hence can work as 'Universal memory'. In these devices, the data is stored in a ferromagnet (FM) based structure 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 operation. In a few promising MRAM structures, a Ferromagnetic/nonmagnetic (FM/NM) is also utilized in which efficient generation and propagation of spin current is crucial for the energy efficient fast operation. In such cases, the quality of the NM and the interface becomes vital.

In this thesis work, we have grown and studied two Co-based FM alloys thin films, an Amorphous alloy: $Co_{60}Fe_{20}B_{20}$ (CFB) and a Full Heusler alloy: Co_2FeAl (CFA). The study includes structural, magnetic (both static and dynamic) and transport properties. Along with this, the effect of the presence of an adjacent NM layer adjacent to these FM layers on the spin dynamic properties of the resulting FM/NM heterostructure is also explored through spin pumping measurements. As NM different heavy metals (HMs) such as W, Ta, Mo, *etc.* are used. In addition, a W-based dichalcogenide (WS_2) is also explored as NM in FM/NM.

First, the crystalline quality of the CFA thin film (60 nm) is optimized to achieve low damping constant through two different thermal treatment mechanisms, *viz.*, in-situ post-deposition annealing and temperature during growth. The post-annealed films were found to be structurally superior in terms of crystallinity, density and interface width compared to the films grown at higher temperatures. A record lowest damping constant value of 0.00173 was found for as deposited CFA thin film which is further lowered to 0.00119 through post annealing at 773 K. Later, similar approach of post-deposition annealing was performed on comparatively thinner CFA thin films (10 nm), and the structural, magnetic (both static and dynamic) and transport properties were explored. The magnetic and transport properties were found to be very sensitive to the crystallinity and interface width of the samples. A lowest damping constant of 0.0051 was observed for the sample annealed at 573 K. The temperature-dependent transport analysis reveals the dominance of the weak-localization effect at lower temperatures and electron-phonon scattering at higher temperatures.

After the optimization of CFA thin films, a detailed spin transport analysis is performed on W/CFA heterostructures through Ferromagnetic resonance (FMR) base spin pumping measurements. From this analysis, an effective *spin mixing conductance* of 6.51±0.39 nm^{-2} and *transparency* of 60.27±5.41% is obtained for the W/CFA interface, while the spin diffusion length of W is found to be 2.12±0.27 nm. Later the effect of different interlayers (ILs) such as Al, Mg, Mo and Ta on the spin pumping efficiency was also explored, which revealed that appropriate IL with the appropriate thickness could enhance the spin pumping efficiency by a considerable margin. After spin pumping study in W/CFA-based heterostructures, similar measurements were carried out on Ta and CFB-based heterostructures and the spin pumping efficiency parameters are estimated. The effective *spin mixing conductance* of 11.74 ± 1.14 nm^{-2} and *transparency* of 60.61±5.88% are obtained for the Ta/CFB interface, while the spin diffusion length of Ta is found to be ~2.1 nm.

After exploring the HMs as the NM, a 2D dichalcogenide material, WS_2 is also explored. Different numbers of layers (1-4 monolayers) of WS_2 were grown and its effect on spin pumping in WS_2/CFA heterostructures is observed. The effective *spin mixing conductance* of 7.47 ± 0.97 nm^{-2} is obtained for the WS_2/CFA interface. Thus, this thesis work links the relation between the growth and spin dynamic properties of different FM and FM/NM heterostructures and also provides a fundamental understanding about the interfacial effects on the spin transport phenomena in FM/NM interfaces. This will be crucial for material optimizations in practical spintronic memory applications.

सार

पारंपरिक इलेक्ट्रॉनिक्स-आधारित मेमोरी डिवाइस जो इलेक्ट्रॉन के चार्ज एट्रिब्यूट के माध्यम से संचालित होते हैं, वे अस्थिरता के मुद्दों और विभिन्न मेमोरी चरणों में प्रदर्शन गति में अंतर की अतिरिक्त समस्याओं से ग्रस्त हैं। स्पिंट्रोनिक्स-आधारित मेमोरी डिवाइस, जैसे मैग्नेटोरेसिव रैंडम एक्सेस मेमोरी (MRAM), सूचना भंडारण और इसके प्रसंस्करण उद्देश्य के लिए चार्ज विशेषता के साथ इलेक्ट्रॉन की स्पिन विशेषता का उपयोग करता है। ये MRAM गैर-वाष्पशील और तेज हैं, इसलिए 'यूनिवर्सल मेमोरी' के रूप में काम कर सकते हैं। इन उपकरणों में, डेटा को फेरोमैग्नेट (FM) आधारित संरचना में संग्रहीत किया जाता है और इसलिए FM के आवश्यक गुण जैसे उच्च क्यूरी तापमान (T_c), निम्न गिल्बर्ट पैरामीटर (α), उच्च स्पिन ध्रुवीकरण (η), आदि बन जाते हैं। तेज और ऊर्जा कुशल संचालन के लिए महत्वपूर्ण। कुछ आशाजनक MRAM संरचनाओं में, एक फेरोमैग्नेटिक/गैर-चुंबकीय (FM/NM) का भी उपयोग किया जाता है जिसमें ऊर्जा कुशल तेज संचालन के लिए कुशल उत्पादन और स्पिन करंट का प्रसार महत्वपूर्ण है। ऐसे मामलों में, NM और इंटरफ़ेस की गुणवत्ता महत्वपूर्ण हो जाती है।

इस थीसिस कार्य में, हमने दो सह-आधारित FM मिश्र पतली फिल्मों, एक अनाकार मिश्र धातु: $Co_{60}Fe_{20}B_{20}$ (CFB) और एक पूर्ण Heusler मिश्र धातु: Co_2FeAl (CFA) का विकास और अध्ययन किया है। अध्ययन में संरचनात्मक, चुंबकीय (स्थिर और गतिशील दोनों) और परिवहन गुण शामिल हैं। इसके साथ ही, परिणामी FM/NM हेटरोस्ट्रक्चर के स्पिन गतिशील गुणों पर इन FM परतों से सटे एक आसन्न NM परत की उपस्थिति के प्रभाव का भी स्पिन पंपिंग माप के माध्यम से पता लगाया जाता है। NM के रूप में विभिन्न भारी धातुओं (HMs) जैसे W, Ta, Mo, आदि का उपयोग किया जाता है। इसके अलावा, FM/NM में NM के रूप में W-आधारित डाइक्लोजेनाइड (WS_2) का भी पता लगाया जाता है।

सबसे पहले, CFA पतली फिल्म (60 nm) की क्रिस्टलीय गुणवत्ता को दो अलग-अलग थर्मल उपचार तंत्रों के माध्यम से कम भिगोना स्थिरांक प्राप्त करने के लिए अनुकूलित किया जाता है, अर्थात, इन-सीटू पोस्ट-डिपोजिशन एनीलिंग और विकास के दौरान तापमान। एनाल्ड के बाद की फिल्मों को उच्च तापमान पर उगाई गई फिल्मों की तुलना में क्रिस्टलीयता, घनत्व और इंटरफ़ेस चौड़ाई के मामले में संरचनात्मक रूप से बेहतर पाया गया। जमा CFA पतली फिल्म के रूप में जमा CFA पतली फिल्म के लिए 0.00173 का एक रिकॉर्ड सबसे कम भिगोना स्थिर मूल्य पाया गया था, जिसे बाद में 773 K. , और संरचनात्मक, चुंबकीय (स्थिर और गतिशील दोनों) और परिवहन गुणों का पता लगाया गया। चुंबकीय और परिवहन गुण नमूनों की क्रिस्टलीयता और इंटरफ़ेस चौड़ाई के प्रति बहुत संवेदनशील पाए गए। 573 K पर एनाल्ड नमूने

के लिए 0.0051 का सबसे कम गिल्बर्ट पैरामीटर देखा गया था। तापमान पर निर्भर परिवहन विश्लेषण से कम तापमान पर कमजोर-स्थानीयकरण प्रभाव और उच्च तापमान पर इलेक्ट्रॉन-फोनन बिखरने के प्रभुत्व का पता चलता है।

CFA पतली फिल्मों के अनुकूलन के बाद, फेरोमैग्नेटिक रेजोनेंस (FMR) बेस स्पिन पंपिंग माप के माध्यम से W/CFA हेटरोस्ट्रक्चर पर एक विस्तृत स्पिन परिवहन विश्लेषण किया जाता है। इस विश्लेषण से, W/CFA इंटरफेस के लिए $6.51 \pm 0.39\ nm^{-2}$ की एक प्रभावी स्पिन मिश्रण चालन और $60.27 \pm 5.41\%$ की पारदर्शिता प्राप्त की जाती है, जबकि W की स्पिन प्रसार लंबाई 2.12 ± 0.27 nm पाई जाती है। बाद में स्पिन पंपिंग दक्षता पर विभिन्न इंटरलेयर्स (ILs) जैसे कि Al, Mg, Mo और Ta के प्रभाव का भी पता लगाया गया, जिससे पता चला कि उपयुक्त मोटाई के साथ उपयुक्त IL स्पिन पंपिंग दक्षता को काफी अंतर से बढ़ा सकता है। W/CFA - आधारित हेटरोस्ट्रक्चर में स्पिन पंपिंग अध्ययन के बाद, इसी तरह के माप Ta और CFB-आधारित हेटरोस्ट्रक्चर पर किए गए थे और स्पिन पंपिंग दक्षता मापदंडों का अनुमान लगाया गया है। $11.74 \pm 1.14\ nm^{-2}$ की प्रभावी स्पिन मिश्रण चालन और $60.61 \pm 5.88\%$ की पारदर्शिता Ta/CFB इंटरफ़ेस के लिए प्राप्त की जाती है, जबकि Ta की स्पिन प्रसार लंबाई ~ 2.1 nm पाई जाती है।

HMs को NM के रूप में तलाशने के बाद, एक 2D डाइक्लोजेनाइड सामग्री, WS_2 का भी पता लगाया जाता है। WS_2 की विभिन्न परतों (1-4 मोनोलयर्स) को उगाया गया और WS_2/CFA हेटरोस्ट्रक्चर में स्पिन पंपिंग पर इसका प्रभाव देखा गया। WS_2/CFA इंटरफेस के लिए $7.47 \pm 0.97\ nm^{-2}$ का प्रभावी स्पिन मिश्रण चालन प्राप्त किया जाता है। इस प्रकार, यह थीसिस कार्य विभिन्न FM और FM/NM हेटरोस्ट्रक्चर के विकास और स्पिन गतिशील गुणों के बीच संबंध को जोड़ता है और FM/NM इंटरफेस में स्पिन परिवहन घटना पर इंटरफेसियल प्रभावों के बारे में एक मौलिक समझ भी प्रदान करता है। व्यावहारिक स्पिंट्रोनिक मेमोरी अनुप्रयोगों में सामग्री अनुकूलन के लिए यह महत्वपूर्ण होगा।

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