



AFRL-AFOSR-VA-TR-2024-0225

Nanostructures of Magnetic Dirac Metals for RF Electronics

Budhani, Ramesh
MORGAN STATE UNIVERSITY (INC)
1700 E COLD SPRING LN
BALTIMORE, MD, 21251-0002
US

05/17/2024
Final Technical Report

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Air Force Research Laboratory
Air Force Office of Scientific Research
Arlington, Virginia 22203
Air Force Materiel Command

REPORT DOCUMENTATION PAGE

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1. REPORT DATE 20240517		2. REPORT TYPE Final		3. DATES COVERED	
				START DATE 20190301	END DATE 20221231
4. TITLE AND SUBTITLE Nanostructures of Magnetic Dirac Metals for RF Electronics					
5a. CONTRACT NUMBER		5b. GRANT NUMBER FA9550-19-1-0082		5c. PROGRAM ELEMENT NUMBER 61102F	
5d. PROJECT NUMBER		5e. TASK NUMBER		5f. WORK UNIT NUMBER	
6. AUTHOR(S) Ramesh Budhani					
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) MORGAN STATE UNIVERSITY (INC) 1700 E COLD SPRING LN BALTIMORE, MD 21251-0002 US					8. PERFORMING ORGANIZATION REPORT NUMBER
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) Air Force Office of Scientific Research 875 N. Randolph St. Room 3112 Arlington, VA 22203				10. SPONSOR/MONITOR'S ACRONYM(S) AFRL/AFOSR RTA1	11. SPONSOR/MONITOR'S REPORT NUMBER(S) AFRL-AFOSR-VA-TR-2024-0225
12. DISTRIBUTION/AVAILABILITY STATEMENT A Distribution Unlimited: PB Public Release					
13. SUPPLEMENTARY NOTES					
14. ABSTRACT The key achievements of this project are: (i) Building an ambient temperature ferromagnetic resonance FMR and inverse spin Hall effect ISHE spectrometer in the first year of the project for measurements over a frequency band of 1 to 31 GHz; (ii) Design and fabrication of a FMR spectrometer that also allows measurements of inverse spin Hall voltage over the frequency, temperature, and magnetic field ranges of 1 to 15 GHz, 2 to 300 K and 0 to ± 5 tesla respectively. This machine is based on a unique cryostat design that allow condensation of ~ 40 liters of liquid helium from the welding-grade He gas; (iii) Growth of Fe78Ga13B9, Fe60Co20B20, Fe75Co25 and Ni81Fe19 thin films with dc magnetron sputtering and their characterization using vibrating sample magnetometry (VSM) and FMR; (iv) Growth of Bi85Sb15 and Bi2Te3 films by sputter deposition from melt-grown targets, and measurement of Raman scattering, X-ray crystallography and low temperature electronic transport of these films; (v) Growth of FeGaB/Ag/BiSb, Bi2Te3/FeCo FeCoB/Bi2Te3, and b-W/FeCo heterostructures of varying thickness and interface character; (vi) Measurements of static and dynamic magnetization with VSM and FMR of the bilayers listed in the preceding sentences, together with the measurements of spin pumping across interfaces manifested in the FMR line-width and inverse spin Hall voltage; (vii) Measurements of spin textures, structural and magnetic periodicity and topological Hall effect in [?-W/FeCoGd /Ho]10 multilayers; (viii) Epitaxial growth of chiral antiferromagnet Mn3Sn and establishing the signatures of various magnetic phases in anisotropic magnetotransport in such films.					
15. SUBJECT TERMS					
16. SECURITY CLASSIFICATION OF:				17. LIMITATION OF ABSTRACT	
a. REPORT U	b. ABSTRACT U	c. THIS PAGE U	UU		18. NUMBER OF PAGES 56
19a. NAME OF RESPONSIBLE PERSON KENNETH GORETTA					19b. PHONE NUMBER (Include area code) 426-7349

Standard Form 298 (Rev.5/2020)
Prescribed by ANSI Std. Z39.18

Project Completion Report
Period covered 3/01/2019 to 03/31/2023.

Project Title:
Nanostructures of Magnetic Dirac Metals for RF Electronics

Funding Agency:
Air Force Office of Scientific Research (AFOSR)
(Award No. FA9550-19-1-0082)

Principal Investigator:
Dr. Ramesh C Budhani, Professor, Department of Physics,
Morgan State University
1700 East Cold Spring Lane, Baltimore MD -21251-0001

Govt. Program Manager:
Dr. Kenneth Goretta, AFOSR/RTA1

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ABSTRACT

The Air Force Office of Scientific Research (AFOSR) awarded a grant (# FA9550-19-1-0082) for the period 03/01/2019 to 03/31/2022 with a no-cost extension until March 31, 2023, to conduct fundamental research on the topic entitled “Nanostructures of Magnetic Dirac Metals for RF Electronics”. The Principal Investigator of this project, Dr. Ramesh C Budhani and his research team at Morgan State University gratefully acknowledge this critical support to conducted advanced research and to train underrepresented minority students for research on emergent class of materials. This *Final Report* of the project summarizes the overall achievements of the team in the areas of magnetic thin film heterostructure growth, development of novel measurement techniques, the electromagnetic response of magnetic heterostructures over a wide frequency, temperature, and magnetic field phase space, launching of research collaborations with federal laboratories and academia, and mentoring of a large number of students. The constituent materials of thin film heterostructures addressed here are soft metallic ferromagnets (FM) and non-magnetic metals/semimetals whose electronic properties are dominated by strong spin – orbit interaction (SOI) driven band topology. The ferromagnets of choice are $\text{Fe}_{78}\text{Ga}_{13}\text{B}_9$, $\text{Fe}_{60}\text{Co}_{20}\text{B}_{20}$, and $\text{Fe}_{75}\text{Co}_{25}$. While the films of the former two alloys are non-crystalline when deposited at ambient temperature, $\text{Fe}_{75}\text{Co}_{25}$ yields polycrystalline or epitaxial films, depending on the choice of the substrate and deposition temperature. The non-magnetic SOI compounds are the topological insulators of Bi_2Te_3 and Bi-Sb class, the A-15 phase of tungsten (β -W) and a chiral antiferromagnet Mn_3Sn . In addition to these systems, we have also studied thin films of compensated ferrimagnet FeCoGd multilayered with thin films of β -W and holmium to realize additive spin orbit torque on the ferrimagnet, which displays strong perpendicular magnetic anisotropy. The key achievements of this project are: (i) building of an ambient temperature ferromagnetic resonance (FMR) and inverse spin Hall effect (ISHE) spectrometer in the first year of the project for measurements over a frequency band of 1 to 31 GHz; (ii) Building of another FMR/ISHE spectrometer with measurements capabilities down to liquid helium temperatures; (ii) growth of FeGaB , FeCoB , $\text{Fe}_{75}\text{Co}_{25}$ and $\text{Ni}_{81}\text{Fe}_{19}$ thin films with dc magnetron sputtering and characterization of their magnetic properties using vibrating sample magnetometry (VSM) and FMR; (iii) Studies of superconductivity in β -tungsten (A-15 phase) films, which is an excellent spin-torque material, in collaboration with Johns Hopkins University; (iv) growth of textured thin films of $\text{Bi}_{85}\text{Sb}_{15}$ and Bi_2Te_3 and measurements of their structure, bonding configurations and electronic transport properties over a broad range of temperature and magnetic field strength; (v) synthesis of $\text{FeGaB}/\text{Ag}/\text{BiSb}$, $\text{FeCo}/\text{Bi}_2\text{Te}_3$, $\text{FeCoB}/\text{Bi}_2\text{Te}_3$ heterostructures of varying thickness and interface character; (vi) measurements of static and dynamic magnetization of such heterostructures with VSM and FMR and measurements of spin pumping across interfaces through studies of FMR driven ISHE; (vii) detailed studies of spin textures, structural and magnetic periodicity and topological Hall effect in $[\beta\text{-W}/\text{FeCoGd}/\text{Ho}]_{10}$ multilayers and (viii) epitaxial growth of chiral antiferromagnet Mn_3Sn and establishing the signatures of various magnetic phases in anisotropic magneto-transport in such films. This research has led to ten peer-reviewed articles, several conference presentations, and two invited lectures. This grant from the AFOSR has also enabled training of undergraduate and graduate students of Morgan State in the key areas of thin film growth, electronic transport measurements and studies of high frequency magnetization dynamics. Human resource development and training have been a key objective of this project. It has facilitated mentoring of one post-doctoral associate, four graduate students and four undergraduate students during the past four years. In parallel, Fruitful collaborations have been established with the RF magnetic materials group at the Air Force laboratory - Dayton Ohio, Johns Hopkins University, University of Delaware, Brookhaven National Laboratory, NIST – neutron research center and University of Maryland College park to strengthen the mandate of this project.

A. INTRODUCTION

This project was launched in the month of March 2019 to create novel thin film platforms of quantum materials (QM) and soft ferromagnets (FM) for potential application in GHz and THz spintronic devices. The ferromagnets of choice were polycrystalline and single crystal films of $\text{Fe}_{75}\text{Co}_{25}$, amorphous layers of metglass FeGaB and FeCoB, and polycrystalline films of permalloy. The quantum materials were of the Dirac semimetal family, and any other quantum metal (QM) that had strong effects of spin – orbit coupling (SOC) on its electronic structure. Hence, the focus was on a semimetal or a metal that would pump large spin current into the ferromagnetic film through the process of spin Hall effect (SHE). A yet another key objective of this proposal was to establish a ferromagnetic resonance (FMR) spectroscopy facility to measure the magnetization dynamics and inverse spin Hall effect (ISHE) induced by spin pumping in the FM/QM heterostructures over a wide frequency range. It was also aimed to augment this spectrometer design for measurements down to 2K and magnetic field strength from 0 to ± 5 tesla.

The key achievements of this project are: (i) Building an ambient temperature ferromagnetic resonance FMR and inverse spin Hall effect ISHE spectrometer in the first year of the project for measurements over a frequency band of 1 to 31 GHz; (ii) Design and fabrication of a FMR spectrometer that also allows measurements of inverse spin Hall voltage over the frequency, temperature, and magnetic field ranges of 1 to 15 GHz, 2 to 300 K and 0 to ± 5 tesla respectively. This machine is based on a unique cryostat design that allow condensation of ~ 40 liters of liquid helium from the welding-grade He gas; (iii) Growth of $\text{Fe}_{78}\text{Ga}_{13}\text{B}_9$, $\text{Fe}_{60}\text{Co}_{20}\text{B}_{20}$, $\text{Fe}_{75}\text{Co}_{25}$ and $\text{Ni}_{81}\text{Fe}_{19}$ thin films with dc magnetron sputtering and their characterization using vibrating sample magnetometry (VSM) and FMR; (iv) Growth of $\text{Bi}_{85}\text{Sb}_{15}$ and Bi_2Te_3 films by sputter deposition from melt-grown targets, and measurement of Raman scattering, X-ray crystallography and low temperature electronic transport of these films; (v) Growth of FeGaB/Ag/BiSb, $\text{Bi}_2\text{Te}_3/\text{FeCo}$ $\text{FeCoB}/\text{Bi}_2\text{Te}_3$, and b-W/FeCo heterostructures of varying thickness and interface character; (vi) Measurements of static and dynamic magnetization with VSM and FMR of the bilayers listed in the preceding sentences, together with the measurements of spin pumping across interfaces manifested in the FMR line-width and inverse spin Hall voltage; (vii) Measurements of spin textures, structural and magnetic periodicity and topological Hall effect in $[\beta\text{-W}/\text{FeCoGd}/\text{Ho}]_{10}$ multilayers; (viii) Epitaxial growth of chiral antiferromagnet Mn_3Sn and establishing the signatures of various magnetic phases in anisotropic magneto-transport in such films. This research has led to ten peer-reviewed articles, several conference presentations, and two invited lectures. This grant from the AFOSR has also enabled training of undergraduate and graduate students of Morgan State in the key areas of thin film growth, electronic transport measurements and studies of high frequency magnetization dynamics. Human resource development and training have been a key objective of this project. It has facilitated mentoring of one post-doctoral associate, four graduate students and four undergraduate students during the past four years. In parallel, Fruitful collaborations have been established with the RF magnetic materials group at the Air Force laboratory - Dayton Ohio, Johns Hopkins University, University of Delaware, Brookhaven National Laboratory and NIST – neutron research center to strengthen the mandate of this project. The following sections describe, very briefly, the achievement listed under Roman letters (i) through (viii).

B. RESULTS AND DISCUSSION

B1. Ferromagnetic resonance spectrometer for ambient temperature applications

This equipment has been fully designed and built at Morgan State with cleaver usage of existing and recycled equipment. The important components of this spectrometer are:

1. A precision signal generator (1 – 32.5 GHz, with AM and FM modulation and high-power option (from Keysight Instruments) – New).

2. 1.5 tesla electromagnet with 8-inch diameter pole pieces and associated dc power supply (refurbished equipment acquired from Bid service).
3. Lockin amplifier (SR830), Gauss meter, 20 A dc power supply, low frequency signal generator and oscilloscope (all preexisting in the laboratory).
4. Computer and LabView data acquisition system (refurbished, codes written in the laboratory).

Figure 1 shows a wide-angle view of this spectrometer. The microwave signal, pure or AM/FM modulated, is taken from the rear port of the signal generator via a semirigid copper clad cable, which is connected to a quick-connect 2.92 mm rf connector. The other end of this connector is coupled to another semirigid cable that goes to a grounded coplanar waveguide (GCPW). A sketch of the meander line waveguide used here is shown in Fig. 2(a). The meander pattern allows the required 90-degree angle

between the dc field of the electromagnet and rf field (H_{rf}) of the waveguide. The other end of this waveguide is connected to an RF diode rectifier placed well outside the gap of the electromagnet. The GCPW is mounted on a precision machined plexiglass stand that has provision for anchoring the sample face down on the GCPW, for applying gate bias and for measuring the ISHE voltage. The GCPW is placed between two Helmholtz coils that generate an ac magnetic field $H_{ac} < 25$ Oe at the maximum frequency of 700 Hz. Operation at still higher frequencies limits the field as the impedance of the circuit goes up. Fig. 2(b) shows a photograph of the Helmholtz coil assembly. The 2.92 mm quick connect on the semirigid cable allows rotation of this entire assembly between the pole pieces of the electromagnet,

thus permitting FMR measurements as a function of angle. For the measurements of the FMR driven ISHE, the sample is mounted on a tinny plexiglass block that has pins to electrically connect the two ends of the 10 mm long and 1.5 mm

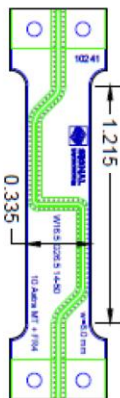


Fig. 2(a): Meander line grounded coplanar Waveguide of copper conductor



Fig. 1: Home-built broadband ferromagnetic resonance spectrometer (front view)

wide sample with gold wires. Two such ISHE holders have been made. In one of which the sample output voltage pins are connected to a thin coaxial cable that goes to a transformer preamplifier and then to a lockin amplifier. In the other holder, a well-shielded twisted pair of copper wires carry the signal to a



Fig. 2(b): Top view of the waveguide placed between Helmholtz coils and the pole pieces of a 1.5 tesla electromagnet.

10 mm long and 1.5 mm wide sample with gold wires. Two such ISHE holders have been made. In one of which the sample output voltage pins are connected to a thin coaxial cable that goes to a transformer preamplifier and then to a lockin amplifier. In the other holder, a well-shielded twisted pair of copper wires carry the signal to a

nanovoltmeter. The plexiglass block also has pins that can be used to apply a gate voltage on the back of the sample.

The output of the barrier diode detector is fed to a SR830 lockin amplifier in A-B mode and referenced to the drive frequency of the Helmholtz coil, which is driven by a ≤ 700 Hz signal taken from the reference output of the lockin and fed to a Kepco power amplifier.

While the modulated absorption appears as a derivative signal, for strong absorbers one can directly measure the absorption by AM modulation of the RF source and phase sensitive detection of the diode output. We have used this method for thicker films of FeGaB. The AM modulation of the source at ~ 5 KHz is also used for phase sensitive detection of the ISHE signal. The power supply of the 1.5 tesla electromagnet, gauss meter, lockin amplifier and signal generator are controlled with LabView data acquisition and control platform whose codes were developed by Mr. Anthony Johnson, Sr. Technician working for this project. For the measurements of ISHE voltage, direct measurements of dc voltage and RF modulation technique, both are being used.

B2. Development of cryogenic temperature (≥ 2 K) FMR and ISHE spectrometer

Figure 3(a) shows a picture of the cryogenic temperature FMR spectrometer developed during the period 3/01/2021 to 02/29/2022. The Cryo-Industries of America made the Dewar and closed cycle helium condensing system. This machine liquifies and stores ≈ 40 liters of liquid helium from welding – grade helium gas which is used to cool a 5-tesla superconducting solenoid magnet of bore diameter 2 inches, located at the bottom of the cryostat. A built-in variable temperature insert allows positioning of a coplanar waveguide (CPW) at the center of the magnet. We have developed two separate waveguide systems that allow measurements of FMR and FMR driven ISHE voltage with dc field in-plane or perpendicular to the plane of the sample. A picture of the CPW insert is also shown in the figure. The CPW has been modified to allow measurement of ISHE voltage. This modification is sketched in Fig. 3(b). The ISHE voltage is commonly measured with a nanovoltmeter. However, when the signal is very weak, we use a lockin amplifier. In this case, the CPW excitation is done in the pulsed mode and the pulse frequency serves as the reference signal for phase sensitive detection. Fig. 4(a) illustrates the high quality of ISHE data obtained at several temperatures down to 2.5 K from a 10 nm thin film of permalloy (Py) deposited on sapphire and a (111) MgO/Mn₃Sn/Py heterostructure. Fig. 4(b) shows the ISHE data of the same structures at 300 K collected at several excitation frequencies for field scans of positive and negative polarity.

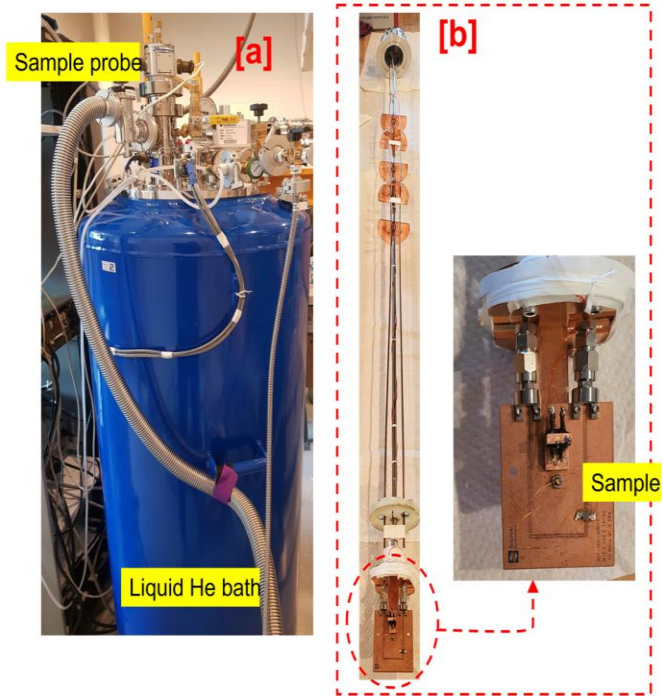


Fig. 3(a): Facility for FMR and ISHE measurements down to 2 K in field 0 to ± 5 tesla. FMR is performed in a pulse modulation mode with PSD. (b) FMR probe and waveguide.

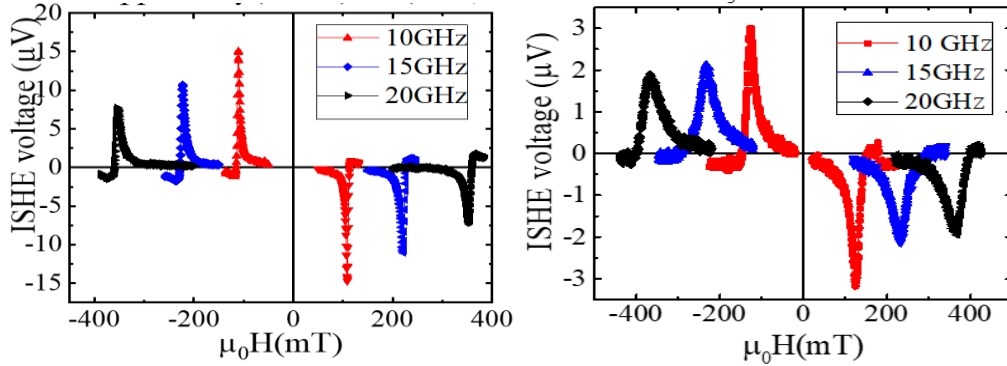


Fig. 4: (a) ISHE measurements on (0001)sapphire/Py(10 nm)/Nb(2nm) (b) on (111)MgO/Mn₃Sn(10nm)/Py(6nm)/Nb(2nm). Data taken at 20 dBm.

B3. Deposition of Dirac semimetal and strong SOC thin metal films.

B3.1: Thin films of topological insulators Bi₈₅Sb₁₅ and Bi₂Te₃:

B3.1.1 Planar Hall effect and anisotropic magnetoresistance of Bi₈₅Sb₁₅ films

Bismuth – antimony alloys (Bi_{1-x}Sb_x) are well-known thermoelectric (TE) materials [1,2]. Their TE characteristics emanate from a tunable electronic band structure achieved by adjusting the Bi/Sb ratio in the alloy. This material has attracted much attention in recent years on the recognition of a strong spin – orbit interaction (SOI) driven band crossing in the composition range of $0.03 < x < 0.22$ [3,4]. For $x \approx 0.03$, it acquires a Dirac-like metallic state, which changes to a 3D Weyl semimetal on application of a magnetic field [5]. For $0.09 < x < 0.22$, (Bi_{1-x}Sb_x) is a 3D topological insulator, as established by angle resolved photoemission measurements on single crystals [6] and epitaxial thin films [7]. Electronic transport measurements on such crystals are characterized by a metal-like resistivity at low temperatures and the presence of weak Shubnikov de Haas oscillations in the magnetic field dependence of longitudinal (r_{xx}) and Hall (r_{xy}) resistivity [8]. These features of electronic transport have been attributed to spin-momentum locked surface states. The topological phase of (Bi_{1-x}Sb_x) has been identified as an excellent spin-orbit-torque (SOT) material for spintronic applications [9-11]. In bilayers of BiSb and a ferromagnet like MnGa, the spin – momentum locked surface states of the former pump a large spin current into the FM layer under the action of a charge current driven Rashba – Edelstein effect (REE) [12]. The REE torque on the magnetization of the FM layer has been established to be much larger than the spin Hall effect driven torque of a heavy metal like Pt. This property, combined with compositional flexibility, has made Bi_{1-x}Sb_x a promising spintronic material. However, the requirement of epitaxy is a bottleneck for scalable and CMOS compatible applications of BiSb as a SOT layer.

Interestingly, two recent studies [11,13] have indicated that the polycrystalline films of Bi_{1-x}Sb_x made by a scalable process like sputtering are quite effective in producing spin currents to torque the magnetization of FeMn, FeCoB and CoTb thin films. This observation has motivated us to undertake a detailed study of electronic transport in sputter-deposited polycrystalline films of BiSb. Although polycrystalline films of (Bi_{1-x}Sb_x) alloys have been studied previously by several groups [14-16], the focus of these studies has been their applicability as a thermoelectric material. Here we compare the low temperature ($T \geq 2$ K) magneto-transport in polycrystalline films of Bi₈₅Sb₁₅ with that of epitaxial films and single crystals where the existence of a topological phase has been established.

$\text{Bi}_{85}\text{Sb}_{15}$ films were deposited on thermally oxidized silicon wafers by magnetron sputtering of a stoichiometric 2 – inch diameter alloy target in a laser deposition/sputtering hybrid load-lock chamber shown in Fig. 5. The $\text{Bi}_{85}\text{Sb}_{15}$ alloy has a low melting temperature ($T_m \sim 300^\circ\text{C}$) [17] and high sputtering yield [18]. The sputter gun was operated at low power (≈ 25 Watts) to avoid surface melting of the target and to ensure a low growth rate (~ 0.15 nm/s). Films were deposited at ambient temperature and at 100 and 150°C . It was noticed that the higher deposition temperature and excessive growth rates result in rough films.

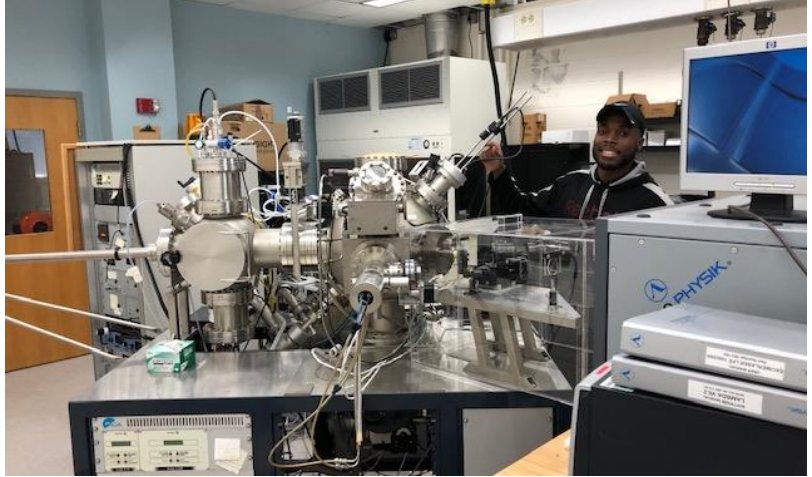


Fig. 5: Integrated High vacuum system (base pressure $\approx 1 \times 10^{-7}$ mbar) used for PLD and sputter deposition of FeGaB/BiSb/MgO type heterostructures.

The X-ray diffraction profile of the film deposited at 35°C shows peaks corresponding to several allowed hkl -indices with (001) reflections being the most prominent. The higher intensity of such reflections suggests a predominantly oriented growth along the c -axis of the rhombohedral cell (See Fig. 6). Films deposited at higher temperatures show a much-improved c -axis texture albeit with enhanced surface roughness. Since the smoothness of the interface between BiSb and FM films is important to address SOT physics, we have focused on addressing the properties of films grown in the vicinity of ambient temperature ($T \leq 50^\circ\text{C}$).

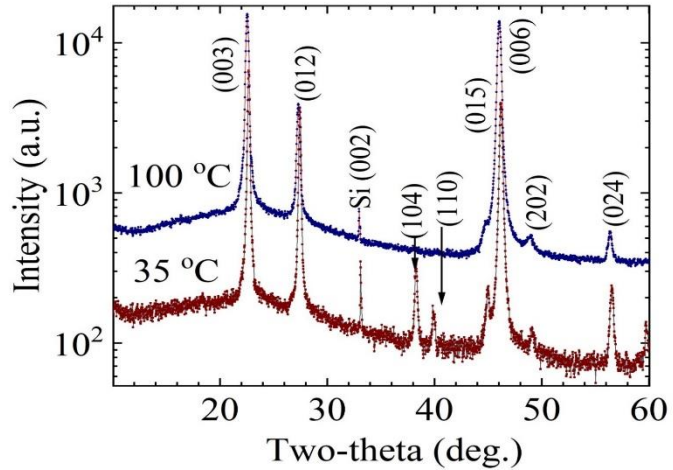


Fig. 6: X-ray diffraction profile of $\text{Bi}_{85}\text{Sb}_{15}$ thin film deposited on thermally oxidized silicon at 35°C and 100°C . The diffraction peaks have been indexed for the Miller indices of the rhombohedral structure of BiSb alloy. A preferred orientation of the film along the c -axis is evident in these data.

The temperature dependence of electrical resistivity and Hall coefficient of these films was reported in the Year – I progress report. While there is a lack of carrier concentration and mobility data on epitaxial and polycrystalline films of $\text{Bi}_{85}\text{Sb}_{15}$, the carrier concentration in our films (1.97×10^{19} electrons/ cm^3) is $\approx$ two orders of magnitude higher than that reported by Taskin and Ando [8] for $x = 0.09$ BiSb single crystals and the carrier mobility of these films ($\sim 203 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$) is smaller by the same factor.

The measurements of electrical resistivity of 3D topological insulators and Dirac/Weyl semimetals in a configuration where the current density, magnetic field and induced electric field are in the same plane have generated considerable interest due to the presence of anisotropic magnetoresistance and planar Hall

effect [19-26], which have traditionally been the signatures of electronic transport in a magnetically ordered metal [27]. We carried out the measurements of electrical resistivity of BiSb thin film in a field-in-plane geometry as shown in Fig. 7. Here the transport current J_x ($= 25 \text{ A/cm}^2$) flows in the x-direction and the induced electric fields E_{xx} and E_{xy} in the x and y directions respectively are measured as the magnetic field is rotated in the xy-plane from -90 to 270 degree. The angle $\phi = 0$ corresponds to the situation when H and J_x are parallel.

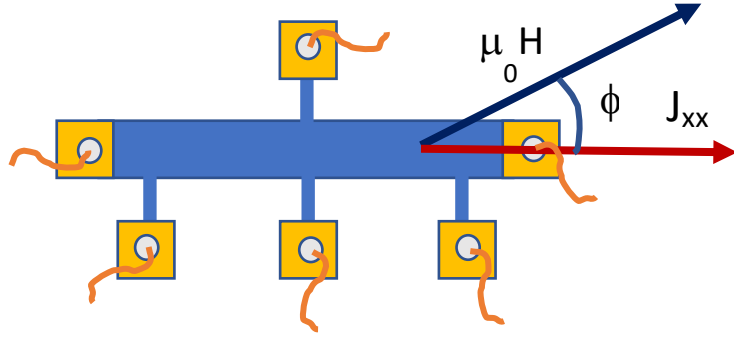


Fig. 7: Sketch of sample geometry used for measurements of the angular dependence of R_{xx} and R_{xy} . The magnetic field is rotated in the plane of the film from 0 to 2π with respect to the direction of the current density.

The induced electric field in the direction of J_x yields magnetoresistivity whereas the orthogonal field E_{xy} results in the Planar Hall effect. The components of the resistivity tensor are expressed as [27]

$$\rho_{xx} = \rho_{\perp} - \Delta\rho \cos^2 \phi \quad (1)$$

$$\rho^{\text{PHE}} = -\Delta\rho \sin \phi \cos \phi \quad (2),$$

where $\Delta\rho = \rho_{\perp} - \rho_{\parallel}$, with $\rho_{\perp}$ and $\rho_{\parallel}$ corresponding to H perpendicular to J_x and H parallel to J_x respectively. The anisotropic magnetoresistance ($\Delta\rho$) and ρ^{PHE} are characteristic features of spin-orbit scattering dominated electronic transport in magnetic alloys due to the coexisting s and d bands near the Fermi energy [27]. Interestingly however, PHE and anisotropic magnetoresistance have been found in non-magnetic 3D topological insulators like Bi_2Te_3 [19-22]. Appreciable values of AMR and PHE have been observed in Dirac and Weyl semimetals as well [23-26]. While in the latter class of semimetals, this anisotropic transport has been attributed to breaking of chiral symmetry, which results in a large negative longitudinal magnetoresistance (LMR), varied interpretations have been proposed for PHE in 3D TIs where no negative LMR is seen. $\text{Bi}_{1-x}\text{Sb}_x$ is one of the first reported 3D TIs. While the measurements of Hall resistivity and orbital MR in single crystals and epitaxial films of $\text{Bi}_{1-x}\text{Sb}_x$ have been reported earlier [6,8,10], data on PHE and AMR is lacking. In the following sections, we present results of in-plane anisotropic electronic transport from 2 K to 270 K in a field range of 0 to ± 9 tesla. Figure 8(a) shows the variation of R_{xx} and R_{xy} at 2 K and $+9$ tesla as the sample is rotated to change the angle between magnetic field and current density directions from -90 degree to 270 degree. A similar measurement of R_{xx} and R_{xy} at 2 K with the field direction reversed is shown in Fig. 8(b). We note that the positions of extrema in R_{xx} and R_{xy} data of Fig. 8(a&b) are consistent with Eq. 1 & 2. However, there is a noticeable asymmetry in the behavior of R_{xy} for the two field orientations. There are two factors that contribute to this asymmetry. First is a normal Hall voltage that results from a non-zero out-of-plane component of the magnetic field due to a misalignment of the film plane and the plane of rotation. This contribution is antisymmetric in field and can be eliminated on symmetrization of the $(+H)$ and $(-H)$ data. Second, a zero-field misalignment voltage across the Hall contacts, which will add to the PHE voltage on symmetrization of R_{xy} [$\{(R_{xy}(+H) + R_{xy}(-H))/2\}$]. This constant resistance can be subtracted from the measured $R_{xy}(H)$ provided its value is small such that the AMR induced change in it is insignificant compared to the true $R_{xy}(H)$. In addition to these two factors, another contaminant of R_{xy} comes from the orbital magnetoresistance (OMR) of the misaligned section of

the transverse contacts when the sample is tilted with respect to the plane of rotation. This would add a $\cos^2\phi$ term in Eq. 2. The misalignment of the plane of rotation and the plane of the sample adds an error in the value of AMR as well. If the OMR of the sample is large, then the normal component of the field will add an OMR contribution to $R_{xx}^\perp$ and R_{xx}' . We address these errors by first symmetrizing the $R_{xx}(\phi)$ and $R_{xy}(\phi)$ data of Fig. 8(a) and Fig. 4(b). The result of this procedure is displayed in Fig. 8(c), along with the fits of $R_{xx}(\phi)$ to Eq. 1 and of $R_{xy}(\phi)$ to a function of the type $R_{xy}(\phi) = A + B \sin(\phi) \cos(\phi) + C \cos^2(\phi)$. The last term of this equation considers the error in R_{xy} due to OMR of the misaligned section, as discussed earlier. The magnitude of this error ($C \cos^2(\phi)$) and the antisymmetric contribution to R_{xy} are displayed in

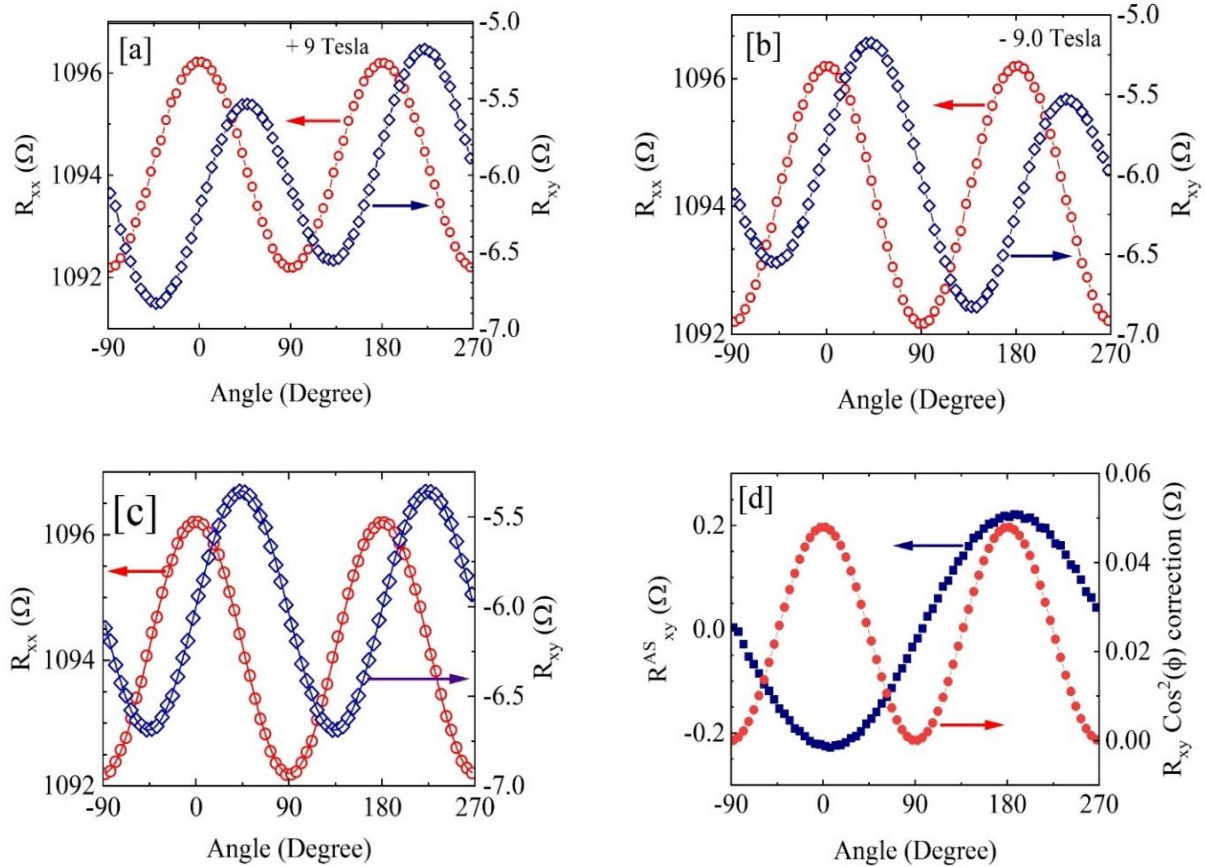


Fig. 8: The variation of R_{xx} and R_{xy} as a function of the angle (ϕ) between the direction of a 9-tesla field and the direction of current density J_{xx} . (a) + 9 tesla, (b) - 9 tesla, (c) R_{xx} and R_{xy} on symmetrization of the data shown in (a) and (b). Solid lines in Fig.4(c) are fits to equation of the type $R_{xx}(\phi) = R_{xx}(0) - D R_{xx} \cos^2(\phi)$ and $R_{xy}(\phi) = R_{xy}(0) - D R_{xy} \sin(\phi) \cos(\phi) + B \cos^2(\phi)$ to the R_{xx} and R_{xy} data respectively. Fig. 4(d) shows the variation of antisymmetric R_{xy} extracted from Fig. 4(a) and Fig.4(b) and the variation of coefficient B extracted from the fit in Fig. 4(c).

Fig. 8(d). Comparing the peak amplitude of R_{xy} in Fig. 8(c) and the peak amplitude of the $C \cos^2(\phi)$ term in Fig. 8(d) shows that the maximum error introduced by this term in the measurement of R_{xy} is $\leq \pm 3.5\%$. Moreover, a comparison of the peak value of antisymmetric R_{xy} ($= 0.2 \Omega$) at 9 tesla in Fig. 8(d) with the Hall resistance data linearized in field (slope $= 2\Omega/\text{tesla}$), yields a tilt angle of ≈ 0.6 degree with respect to the plane of the sample.

The angular dependencies of the symmetrized R_{xy} and R_{xx} at 2 K for several values of field are shown in Fig. 9(a) and Fig. 9(b) respectively. In the plot of Fig. 9(a), the constant offset value of ΔR_{xy} at zero-field

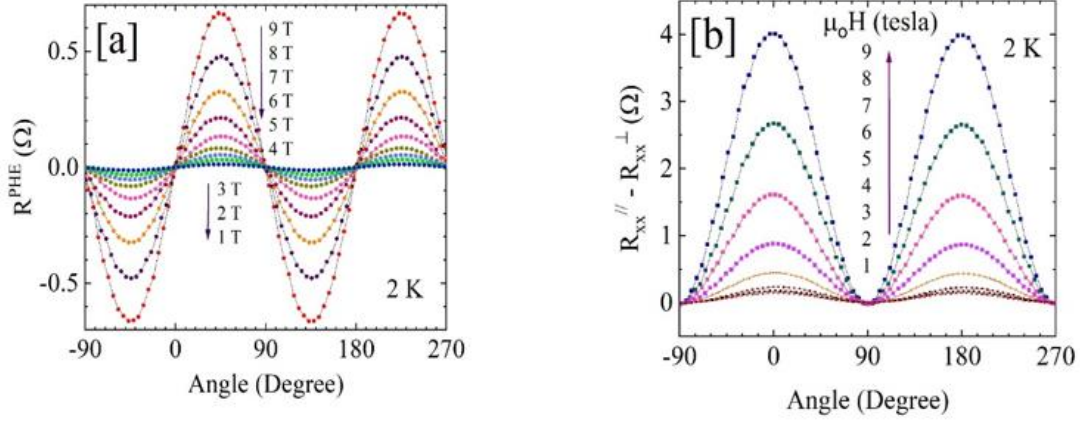


Fig. 9: (a) shows the planar Hall resistance as a function of angle. The difference between $R^//_{xx}$ (f) - $R^{\perp}_{xx}$ is plotted as a function of the angle (f).

has been subtracted, whereas Fig. 9(b) displays the variation of $R_{xx}(\phi)$ with respect to the average value of R_{xx} at $\phi = \pm 90$ degree. The dominant $\sin(\phi)\cos(\phi)$ and $\cos^2(\phi)$ dependencies of these quantities are evident in Fig. 9(a) and 9(b) respectively. The variation of the peak amplitudes of ΔR^{PHE} and ΔR_{xx} ($R^//_{xx} - R^{\perp}_{xx}$) extracted from these plots as a function of magnetic field is shown in Fig. 10. The two important conclusions that can be drawn from these data are: (1) the longitudinal magnetoresistance is positive with a field dependence of the type $\Delta R_{xx} \sim H^{\alpha}$ ($\alpha = 1.5$), and (2) the PHE amplitude is smaller than the ΔR_{xx} .

In the inset of Fig.10 we plot the variation of orbital magnetoresistance ($\Delta R^{OMR} = [\{R_{xx}(H) - R_{xx}(0)\}/R_{xx}(0)] \times 100$) with field normal to the film plane, longitudinal magnetoresistance (LMR) ($\Delta R^{LMR} = [\{R^//_{xx}(H) - R_{xx}(0)\}/R_{xx}(0)] \times 100$) with field parallel (//) to J_x and anisotropic magnetoresistance ($\Delta R^{AMR} = [\{R^//_{xx}(H) - R^{\perp}_{xx}(H)\}/R^{\perp}_{xx}(H)] \times 100$). Two noteworthy conclusions that can be drawn from these data are: (1) the LMR is larger than OMR, and (2) the AMR is small but positive, indicating the absence of any chiral anomaly.

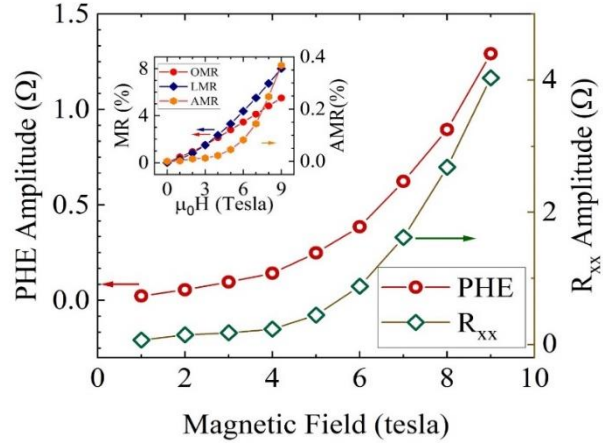


Fig. 10: Amplitudes of $\Delta R^{PHE}(H)$ and $\Delta R_{xx}(H)$ at 2 K plotted as a function of magnetic field. Inset show the variation of out-of-plane and in-plane magnetoresistance as a function of applied field.

The resistance tensor of a two-dimensional magnetic film for in-plane magnetic field predicts that the amplitudes of ΔR^{PHE} and ΔR_{xx} should be the same. The large difference seen in the values of ΔR^{PHE} and ΔR_{xx} in this case points towards some subtle differences in the origin of these components in the resistance tensor. One might argue that in systems with large orbital magnetoresistance, the out of film plane magnetic field due to sample misalignment may significantly change the value of ΔR_{xx} . An estimation of this error can be made by looking at the results of the orbital MR measurements shown in Fig. 10 and the estimated tilt of 0.6 degree from the antisymmetric R_{xy} in Fig. 8(d). This much tilt at 9 tesla would result in a ≈ 95 mT perpendicular field. From Fig. 10 we conclude the effect of this misalignment of field on the measurement of ΔR_{xx} is negligible.

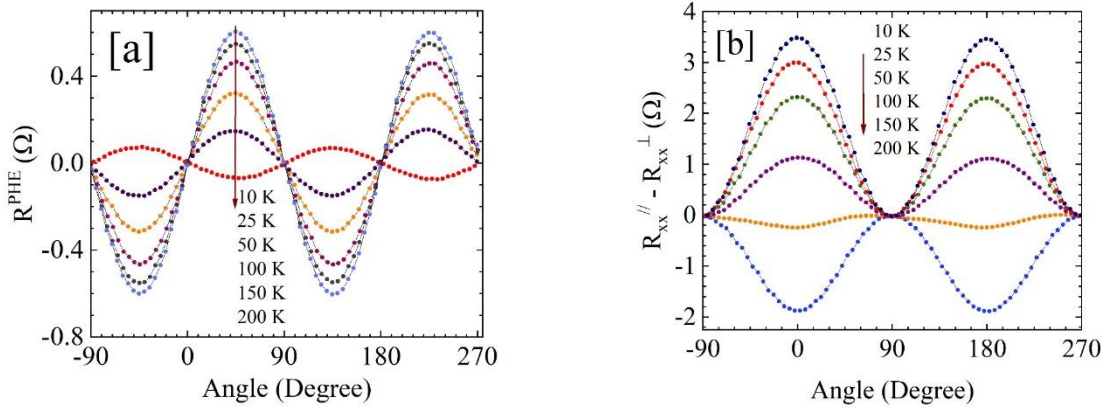


Fig. 11: Angle (θ) dependence of the ΔR^{PHE} and ΔR_{xx} at several temperatures from 2 K to 160 K are shown in (a) and (b) respectively. The data have been obtained by symmetrization of the plus and minus field responses.

Fig. 11(a) and Fig. 11(b) respectively show the angular dependence of ΔR^{PHE} and ΔR_{xx} at several temperatures in the range of 10 K to 160 K. A significant drop in the amplitude of PHE and AMR are seen on increasing the temperature, followed by a sign change in the temperature window of 150 K to 200 K. The amplitude of ΔR^{PHE} and ΔR_{xx} are plotted in Fig. 12. The change in the sign of these two quantities appears to correlate with the inflection point in the temperature dependence of R_{xx} , as seen in the inset where we have plotted dR_{xx}/dT vs. T . The noteworthy features of the in-plane magnetoresistance of these $\text{Bi}_{85}\text{Sb}_{15}$ thin films are: (1) observation of a planar Hall effect, (2) a large difference in the amplitudes of ΔR^{PHE} and ΔR_{xx} , and (3) a sign reversal of these two quantities in the

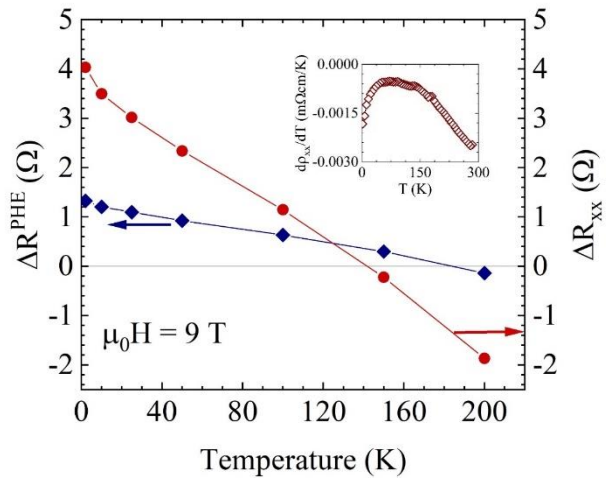


Fig. 12: Variation of the PHE amplitude ΔR^{PHE} and ΔR_{xx} amplitude as a function of temperature. Inset of the figure shows dR/dT vs T in zero-field.

vicinity of 150 K. Although BiSb is non-magnetic, these first observations of PHE in BiSb is consistent with the recent reports of PHE in non-magnetic semimetals like $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3$ [20], bismuth [28] and MoTe_2 [23,24] with non-trivial band topology. The observation of PHE in $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3$ has been attributed to scattering by magnetic impurities present in the sample [20]. The anisotropy of the Fermi surface and the resulting large difference in the magnetic field dependence of $R_{xx}^\perp$ and $R_{xx}^\parallel$ has also been argued to be the source of PHE in some systems [24,28]. We have analyzed the field dependence of $R_{xx}^\perp$ and $R_{xx}^\parallel$ at 2 K. The resistance rises as $\sim H^\alpha$, with α as 1.46 and 1.51 for the $\perp$ and $\parallel$ measurements, respectively. A treatment of electron transport in the framework of semiclassical Boltzmann transport equation attributes PHE in topological insulators to orbital magnetism of Bloch electrons, which is non-zero because of the symmetry breaking in-plane magnetic field [22,25]. This model, however, does not predict a difference in the value of ΔR^{PHE} and ΔR_{xx} . The experimental data of Taskin et al. [20] on $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3$ crystal shows a large difference in these quantities, which the authors have attributed to a contamination of the signal by an OMR contribution arising from the misalignment of the sample plane and the plane of rotation. However, this scenario does not apply in the present case as we have shown that the large difference in ΔR^{PHE} and ΔR_{xx} cannot be attributed to a tilt of the sample plane. Similarly, a change in the sign of planar Hall and anisotropic magnetoresistance at higher temperatures is difficult to explain based on misalignment and/or impurity scattering. A plausible description of this difference in the amplitudes of ΔR^{PHE} and ΔR_{xx} as well as of the sign change has been given by Zheng et al. [26] where they consider the tilt of Dirac cones in the TI induced by the in-plane magnetic field. This tilt contributes to anisotropic backscattering, which is enhanced further by impurity resonant states and may lead to a sign change in AMR. The change in the sign of AMR of our BiSb films is consistent with this picture.

B3.2 Structure and electronic transport in sputter – deposited Bi_2Te_3 films.

High quality polycrystalline films of Bi_2Te_3 were deposited by dc magnetron sputtering on $5 \times 5 \text{ mm}^2$ c-plane (0001) Al_2O_3 substrates in an ultrahigh vacuum system (Fig. 13). A two-inch diameter melt-cast 99.999 % pure target of Bi_2Te_3 was sputtered at 5 mTorr pressure of research grade Ar gas. The substrates were ultrasonically degreased in acetone and alcohol prior to loading in the deposition chamber. Three different types of 25 nm thick Bi_2Te_3 films were deposited at a growth rate of $\approx 2.79 \text{ \AA/s}$ with a choice of growth temperature and nitrogen concentration in the sputtering plasma. The films deposited at 150 °C in pure Ar are labeled as S_1 , whereas those deposited at ambient temperature are named S_2 . Lastly, the films deposited at 150 °C in a mixture of Ar and N_2 (31% N_2 and 69% Ar) are denoted as S_3 .

The measurements of PHE, AMR, OMR and Hall resistivity were performed on films deposited through a shadow mask that created Hall bars of dimensions of $300 \times 3000 \text{ }\mu\text{m}^2$. A physical properties measurement system equipped with a vertical rotator was used to measure electronic conduction in the temperature and field ranges of 2 - 300 K and 0 to $\pm 9 \text{ T}$,



Fig. 13: Multitarget sputtering system with load-lock used for deposition of FeCo , $\beta\text{-W}$ and Bi_2Te_3 films (base pressure $\approx 8 \times 10^{-9} \text{ mbar}$).

respectively. The crystallographic structure of the films was determined using X-ray diffraction measurements performed using a Bruker D-8 Advance X-ray diffractometer with Cu-K α_1 radiation and an Eiger2R 500K detector. A parallel X-ray beam was obtained using a Göbel mirror and Cu-K α_1 radiation was selected using a Ge (004) monochromator. Raman scattering spectra were measured using a Horiba JY T64000 spectrometer equipped with Olympus microscope and liquid nitrogen cooled CCD-detector. The laser probe was 2 microns in diameter. Spectra were excited with the 514.5 nm line of Ar-Kr laser using power of 0.12 mW. Spectral resolution of 2 cm $^{-1}$ was used.

Bi $_2$ Te $_3$ crystallizes in a tetradymite type structure with space group R-3m, which is formed by stacking three

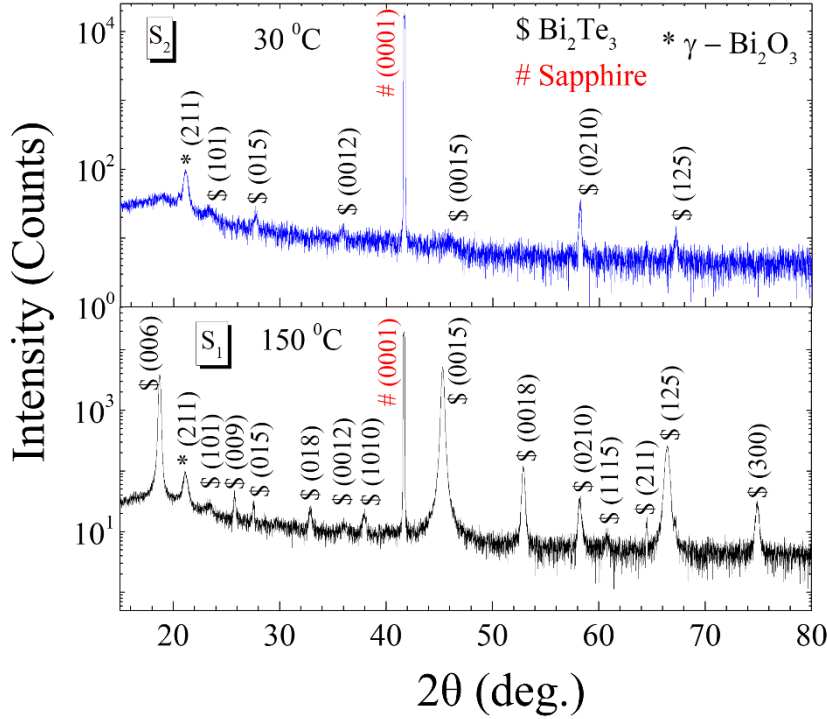


Figure 14: X-ray diffraction profiles of Bi $_2$ Te $_3$ thin films deposited on (0001) sapphire at 30 °C (upper panel) and 150 °C (lower panel). The diffraction peaks have been indexed for the Miller indices of the rhombohedral phase of Bi $_2$ Te $_3$. A marked enhancement in the intensity of diffraction peaks is seen in the film grown at 150 °C.

quintuple layers (QLs), each containing five planes of Bi and Te atoms along the c direction with a c-axis lattice parameter of 30.474 Å [29, 30]. The a-axis lattice parameter of this hexagonal structure is 4.3 Å, which is 7 % larger than the a-axis lattice parameter of sapphire. Notwithstanding this large lattice mismatch, twinning-free films of Bi $_2$ Te $_3$ with low surface roughness have been deposited earlier on (0001) sapphire [31, 32]. Fig.14 compares the θ - 2θ X-ray diffraction profiles of our films grown at room temperature and 150 °C. The diffraction pattern of the film deposited at 150 °C displays a prominent (000n) texture. We have also observed a weak intensity peak of γ -Bi $_2$ O $_3$, which is presumably formed due to the interaction of surface adsorbed oxygen with Bi on prolonged exposure of the film surface to ambient atmosphere [33,34]. The highly c-axis textured pattern of the polycrystalline films has been indexed for the Miller indices of the rhombohedral space group R-3m. The diffraction profile of the film deposited at room temperature shows broader peaks of lesser intensity, indicating their low crystallinity. The average crystallite size of the 150 °C film (S $_1$) calculated using the Debye - Scherrer formula is $\sim$ 7.5 Å and its c-axis lattice parameter is $\sim$ 29.3 Å. The lattice parameter of the films grown at room temperature is 3 % smaller and their crystallite size is $\sim$ 7.1 Å.

Bi_2Te_3 films of 100 nm in thickness grown in the same conditions were examined by Raman scattering spectroscopy, which is routinely used for characterization of Bi_2Te_3 bulk and films. Raman spectra of the films presented in Fig. 15 show phonon lines in the spectral range typical for Bi_2Te_3 . The bands are broadened and are shifted to lower frequencies as compared to the Raman spectra of bulk Bi_2Te_3 . For comparison, the position of the modes of Bi_2Te_3 are marked on the plot with the dashed lines and symmetry notations. Typically, the presence of the A_{1u} mode in the spectra of Bi_2Te_3 signals broken local inversion symmetry due to structural disorder. The A_{1g} mode signifies the symmetric out of plane stretching of Bi-Te bonds in the opposite direction while E_g mode arises due to the symmetric in plane stretching of Te-Bi atoms in the same direction [35]. The broad Raman bands of the films at 96 cm^{-1} , 108 cm^{-1} , and 123 cm^{-1} indicate the presence of structural disorder and smaller crystallite size in the samples which results in the decrease of vibrational frequencies compared to the bulk Bi_2Te_3 [36]. Broader bands of the film grown at 30 °C suggest a higher amount of structural disorder.

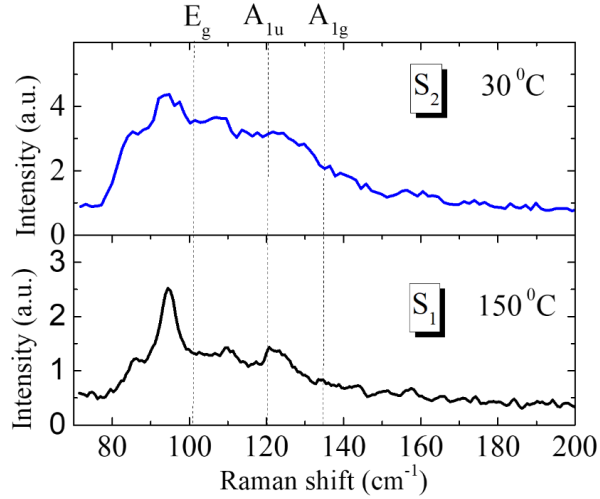


Figure 15: Raman scattering spectra of Bi_2Te_3 100 nm films deposited on (0001) sapphire at 30 °C (upper panel) and 150 °C (lower panel). Phonons of Bi_2Te_3 are marked.

Electronic structure calculations for rhombohedral Bi_2Te_3 yield a bandgap of ~ 0.15 eV [37]. While this gap exists in the bulk of the material, the conduction and valence bands cross near the Fermi energy at a single Dirac point, making Bi_2Te_3 a topological insulator. These Dirac-like states are known as topological surface states (TSS) [37], which remain robust when the film thickness is reduced to 3QL. However, below this thickness the TSS cease to exist and a bandgap opens [37]. Defects, such as Te vacancies, can dope Bi_2Te_3 , moving the Fermi energy, sometimes into bands. To ascertain the number density of charge carriers, their type, and mobility, we have performed measurements of Hall resistivity (ρ_{xy}) of the S_1 , S_2 and S_3 type of samples over a wide range of temperature and magnetic field. Panels *a*, *b* and *c* respectively of Fig. 16 show the plots of ρ_{xy} as a function of magnetic field for the samples S_1 , S_2 and S_3 at several values of temperature. The sign of the ρ_{xy} for all three samples is negative, and it is linear in field over the field range of 0 to 9 tesla. This behavior of ρ_{xy} is indicative of a single band transport with electrons as majority charge carriers. We calculated the carrier density (n_s) from the slopes of the ρ_{xy} vs field curves. The results of such calculations are displayed in Panel d of Fig. 16. The large value of n_s ($\sim 9 \times 10^{20}/\text{cm}^3$) at 2 K for the film deposited at 150 °C (S_1) is a noticeable feature. We also note that the n_s of this sample remains nearly the same on increasing the temperature to 300 K. However, the n_s of the ambient temperature deposited film (S_2) increases by $\sim 20\%$ on raising the temperature from 2 K to 300 K. A similar increase is also seen for the sample prepared in $\text{N}_2 + \text{Ar}$ mixture (S_3). The inset of the panels '*a*' through '*c*' show the temperature dependence of the longitudinal resistivity (ρ_{xx}) of the films measured in zero field and at 1 tesla. The ρ_{xx} of sample S_1 is distinctly metallic down to 2 K. The resistivity increases by a factor of ~ 3 when the growth is performed at ambient temperature. The ρ_{xx} of this sample (S_2) show a distinct increase as the temperature is lowered below ~ 20 K. A similar trend in $\rho_{xx}(T)$ is also seen for the sample S_3 . The Hall mobility (μ_H) of the samples has been calculated using the Drude's relation $\mu_H = 1/en_s\rho_{xx}$. The mobility of the 150 °C deposited film is 16.7 $\text{cm}^2 \text{V}^{-1} \text{S}^{-1}$ at 2 K. These transport parameters are comparable to the results of other groups on sputter-deposited films of Bi_2Te_3 [38, 39]. For example, Kim et. al. [38] report a Hall mobility

of $20 \text{ cm}^2 \text{ V}^{-1} \text{ S}^{-1}$ for their 150°C deposited film along with n_s and ρ_{xx} of $2.5 \times 10^{20} \text{ cm}^{-3}$ & $12 \text{ } \mu\Omega \text{ m}$ respectively. We further note that the n_s and μ_H of the film deposited at 150°C in a $\text{Ar}+\text{N}_2$ gas mixture are nearly the same as for the film deposited at room temperature. The ρ_{xx} of these films also shows an upturn

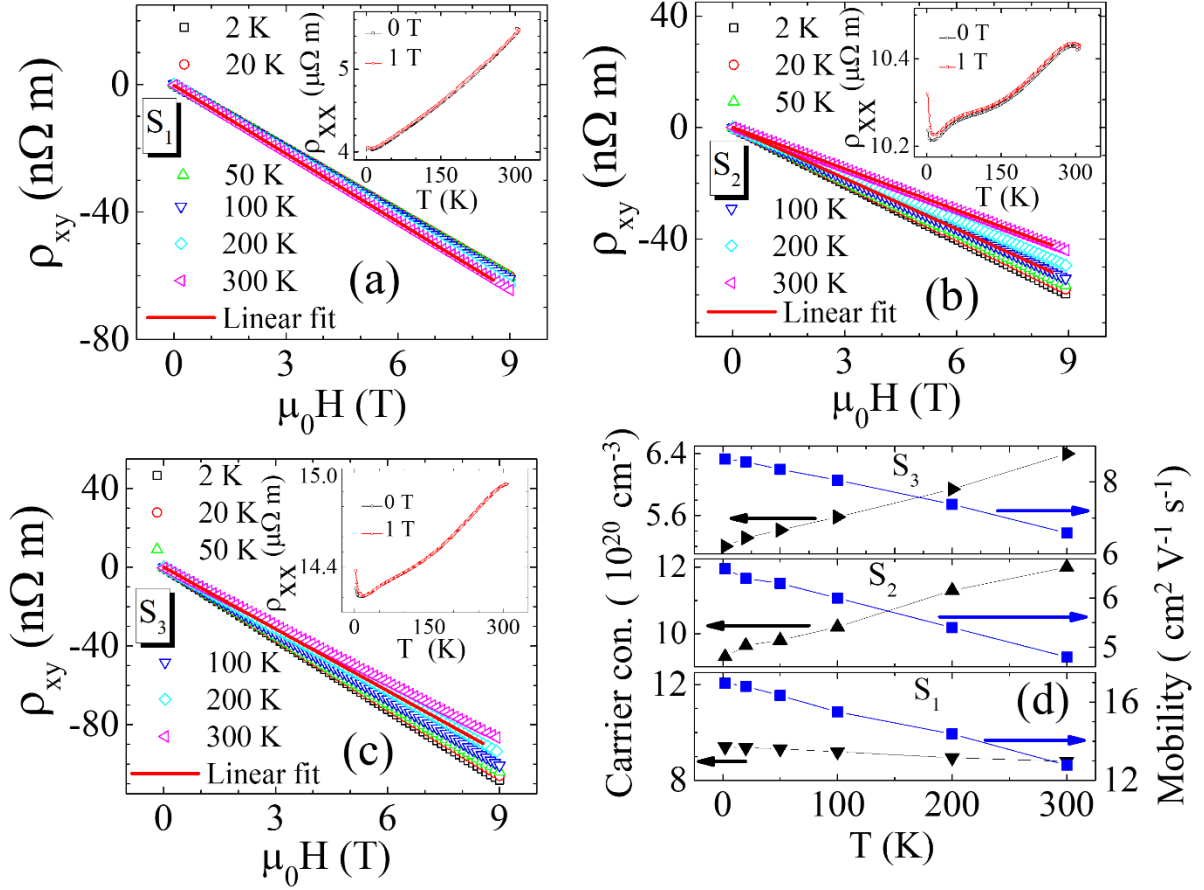


Figure 16: Hall resistivity (ρ_{xy}) of Bi_2Te_3 measured between 2-300 K as a function of magnetic field for (a) S_1 (b) S_2 (c) S_3 and each inset of figure shows resistivity (ρ_{xx}) as a function of temperature in 0 T & 1 T field. Red line in the figure shows linear fitting. (d) Carrier concentration and mobility of sample S_1 , S_2 and S_3 .

on lowering the temperature below $\sim 20 \text{ K}$ which may be a signature of weak localization. We also note that the mobility of our best film (S_1) is still lower by a factor of ~ 18 compared to the mobility of the films made by MBE [40].

In Fig. 17, we show the magnetic field dependence of the longitudinal resistivity (ρ_{xx}) of the S_1 film at several temperatures measured with the field directed perpendicular to the plane of the film. A small OMR ($(\rho_{xx}(H) - \rho_{xx}(0)) / \rho_{xx}(0)$) is observed, reaching only 0.4 percent at 20 K and 9 T. The OMR measured at 2 K also shows a distinct cusp which develops in the field range of 0 to $\pm 0.5 \text{ T}$. This cusp is not seen in the data taken at $T \geq 20 \text{ K}$. A similar low temperature feature in OMR is also seen in MBE-grown epitaxial films [16], which was interpreted as a signature of weak antilocalization [40,41]. The other interesting feature of the OMR seen in Fig.10 is its non-saturating nature even at the highest field of 9 T. The non-saturating OMR is a prominent feature of several semimetals, including the Dirac and Weyl semimetals [42, 43]. A diverse set of explanations are available for its origin including the quantum character of

electronic transport in 2D gapless topological surface states of a structural pure material [42-44] to charge inhomogeneities and conductivity fluctuations in polycrystalline samples [45, 46].

The OMR of semimetals and narrow bandgap semiconductors has been traditionally understood in the framework of the Kohler rule [47, 48], which stipulates that the OMR should scale with field as $\sim f(\mu_0 H / \rho_{xx}(0))$, where the OMR is defined as $[\rho_{xx}(H) - \rho_{xx}(0)] / \rho_{xx}(0)$ with $\rho_{xx}(H)$ and $\rho_{xx}(0)$ being the in-field and zero-field longitudinal resistivities. In Fig. 18(a to c) we plot the OMR of samples S₁, S₂ and S₃ respectively as a function of $\mu_0 H / \rho_{xx}(0)$ on a log – log scale and fit the data with the Kohler function i.e., $MR \propto (\mu_0 H / \rho_{xx}(0))^m$ where m is a scaling factor. While the data for all three samples at $T \geq 20$ K show a power law behavior typical of normal non-magnetic metals, the exponent, m , does not have a universal value. The variation of m as a function of temperature is shown in Fig. 18(d). The exponent m is equal to two for a perfectly electron – hole compensated system [49]. Here $m = 1.7 \pm 0.1$ in the temperature range of 50 K to 300 K. A substantial drop in m is seen for $T \leq 50$ K. A similar behavior of m has been noticed in other semimetals as well [50, 51]. It has been argued that this deviation is due to a change in the relative concentration of electrons and holes or a change in their relative mobilities [49, 50]. However, the constant slope of the ρ_{xy} vs field curves in Fig.16 does not indicate a change in n_e/n_h . We suggest that the change in m below 50 K is presumably due to the field induced antilocalization effect, which becomes progressively stronger at lower temperatures.

We now present the results of ρ_{xx} and ρ_{xy} measurements as the external magnetic field is rotated in the plane of the film. A sketch of the experimental geometry used for the anisotropic resistivity measurements is shown earlier in Fig. 7. Here a constant current of density ≈ 133 A/cm² runs in the x-direction and the induced electric fields E_{xx} and E_{xy} along the x and y-directions respectively, are measured as a function of the angle Φ which varies from -90° to 270° . At $\Phi = 0$ the applied magnetic field is parallel to the direction of current density J . For thin films of well-known ferromagnets like permalloy, the Φ -dependence of the longitudinal resistivity $\rho_{xx} (= E_{xx}/J_x)$ and transverse resistivity $\rho_{xy} (= E_{xy}/J_x)$ follow [20, 26].

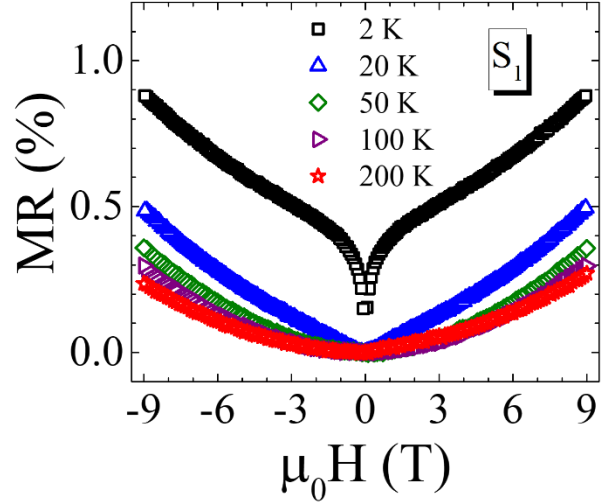


Figure 17: OMR of S1 at different temperatures where magnetic field is applied perpendicular to the sample plane. MR% is calculated as $((\rho_{xx}(H) - \rho_{xx}(0)) / \rho_{xx}(0)) \times 100$.

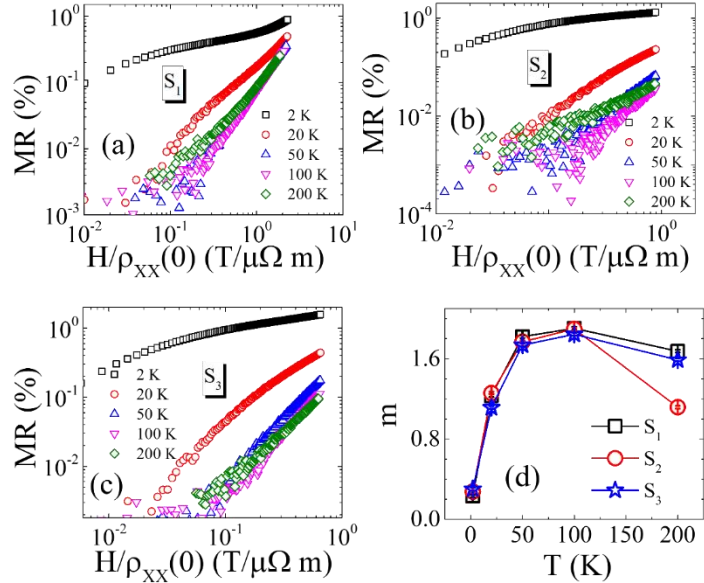


Figure 18: OMR scaling and fitting using Kohler's rule for sample (a) S₁, (b) S₂ and (c) S₃ with m taking as a free parameter. (d) derived m parameter as function of temperature.

$$\rho_{xx}(\Phi) = \rho_{\perp} + \Delta\rho \cos^2(\Phi) \quad (1a)$$

$$\rho_{xy}(\Phi) = \Delta\rho \sin(\Phi) \cos(\Phi) \quad (2a)$$

Here $\Delta\rho = \rho^{\parallel} - \rho_{\perp}$ where $\rho_{\perp}$ and $\rho^{\parallel}$ correspond to the value of ρ_{xx} for $\mu_0 H$ perpendicular to J_x and for $\mu_0 H$ parallel to J_x respectively. In this geometry, the induced E_{xy} is also called the planar Hall effect (PHE). The source of anisotropic magnetoresistance $\rho_{xx}(\Phi)$ and PHE $\rho_{xy}(\Phi)$ in magnetic alloys is the s-d scattering near the Fermi energy [27, 51]. However, in recent years, as discussed earlier in section B3.3.1, robust $\rho_{xx}(\Phi)$ and $\rho_{xy}(\Phi)$ have been measured in non-magnetic topological insulators and Dirac/Weyl semimetals and their origin has been attributed to non-trivial band topology and conduction in topologically protected surface states. However, the band topology and surface states are well-defined only in single crystals and epitaxial films. Alternatively, the anisotropic transport may be a manifestation of inhomogeneous current distribution in a disordered material [51, 52] or a result of strong spin-orbit coupling in these heavy element-based compounds. To address this issue, it is worthwhile to measure the anisotropic magnetoresistance and PHE in polycrystalline films of Bi_2Te_3 with different degrees of structural disorder.

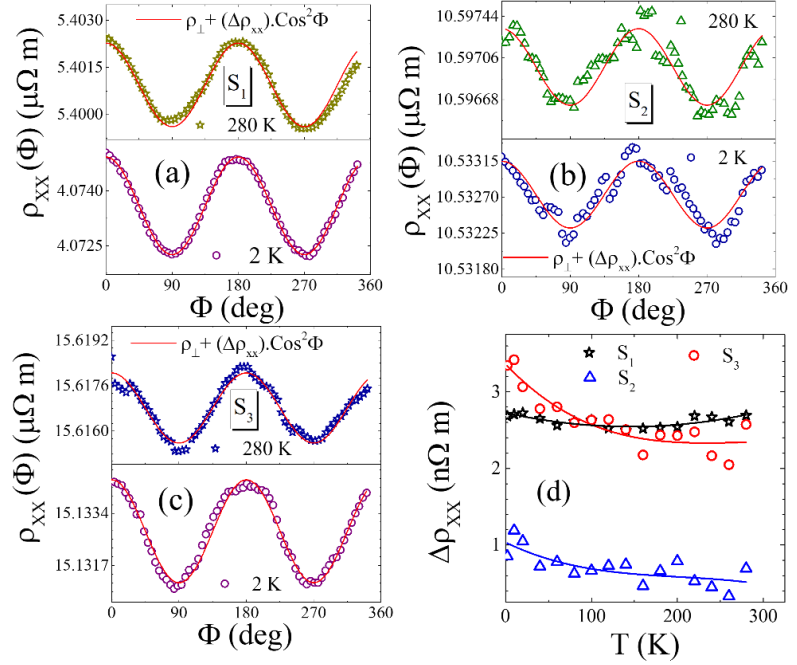


Figure 19: The variation of ρ_{xx} as a function of the angle (ϕ) between the direction of a 9 T field and the direction of current density J_x . The data are obtained from symmetrization of plus and minus field response and symmetrize data at 2 K and 280 K are shown for ρ_{xx} . (a) S_1 (b) S_2 (c) S_3 The solid red lines in a, b and c are fits to the equation of the type $\rho_{xx}(\phi) = \rho_{xx}(0) + \Delta\rho_{xx} \cos^2 \phi$. (d) AMR as a function of temperature for S_1 , S_2 and S_3 . At lower temperature below 50 K sample S_3 has 1.2 times higher AMR compared to sample S_1 .

In Fig. 19(a) through Fig. 19(c) we show the $\rho_{xx}(\Phi)$ data at two representative temperatures, 2 K and 280 K for samples S_1 , S_2 and S_3 respectively. These data have been extracted by symmetrizing the longitudinal resistance R_{xx} measured at +9 T and -9 T. The angular dependence of ρ_{xx} in all three cases follows the dependence expressed in Eq. 1. Fig. 19(d) summarizes the temperature dependence of AMR ($\Delta\rho = \rho^{\parallel} - \rho_{\perp}$) for all three samples. While the $\Delta\rho$ remains constant over the measured temperature range, its value is much larger in the sample deposited at 150 °C. It is worth mentioning here that the percentage change in AMR in these polycrystalline thin films of Bi_2Te_3 thin films (0.05%) is smaller than the ferromagnets like permalloy (2%) [53] and FeGaB (0.4%) [54], where s-d scattering contributes to this anisotropy. Interestingly however, it is comparable to the maximum AMR of 0.06% reported for epitaxial films of Bi_2Te_3 [40]

Unlike the case of ρ_{xx} , the measurement of ρ^{PHE} has two sources of error; one comes from the normal Hall effect due to misalignment of the field direction with respect to the plane of the sample, and the other from the misalignment of the E_{xy} measurement leads, which results in a voltage drop across them even at zero-field. We have corrected the ρ_{xy} data for these errors by symmetrizing the $+\mu_0 H$ and $-\mu_0 H$ data and by setting the zero-field value of ρ_{xy} to zero.

In Fig. 20(a) through Fig. 20(c), we show the Φ -dependence of ρ_{xy} at four representative temperatures for samples S_1 , S_2 and S_3 respectively. The field symmetrized $\rho_{xy}(\Phi)$ fits the equation $\rho_{xy}(\Phi) = \Delta\rho_{xy} \cos \Phi \sin \Phi$. The value of planar Hall resistivity ($\Delta\rho_{xy}$) extracted from these fits for all three samples is plotted as a function of temperature in Fig. 20(d). The PHE of the well-crystallized sample is the largest (≈ 2.8 n Ω m) and remains constant over the entire temperature range. For the other two samples, which show a low-temperature upturn in resistivity (see inset of Fig. 16), a marginal increase in $\Delta\rho_{xy}$ is seen at $T \leq 50$ K. We also note from Fig. 19 (d) and Fig. 20(d) that the amplitudes of AMR and PHE for each sample are nearly the same, as one sees in ferromagnetic alloys. Earlier measurements of PHE in Bi_2Te_3 and related topological insulators have evoked the role of surface states in leading to the AMR and PHE. However, our experiments show that this anisotropic in-plane transport is seen in polycrystalline films as well. This observation suggests the role of bulk states in generating the PHE voltage. Indeed, a recent study Zhang et. al. [26] shows that the strong spin orbit coupling drives non-linearity in charge carrier momentum in TIs, which may contribute to a planar Hall effect.

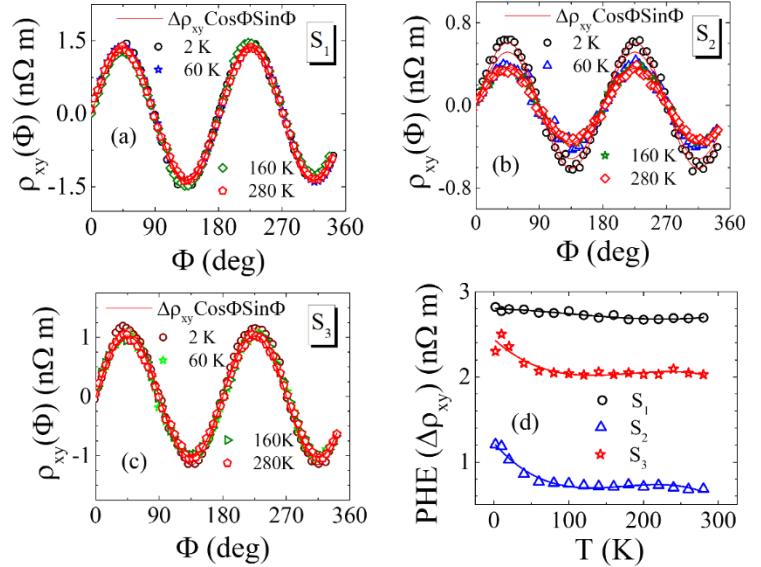


Figure 20: The variation of ρ^{PHE} as a function of the angle (ϕ) between the direction of a 9 T field and the direction of current density J_x . The data are obtained from symmetrization of plus and minus field response and symmetrize data are shown for ρ_{xy} at temperature 2 K, 60 K, 160 K and 280 K (a) S_1 (b) S_2 (c) S_3 . The solid lines in a, b and c are fits to the equation of the type $\rho_{xy}(\phi) = \Delta\rho_{xy} \sin \phi \cos \phi$ to the ρ_{xy} data. (d) PHE as a function of temperature for S_1 , S_2 and S_3 . PHE

B3.4 Deposition of β -tungsten films

The non-equilibrium A-15 phase of tungsten has the largest spin Hall angle ($\Theta_H \sim 0.33$) amongst all 5d metals [6]. This property, together with low electrical resistivity, low electromigration and corrosion resistance of tungsten make β -W a very promising spin-orbit-torque (SOT) material. However, the stabilization of this non-equilibrium phase requires special deposition conditions. Earlier, it was found that low growth rate and oxygen impurities in the sputtering plasma stabilize the β -phase. However, oxygen contamination is difficult to control, and the oxygen-doped films tend to have high electrical resistivity. We have successfully deposited nearly phase pure β -W films by injecting controlled amount of nitrogen gas in the Ar sputter plasma. This was achieved with two separate mass flow controllers, one for pure Ar and the other for 30% nitrogen in Ar. Fig.21 shows the X-ray diffraction profile of two films, one made in controlled $\text{N}_2 + \text{Ar}^+$ plasma and other in pure Ar at low power. The pure Ar film still has some traces of the most intense (110) reflection of the α -phase. The electrical resistivity of some of these films has been measured down to 2 K. Fig.22 shows clear signatures of robust superconductivity in one of such films where the T_c is 3.6 K. We are currently in the process of depositing a series of β -W films of different thickness on

epitaxial Fe₇₅Co₂₅ to measure FMR driven ISHE. These experiments will establish the spin diffusion length in β -W.

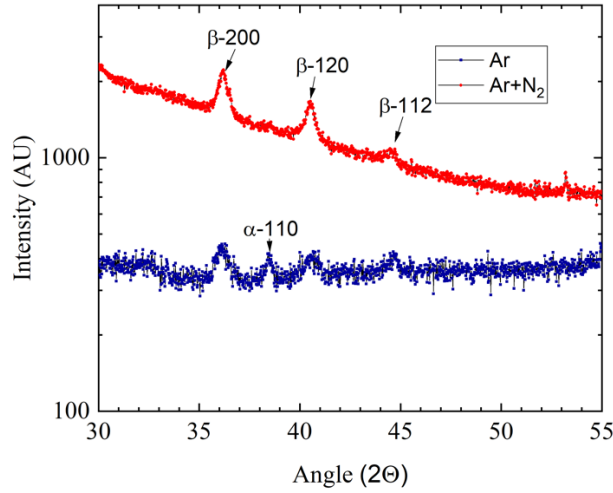


Fig. 21: X-ray (Cu K α) diffraction profile of two films of tungsten deposited at low rate (~ 0.04 nm/s), one in pure Ar and other in Ar diluted with $\sim 5\%$ N₂.

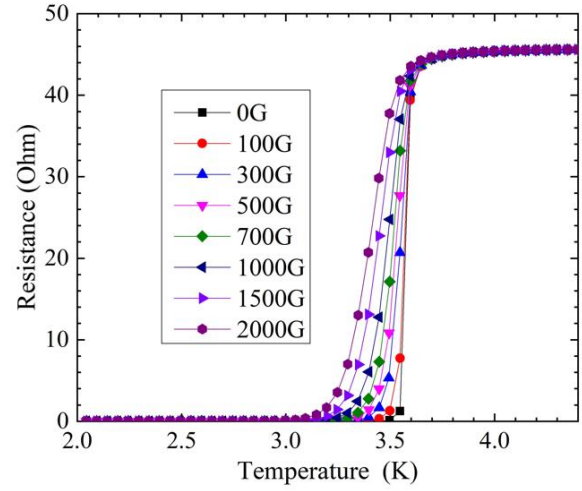


Fig. 22: Superconducting transition in beta-tungsten film at several out-of-plane fields.

B.4 Ferromagnetic resonance driven inverse spin Hall effect in FeGaB-BiSb bilayers

The resonant precession of the macroscopic magnetization vector $\mathbf{M}$ of a ferromagnetic layer under a radio frequency field H_{rf} applied orthogonal to a dc magnetic field (H_{dc}) is damped as the angular momentum is lost through emission of magnons and interactions with magnetic inhomogeneities in the system. This loss of angular momentum is significantly higher when the FM film is interfaced with a spin orbit coupled non-magnetic metal (SOC-NM). The spin current density injected into the SOC-NM is expressed as [55],

$$\mathbf{J}_s = (\hbar/4\pi) g(1/M_s^2) < \mathbf{M}(t) \times d\mathbf{M}/dt > \quad \text{----- (3),}$$

where g and M_s are spin transfer conductance of the interface and saturation magnetization of the FM layer respectively. This flow of spin current across the interface leads to the generation of a charge current in the SOC-NM through the process of ISHE. The charge and spin current densities are related as [12],

$$\mathbf{J}_c = \Theta_{SH} (\mathbf{J}_s \times \boldsymbol{\sigma}) \quad \text{----- (4),}$$

where Θ_{SH} is the spin Hall angle of the SOC-NM and $\boldsymbol{\sigma}$ the spin polarization vector of the spin current set by the external magnetic field. Under open circuit conditions, the charge current leads to the development of an electric field in the SOC-NM layer orthogonal to the direction of the magnetic field and is antisymmetric on field reversal.

Measurements of ISHE response of a SOC-NM under RF excitation have been used extensively to extract the fundamental quantity Θ_{SH} of SOC-NMs [37]. However, a precise determination of the spin Hall angle with this method is plagued by several other effects that too lead to the generation of an open circuit voltage orthogonal to dc magnetic field. The dominant spurious effects that contaminate the true ISHE voltage are as follows [56].

- (i) Non-zero spin orbit coupling in the FM layer leads to anisotropic magnetoresistance and planar Hall effect. The rf current in the sample results in a rectified dc voltage due to AMR and PHE through the $\mathbf{J} \times \mathbf{m}$ term in the force equation whose time average gives a constant voltage. This contribution depends on the phase difference between $\mathbf{J}$ and $\mathbf{m}$. It is also antisymmetric across H_{res} .
- (ii) In inhomogeneous FM films with spin absorbing surfaces and interfaces with SOC, the spin current produced by a precessing magnetization may generate a voltage through ISHE. The magnitude of this voltage is very small and in contrast to the signal of phase sensitive rectification, it is symmetric in field as this process involves incoherent spins and hence the voltage signal is phase independent. In literature, this effect has been labeled as magnonic charge pumping in broken inversion symmetry systems like GaAsMn [57].
- (iii) If the magnetic dead layer has strong SOC, then the rf current in it can produce a spin current through the process of spin Hall effect. This alternating spin current may affect the magnetization of the film and produce a voltage through PHE/AMR on entering the better part of the FM layer.
- (iv) Thermal gradients across the interface may also lead to a Nernst voltage that will be symmetric in field across H_{res} .

To see if these effects are dominant in a plain film of FeGaB, we have measured ISHE voltage in FeGaB films of various thickness. In the following, we present the method of preparation of FeGaB films, measurements of their magnetization with vibrating sample magnetometry, measurements of magnetization dynamics with FMR and finally the ISHE data. The large microwave induced dc voltage in plain films of FeGaB is a very important result. It allows us to correctly interpret the effects of spin angular momentum transferred by the precessing magnetization in FeGaB layer across the FeGaB/BiSb interface. Such measurements are discussed in section B 4.2.2 of this report. Here, we present the dc voltage generated in plain films of FeGaB when their magnetization is driven resonantly by an orthogonal rf magnetic field.



Fig. 22: 2.2 Tesla vector vibrating sample magnetometer used for measurements of field and temperature (100 K to 350 K) dependence of magnetization.

B4.1 Spin pumping and ISHE voltage in FeGaB films:

FeGaB films were grown by DC sputtering at 50 watts on sapphire substrates at room temperature using a stoichiometric $\text{Fe}_{75}\text{Ga}_{15}\text{B}_{10}$ target in the integrated PLD/sputtering system shown in Fig.5. The target was sputtered in $\approx 3 \times 10^{-3}$ mbar of ultra-high purity argon gas. The static magnetization of these films was measured in both in-plane and out of film plane field geometries using the vector VSM shown in Fig. 22. Schematic diagrams of the experimental arrangement used for measurements of FMR and FMR driven ISHE in the FeGaB are shown in Fig. 23 and 24. We have used the home built FMR spectrometer, shown in Fig.1 for this purpose. A 2 to 2.5 mm wide and ~ 8 mm long sample is mounted on the meander line GCPW (Fig. 24) in a flip-chip configuration such that the rf and dc magnetic fields are orthogonal to each other. A 100 μm thick mica sheet is inserted between the sample surface and signal line of the GCPW for electrical isolation. This precaution avoids the shorting of GCPW due to the conducting nature of FeGaB films.

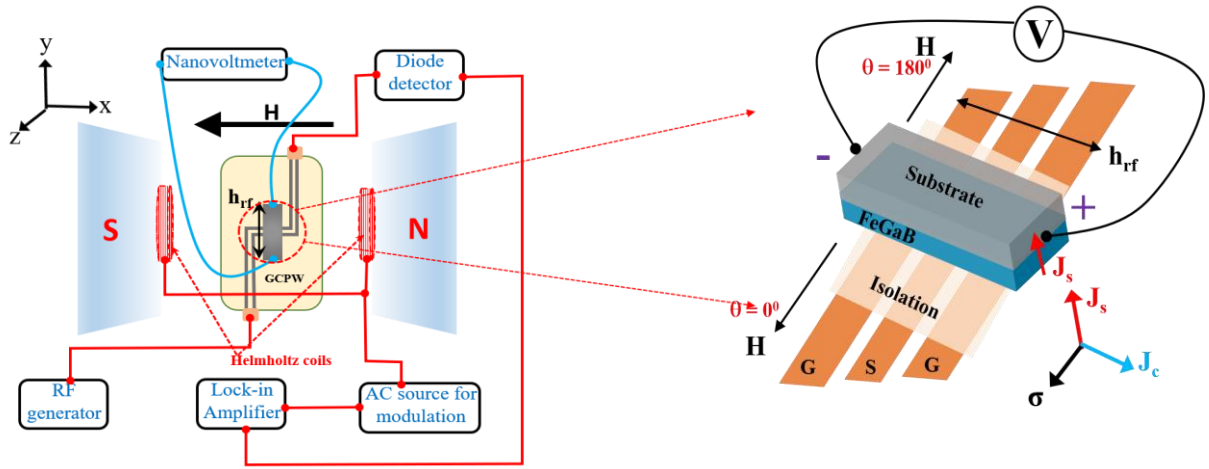


Fig. 23 & 24: Schematic illustration of experimental setup used for FMR and ISHE measurements. FMR measurements are done using the phase sensitive detection technique where the external magnetic field is modulated 855 Hz. FeGaB thin films are placed in a flip chip configuration on the signal line of a grounded coplanar waveguide (GCPW). The DC voltage generated due to Inverse spin Hall effect (ISHE) is measured using a nanovoltmeter.

Two fine ($\sim 50 \mu\text{m}$ diameter) copper wires attached to the end of the sample with silver paint allowed measurements of the ISHE induced dc voltage with a nanovoltmeter. Both FMR and ISHE measurements were performed at a fixed frequency while the dc field was scanned from 0 to $\pm H_{\text{max}}$. The RF source power was varied from -15 dBm to $+15 \text{ dBm}$ to check for nonlinearities in the sample response and to optimize the signal to noise ratio while staying in the linear response regime. The FMR data were collected phase sensitively by modulating the dc field with a pair of Helmholtz coils. But for the ISHE, the signal strength was good enough to measure without modulation with a dc nanovoltmeter.

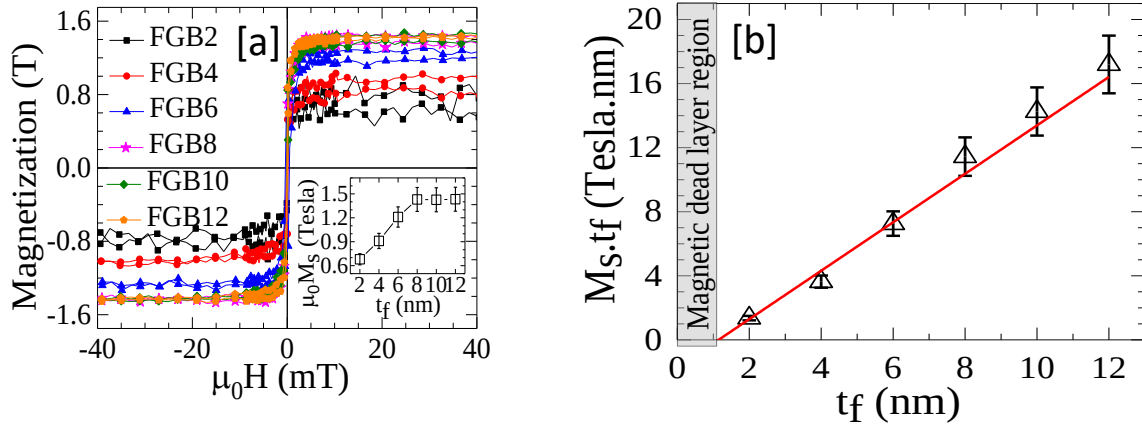


Fig. 25: **(a)** M-H loops of FeGaB thin films of different thickness ($t_f = 2, 4, 6, 8, 10$ and 12 nm) measured at room temperature. Magnetic field is applied parallel to the plane of film. Inset shows saturation magnetization ($\mu_0 M_s$) as a function of film thickness. **(b)** Change in product of M_s and t_f as a function of film thickness. Red line is a linear fit to show the point of magnetic dead layer formation for $t_f \leq 1.2 \text{ nm}$.

Fig. 25(a) shows the in-plane magnetic hysteresis loops of FeGaB films with varying thickness (t_f) at room temperature. Samples with different thickness are named FGB t_f . For example, a 4 nm FeGaB film is named FGB4. These films are soft magnets with a saturation magnetization ($\mu_0 M_s$) that varied from 0.68 T for the lowest thickness sample FGB2 to 1.43 T for the 12 nm film (FGB12). The M_s value increases linearly with film thickness up to 8 nm and then saturates at higher thickness (inset of Fig. 25(a)). The lower values of M_s in thinner films suggest the existence of a magnetic dead layer of thickness 1.2 nm (see Fig. 25(b)).

The results of FMR measurements on FeGaB films of different thickness are presented in Fig. 26. The RF absorption is characterized by a differential Lorentzian line-shapes (inset of Fig. 26). The reduction in thickness of FeGaB film tends to increase the resonance field (H_r) and FMR linewidth (ΔH_r) due to the change in saturation magnetization and induced surface anisotropy. The FMR line-shapes were fitted to the following equation.

$$\frac{dP}{dH} = K_1 \frac{4\Delta H(H-H_r)}{[4(H-H_r)^2 + (\Delta H)^2]^2} - K_2 \frac{(\Delta H)^2 - 4(H-H_r)^2}{[4(H-H_r)^2 + (\Delta H)^2]^2} + \text{slope}H \quad (5)$$

where K_1 and K_2 are the symmetric and anti-symmetric coefficients and H the applied dc field. Fitting with Eq. 5 allow us to calculate H_r and ΔH at each microwave frequency. When the dc

magnetic field is in the plane of the film and in the absence of any in-plane magnetic anisotropy, the resonance field H_r depends on frequency, through the Kittel equation.

$$f = \frac{\gamma}{2\pi} \mu_0 \sqrt{H(H + M_{eff})} \quad (6),$$

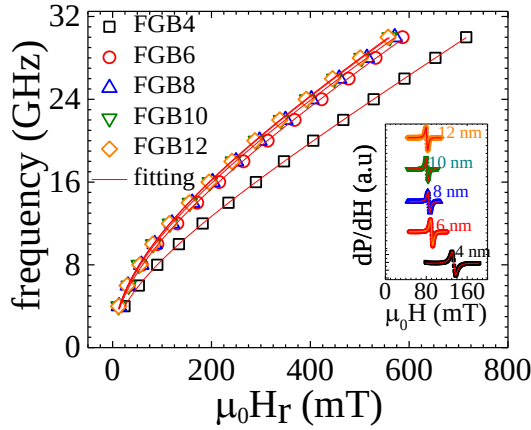


Fig. 26: Change in FMR resonance field ($\mu_0 H_r$) plotted as a function of microwave frequency for FeGaB films of various thickness. Open symbols show the experimental data points calculated after Lorentzian line shape fitting; solid red line is the Kittel fitting. Inset shows the corresponding FMR curves at 10 GHz, red line is Lorentzian line-shape fitting.

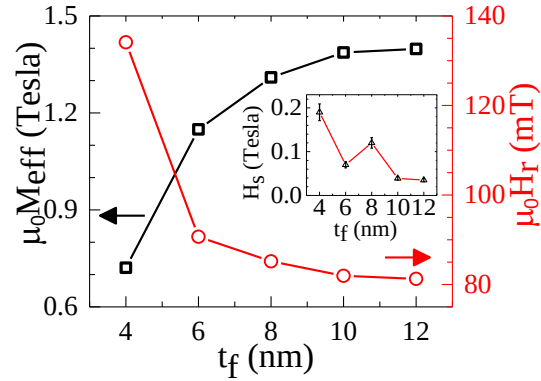


Fig. 27: Change in FMR resonance field ($\mu_0 H_r$) plotted as a function of microwave frequency for FeGaB films of various thickness. Open symbols show the experimental data points calculated after Lorentzian line shape fitting; solid red line is the Kittel fitting. Inset shows the corresponding FMR curves at 10 GHz, red line is Lorentzian line-shape fitting.

where $\mu_0 M_{\text{eff}}$ is the effective saturation magnetization and γ is the gyromagnetic ratio. The change in $\mu_0 M_{\text{eff}}$ and $\mu_0 H_r$ (at $f = 10$ GHz) as a function of t_f are shown in Fig 27. The increasing trend of $\mu_0 M_{\text{eff}}$ with the increasing film thickness is consistent with the results of dc magnetization measurements shown in Fig. 25. $\mu_0 M_{\text{eff}}$ has a contribution from surface anisotropy field and can be written as,

$$\mu_0 M_{\text{eff}} = \mu_0 M_s - H_s \quad \text{----- (7),}$$

where H_s is the induced surface anisotropy field, plotted in inset of Fig. 27. The H_s drops with the increase in film thickness. This induced surface anisotropy presumably helps in breaking structural inversion symmetry in FeGaB thin films.

Fig 28(a) shows the change in the width of the resonance (ΔH) with the applied microwave frequency. Here the data have been fitted to the Landau-Lifshitz-Gilbert (LLG) equation [6,54],

$$\Delta H = \Delta H_0 + \frac{4\pi f \alpha}{\gamma} \quad (8),$$

where ΔH_0 is the inhomogeneous line width broadening caused by sample imperfections and surface defects, and α is the Gilbert damping constant. The frequency dependence of ΔH can derive contributions from (i) Gilbert damping of the precession which is linear in frequency and (ii) two-magnon scattering, which goes as power of frequency. The linear dependence of ΔH on excitation frequency as seen in Fig. 28(a) indicates that the dissipative is dominated by the Gilbert process. We also note in Fig. 28(b) that the α and ΔH_0 decrease with the increase in film thickness, suggesting reduction in surface defects and anisotropy in thicker films. It is instructive to plot α as a function of $1/t_f$. The data shown in Fig. 28(b) follow a dependence of the type,

$$\alpha = \alpha_B + \frac{\alpha_s}{t_f} \quad (9)$$

where α_B is the intrinsic Gilbert damping constant and α_s the interface/surface contribution. A fit of the data to this equation yields $\alpha_B = 1.4 \times 10^{-3}$ which is in close agreement with the previously reported values [58], and $\alpha_s = 34.10 \times 10^{-3} \text{ nm}^{-1}$.

We now present the results of FMR driven dc voltage generated in a direction perpendicular to the direction of dc field. This voltage has been measured for films of thickness 4, 6, 8, 10 and 12 nm. Fig. 29 displays the result of these measurements. This voltage signal reverses sign on reversing the direction of the DC field, which is consistent with the prediction of Eq. 4 for the ISHE voltage. However, the RF rectification effects due to AMR of the magnetic film also leads to a dc voltage that is antisymmetric in

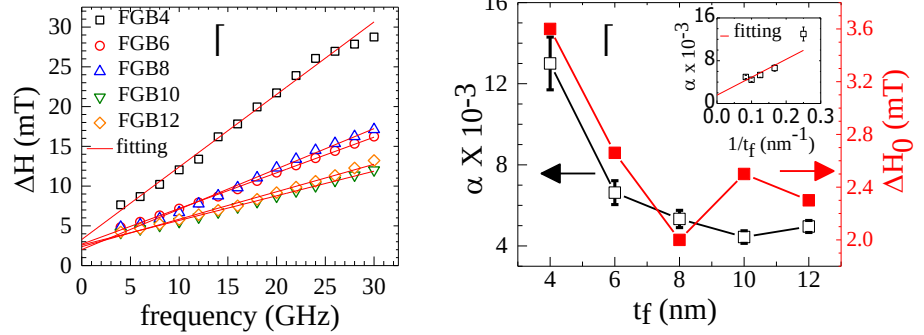


Fig. 28: **(a)** Change in FMR linewidth (ΔH) as a function of microwave frequency for FeGaB films of various thickness. Open symbols are calculated ΔH while red line is the LLG fitting. **(b)** Calculated Gilbert damping constant (α) and inhomogeneous linewidth broadening (ΔH_0) as a function of sample thickness. Inset shows the α vs $1/t_f$ where open symbols are calculated values while red line is linear fitting. Intercept at y-axis shows the α for bulk FeGaB system.

field. To extract the true ISHE contribution to the dc voltage, we carry out an analysis of the peak shape by fitting the response to sum of symmetric and antisymmetric Lorentzian functions [59],

$$V_{mix} = K_s \frac{\Delta H^2}{(H-H_r)^2 + \Delta H^2} + K_{as} \frac{-2\Delta H(H-H_r)}{(H-H_r)^2 + \Delta H^2} \quad (10).$$

The result of this fitting procedure is illustrated in Fig. 30 for the 10 nm thick FeGaB film. This analysis clearly shows that both symmetric and antisymmetric contributions to dc voltage are present.

Taking note of the geometry of our experiment, we can conclude that the antisymmetric contribution is due to the planar Hall effect in these magnetic films whose contribution is maximum when the angle between $\mathbf{J}_x$ and effective field (vector sum of RF and dc magnetic fields) is at 45 degree with respect to the direction of $\mathbf{J}_x$. Neglecting the contribution of possible Nernst effect completely, we attribute the symmetric part of the voltage to ISHE in these films. The symmetric part of the DC voltage for FeGaB films of various thickness is displayed in Fig. 31. The sign of this voltage, when analyzed in the framework of Eq. 4, indicates that the spin current is directed towards the film – substrate interface where the dead layer presumably acts as a sink for dissipation of the spin current.

In the inset of Fig. 31 we plot the room temperature resistivity of the FeGaB films as a function of their thickness. The resistivity approaches a saturation value beyond a thickness of ≈ 8 nm. As the layer thickness increases, ρ value decreases and then saturates for thicker films. We have analyzed the V_{ISHE} data in the framework of a spin diffusion model proposed by Wang et. al [60], which predicts,

$$\frac{V_{ISHE}}{\rho} \propto \lambda_{SD} \tanh\left(\frac{t_f}{2\lambda_{SD}}\right) \quad (11),$$

where λ_{SD} is the spin diffusion length of FeGaB. This analysis yields the value of $\lambda_{SD} = 2.08 \pm 0.28$ nm, which is in close agreement with results on other FM metallic systems. To rule out

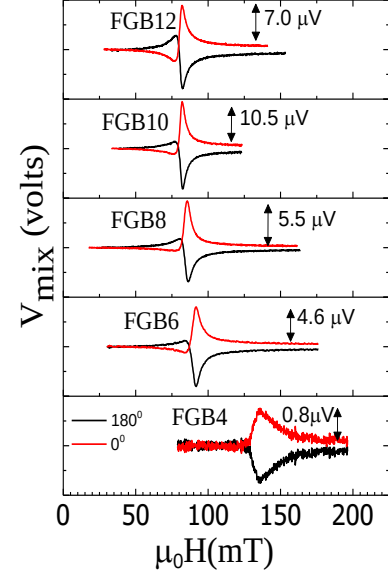


Fig. 29: FMR driven dc voltage signal (V_{mix}) at 10 GHz of 0 and π angle of external magnetic field with respect to the sample length at RF input power = +15 dBm.

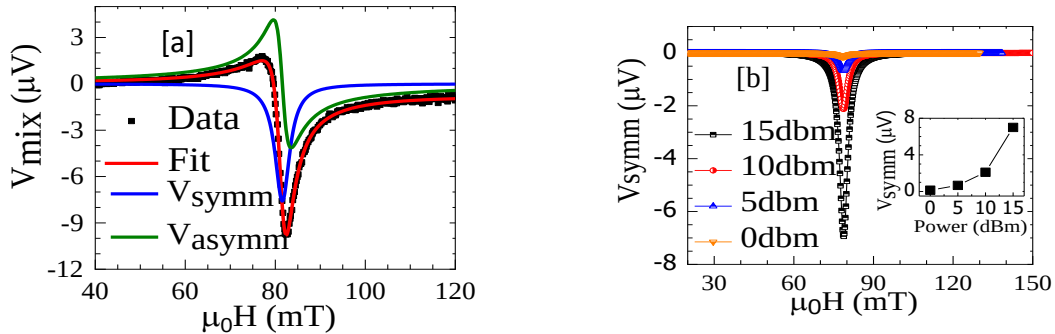


Fig. 30: (a) The FMR driven dc voltage signal of the 10 nm thick FeGaB film is split into symmetric and asymmetric components as discussed in the text. (b) Power dependent symmetric part for the sample FGB10 at 10 GHz, inset shows the increase of maximum voltage as a function of input RF power at 10 GHz.

the effect of the Nernst effect in our measurements, we have performed a control experiment on FGB8 sample with different magnetic field sweep rate. However, the magnitude of voltage signal does not depend upon the sweep rate and remains the same. This supports our finding that symmetric signal generated is primarily from the self-induced ISHE mechanism in FeGaB system.

B4.2 FMR driven spin pumping across FeGaB – BiSb interface

We have carried out extensive studies of FMR, FMR-induced ISHE and time domain terahertz

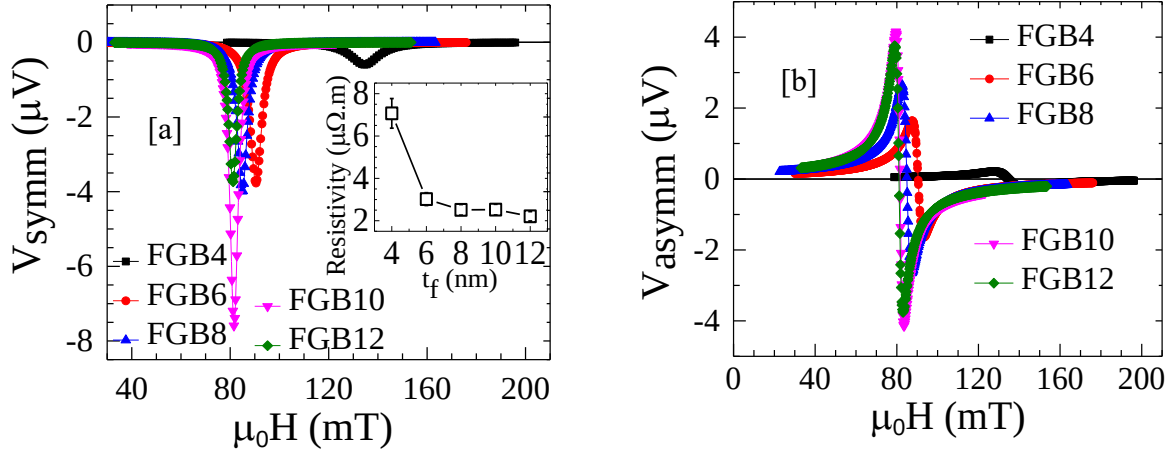


Fig. 31: **(a)** V_{symm} at 10 GHz for different thickness of FeGaB at + 15 dBm input power. Inset shows the change in resistivity (ρ) with the increase in thickness of FeGaB. **(b)** V_{asymm} at the same power and frequency as for (a).

spectroscopy (TDTS) of FeGaB (A, 6 nm)/BiSb (B, d nm) bilayers protected by a 2 nm thick MgO film deposited *insitu* in our integrated PLD/Sputtering growth system (Fig. 5). While the FeGaB and BiSb were made by dc-sputtering of 2-inch diameter targets, the MgO top layer was deposited by laser ablation of a MgO target. All layers were grown at ambient temperature on polished (0001) sapphire substrates.

Fig. 32 shows the magnetic hysteresis loops of three samples with $d = 0, 2$ and 8 nm, measured in an in-plane magnetic field geometry. While a slight increase in loop width and saturation field is discernable for the samples with BiSb, the magnetization loops indicate a magnetically very soft material with in-plane magnetization. The inset of Fig. 33(a) shows the field derivative of the RF power (dP/dH) absorbed by the FeGaB/BiSb samples of different BiSb layer thickness. The FMR spectra of all the samples were characterized by a single derivative peak, symmetric on field reversal. Each spectrum was fit with the derivative of a sum of the symmetric and antisymmetric Lorentzian to extract the resonance field H_r , and the half-width-at-half-maximum linewidth ΔH of the resonance. The behavior H_r as a function of the excitation frequency is plotted in the main panel of Fig. 33(a). The solid lines in the figure are fit to Kittel Equation (Eq.6) for isotropic in-plane FMR response.

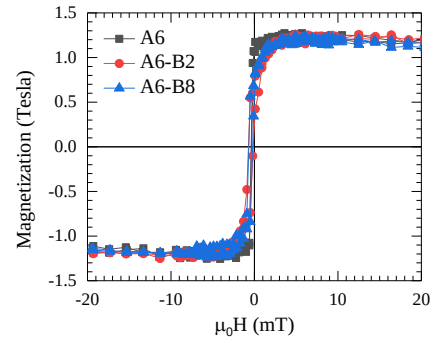


Fig. 32: Magnetization loops of FeGaB (6 nm)/BiSb (d nm) bilayers with $d = 0, 2$ and 8 nm measured at ambient temperature (≈ 300 K) with external magnetic field aligned parallel to the plane of the film.

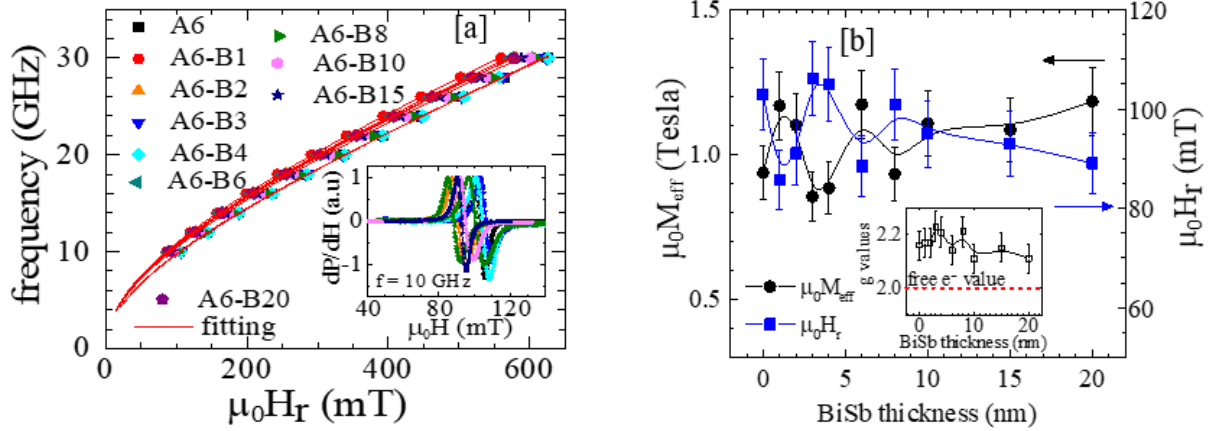


Fig. 33: (a) FMR frequency (GHz) vs resonance field (tesla) for FeGaB (6 nm) / BiSb (d nm) bilayers of $d = 0, 1, 2, 3, 4, 6, 8$, and 15 . Solid lines are fits to Kittel equation. Measurements have been done in intervals of 2 GHz at RF input power of +15dBm. Inset shows the FMR line shape at 10 GHz.

This procedure yields the effective value of magnetization affected by anisotropy fields. The fitting also gives the effective Lande g -factor. The variations of M_{eff} and H_r as a function of BiSb thickness are plotted in Fig. 33(b). It is interesting to note that the H_r and M_{eff} remain constant within the error of measurement for all values of BiSb thickness. The variation of FMR linewidth as a function of the excitation frequency for all bilayer samples is plotted in Fig. 34(a). A fit of these data to LLG equation (Eq. 8) yield the

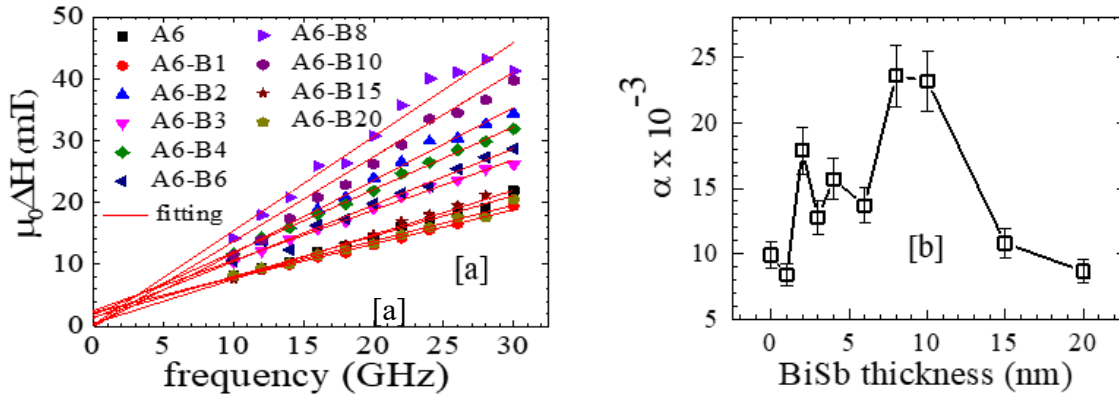


Fig. 34: (a) The FMR linewidth ($m_0\Delta H$) in mT deduced from fitting the measured dP/dH curves is plotted as a function of frequency in GHz. The linear fits to these data yield the Gilbert damping parameter α . The variation of α with the BiSb layer thickness is plotted in Fig. (b).

inhomogeneous linewidth broadening (ΔH_0) and the Gilbert damping coefficient α . Since the transfer of spin angular momentum across the interface due to spin pumping and its absorption by a strong spin – orbit coupled non-magnetic metal provides a channel for damping of magnetization precession, the dependence of α on the BiSb layer thickness is important to understand its spin current absorption characteristics. The variation of α as the BiSb layer thickness is increased is shown in Fig. 34(b).

The damping coefficient increases as more and more RF driven spin current is absorbed by the increasingly thicker BiSb layer. However, this damping eventually reaches a peak value when the BiSb

layer thickness approaches the spin diffusion length. Any further increase in the BiSb thickness reflects the pumped spin current back into the ferromagnet, which seemingly leads to a decrease in α .

The results of FMR driven ISHE voltage measurements at 10 GHz on all the samples are displayed in Fig. 35. Panel (a) of this figure shows the geometry of the ISHE measurement. A $\sim 2 \text{ mm} \times 8 \text{ mm}$ sample is placed on the coplanar waveguide in a flip chip geometry such that the RF magnetic field is along the length of the sample. The dc magnetic field is in the plane of the sample and directed along the width.

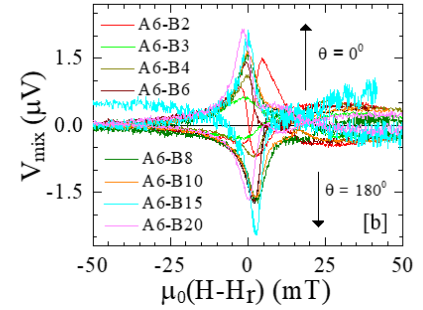
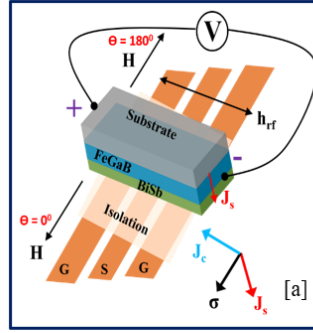


Fig. 35: (a) The experimental arrangement used to measure the dc voltage induced across the length of the sample because of spin pumping and microwave rectification effects. The sample is mounted in a flip-chip geometry. The polarity of the voltage is due to spin pumping driven ISHE. (b) The measured voltage plotted as a function of normalized field. The signal is antisymmetric on field reversal.

The figure also shows the polarity of the dc field and the direction of the ISHE driven charge current (J_c) in the BiSb layer. The measured dc voltages for 0- and 180-degree dc field orientation are shown in panel (b). The x-axis of the plots is normalized with respect to the resonance field for a meaningful comparison of the induced voltage for samples of different thickness. Clearly, the ISHE response is antisymmetric in field. To separate the spurious contributions of AMR from the true ISHE response, the plus and minus field data have been antisymmetrized in field and then analyzed following Eq. 10. The symmetric and asymmetric parts of the dc voltage signal are displayed in Fig. 36 (a) and (b) respectively. We have also measured separately the planar Hall effect and anisotropic

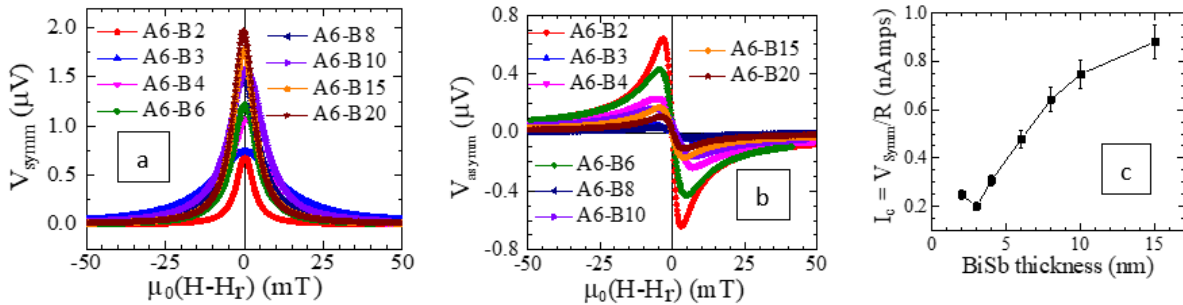


Fig. 36: (a) The symmetric component of the voltage (V_{symm}) calculated after fitting the raw data of Fig. 26 is plotted as a function of the normalized field for samples with various BiSb layer thicknesses. (b) shows the asymmetric voltage (V_{asymm}) arising primarily from planar Hall effect in the ferromagnet. (c) The charge current (I_c) induced by ISHE in BiSb layer is calculated from V_{symm} and the sample resistance R .

magnetoresistance of these samples. These studies indicate that the asymmetric part of the voltage comes primarily from the PHE of FeGaB/BiSb bilayer. Fig. 36(c) shows the variation of the charge current in BiSb layer due to ISHE as a function of the layer thickness. A saturation of I_c is clearly seen as the BiSb layer thickness becomes comparable to spin diffusion length.

B4.3. Spin to Charge conversion at FeGaB/Ag/BiSb heterostructures

A parallel approach for production of spin polarized currents identified in recent years relies on the inverse Rashba – Edelstein effect (IREE) in a two-dimensional electron gas (2DEG) in close proximity of the FM layer [61-64]. The spin-to-charge conversion efficiency of the IREE process is quantified by a parameter λ_{IREE} which is equal to $\alpha_R \tau_S / \hbar$ where α_R is the Rashba coefficient and τ_S signifies the relaxation time to show the balance between spin injections in relation with spin momentum scattering [65,66]. Since λ_{IREE} has the dimensions of length, it is called the IREE length. Currently, the search for new interfaces or surfaces based on topological insulators (TIs) to realize stronger IREE effects at ambient temperature is in focus [66,67]. Most of the earlier studies on FM-TI heterostructures are based on crystalline FMs interfaced with epitaxial films of the Bi_2Se_3 class of TIs [68]. However, an alternative and perhaps superior alternative to Bi_2Se_3 is the three-dimensional (3-D) TI $\text{Bi}_{1-x}\text{Sb}_x$ for x in the range of 0.03 to 0.22. The large spin Hall angle ($\theta_{\text{SH}} \approx 52$) [69] and high electrical conductivity ($\approx 2.5 \times 10^5 \Omega^{-1} \text{ m}^{-1}$) even in a polycrystalline state [70] make it a powerful TI for spin orbit torque applications. A simple structure to address IREE induced

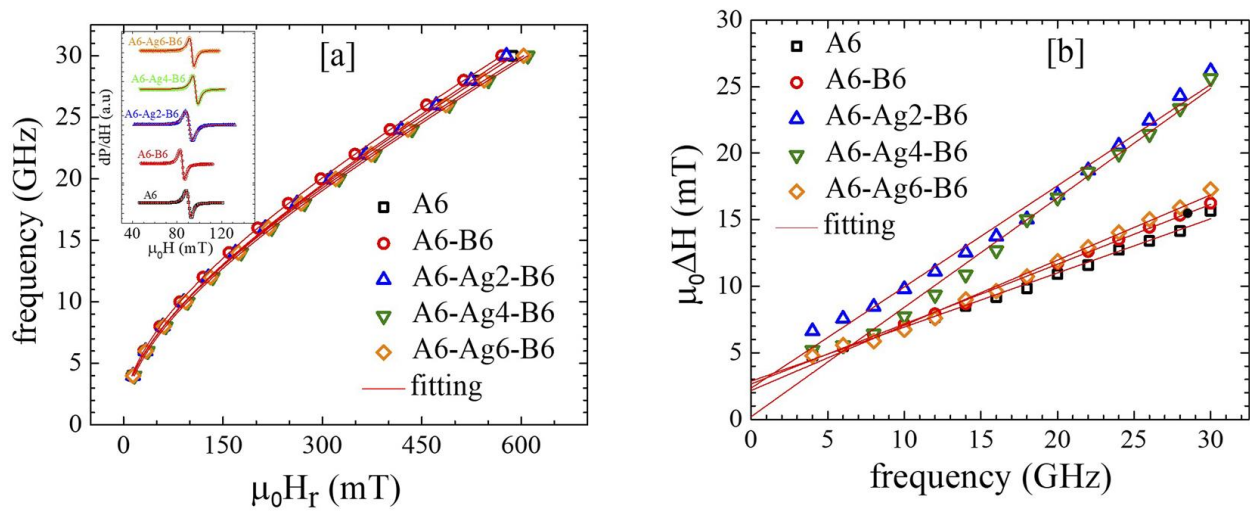


Fig. 37(a) The variation of FMR resonance frequency plotted as a function of magnetic field for FeGaB/Ag/BiSb trilayers of different Ag thickness. Symbols represent the resonance field calculated from FMR line shape analysis and solid lines are the fit to Kittel equation. Inset shows the differential FMR signal at 10 GHz with Lorentzian line-shape fitting. (b) Change in FMR linewidth (ΔH) as a function of microwave frequency for FeGaB/Ag/BiSb multilayer of different Ag layer thickness. Symbols are calculated ΔH while red lines are LLG fitting.

by BiSb can be realized by integrating it with magnetically soft amorphous FM at room temperature like the metallic glass $\text{Fe}_{78}\text{Ga}_{13}\text{B}_9$.

The model system used here is a trilayer of FeGaB/Ag/BiSb with different Ag thickness made *insitu* with a combination of dc magnetron sputtering and pulsed laser ablation. The SCC efficiency has been evaluated by measuring ferromagnetic resonance (FMR) driven ISHE voltage and extraction of the Gilbert damping (α) and interfacial spin-mixing conductance ($g_{\uparrow\downarrow}$) from the FMR line shape analysis. These measurements are carried out at ambient temperature using a coplanar waveguide based FMR spectrometer at several frequencies ranging from 4 to 30 GHz with dc magnetic field parallel to the plane of the film. The enhancement in Gilbert damping and dc voltage output at the resonance confirms efficient spin to charge conversion at the interface. We observe an increase in the charge current with the Ag layer thickness, which suggests that the IREE process at the Ag/BiSb interface dominates over ISHE in the bulk of the BiSb film. The IREE length λ_{IREE} is $\approx 5.13 \times 10^{-12} \text{ m}$ for lowest thickness of Ag and $\approx 18.85 \times 10^{-12} \text{ m}$ for the highest thickness of Ag.

Thin films with trilayer stack structure of Sapphire/FeGaB (6 nm)/Ag (0, 2, 4 and 6 nm)/BiSb (6 nm) were deposited by successive combination of DC magnetron sputtering of Fe₇₈Ga₁₃B₉, pulse laser ablation of Ag and finally DC magnetron sputtering of Bi₈₅Sb₁₅ in a load-lock vacuum system with a base pressure of 1×10^{-7} Torr. The deposition rates of FeGaB, Ag and BiSb are 0.03 nm/s, 0.06 nm/s and 0.11 nm/s respectively. Unless otherwise stated, the labelling A6, A6-B6 will be used for FeGaB and FeGaB/BiSb multilayer respectively, while A6-Ag2-B6 to A6-Ag6-B6 will be used for the identification of the samples with thickness of Ag ranging from 2 to 6 nm respectively. Our samples are of square and rectangular shapes with lateral dimensions of $5.0 \times 5.0 \text{ mm}^2$ and $2 \times 5.0 \text{ mm}^2$, respectively.

The in-plane magnetization loops $[M(H)]$ of all samples of this class were measured and it was observed that the deposition of BiSb and Ag on top of FeGaB does not affect the magnetization of a plain FeGaB film significantly. Also, the angle dependent measurements of magnetization do not reveal any preferred axis of magnetization in the plane of the film [70]. The FMR differential absorption (dP/dH) spectra were measured in a field sweep mode and a distinct single derivative peak was observed over the entire field range. The dP/dH curves of samples with different Ag layer thicknesses collected at 10 GHz for an in-plane magnetic field are shown in the inset of Fig. 37(a). The FMR line shape at different frequencies is considered a superposition of symmetric and anti-symmetric Lorentzian functions as expressed in Eq. 5.

The datasets presented in the insets of Fig. 37(a) at all frequencies have been fitted using Eq. 5 to calculate ΔH and resonance field H_r . The H_r vs f data presented in the main panel of Fig. 16(a) follow the simple Kittel equation (Eq. 6). The absence of any significant change in $\mu_0 M_{eff}$ and $\mu_0 M_s$ on addition of the Ag/BiSb interface with FeGaB layer suggests a metallurgically clean interface. Fig. 37(b) shows the dependence of FMR linewidth ΔH on the excitation frequency. The full width at half maximum of the resonance peak (ΔH) derives contributions from the intrinsic and extrinsic source of damping of the precessing magnetization vector. In the framework of Landau-Lifshitz-Gilbert (LLG) model for damping, the ΔH is expressed as Eq. 7. As seen in Fig. 37(b), the FMR linewidths are clearly enhanced in FeGaB/BiSb bilayers relative to the ΔH of the bare FeGaB film. A large enhancement is also observed in FeGaB/Ag2/BiSb trilayer, which shows the significant spin absorption due to the Rashba coupling. Fitting of experimental data shown in Fig 16(b) to Eq. 10 yields the damping coefficient α , which comes out to be 5.7×10^{-3} for FeGaB sample and increases to 12.01×10^{-3} for FeGaB/Ag4/BiSb sample. While the value of α for the bare FeGaB film is in agreement with previous reports [72], a strong enhancement is seen as the Ag layer is sandwiched between FeGaB and BiSb. The rapid rise of α with Ag interface indicates efficient absorption of spin current due to the IREE effect which will be quantified in the subsequent ISHE measurements.

The spin pumping transparency of an interface can be evaluated by calculating the real part of the spin mixing conductance $g^{\downarrow\uparrow}$ using the following relation [73],

$$g^{\downarrow\uparrow} = \frac{4\pi M_s t_{\text{FeGaB}}}{g\mu_B} (\alpha_{\text{FeGaB/Ag/BiSb}} - \alpha_{\text{FeGaB}}) \quad (11)$$

The maximum $g^{\downarrow\uparrow}$ is calculated to be $2.24 \times 10^{19} \text{ m}^{-2}$ for Ag thickness $t_{\text{Ag}} = 4 \text{ nm}$. This large value of $g^{\downarrow\uparrow}$ suggests a high efficiency of spin pumping into the Ag/BiSb layer.

Observation of IREE effect

To further establish the enhanced spin pumping and observation of IREE, we measure the induced dc voltage in the trilayer structure of FeGaB/Ag/BiSb in the FMR geometry at 10 GHz. Our main interest is now to quantify the SCC efficiency based on the interfacial IREE effect.

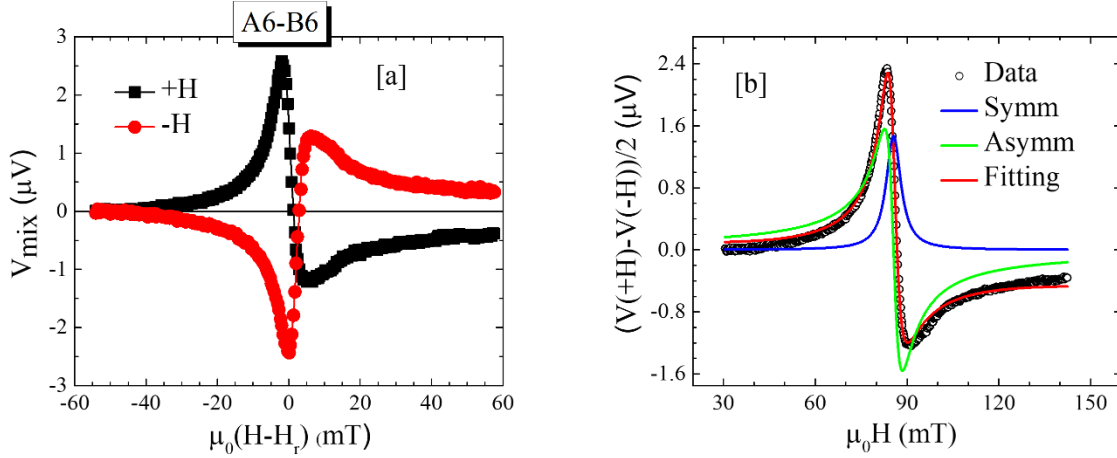


Fig. 38 (a) DC voltage measured for FeGaB/BiSb bilayer structure at 10 GHz. Black and red curve show the voltage measured for magnetic field applied in opposite directions. (b) Anti-symmetrized voltage fitted with equation 5 to separate the symmetric and antisymmetric contribution in the dc voltage.

The dc voltage signal measured at the resonance is shown in Fig. 38(a) for two opposite magnetic field directions. The polarity of the measured voltage is governed by the relation $J_C = \frac{2e}{\hbar} \theta_{SH} (J_S \times \sigma)$ where σ denotes the direction of spin polarization, this relation establishes that BiSb has positive spin Hall angle. The dc voltage signal measured at the resonance can originate from various factors like the ISHE due to spin pumping at FeGaB/BiSb interface, rectification of induced rf currents due to intrinsic SOC and AMR of FeGaB, IREE at the 2-D interface of Ag/BiSb and thermal Seebeck effect. In subsequent analysis, we ignore the small contribution of spin rectification in the bare FeGaB to the symmetric voltage signal. The Seebeck effect can be ignored by anti-symmetrizing the dc signal by taking $V_{mix} = (V(+H) - V(-H))/2$ shown in Fig. 38(b). The final signal is a superposition of symmetric and asymmetric Lorentzian functions as described earlier (Please see Eq. 10).

We first use the standard spin diffusion model to calculate the injected vertical 3-D spin current density as [73],

$$j_S^{3D} = \left(\frac{2e}{\hbar}\right) \cdot \frac{g \gamma^2 \hbar^2 \omega_{rf}^2}{8\pi\alpha^2} \frac{[\gamma 4\pi M_S + \sqrt{(\gamma 4\pi M_S)^2 + 4\omega^2}]}{(\gamma 4\pi M_S)^2 + 4\omega^2} \quad (12)$$

where $\hbar_{rf} \approx 0.2$ Oe is the RF magnetic field component in GCPW. On the other hand, the 2-D charge current density can be calculated by the relation $j_C^{2D} = \frac{V_{symm}}{Rl}$ where V_{symm} is the symmetric voltage contributing to the spin pumping signal, R is the total device resistance and l is the length of the sample and finally using the relation $\lambda_{IREE} = j_C^{2D} / j_S^{3D}$, the IREE length is evaluated. Fig. 18 shows the evolution of 2-D charge current ($I_C = \frac{V_{symm}}{R}$) due to the Rashba interfaces at Ag/BiSb.

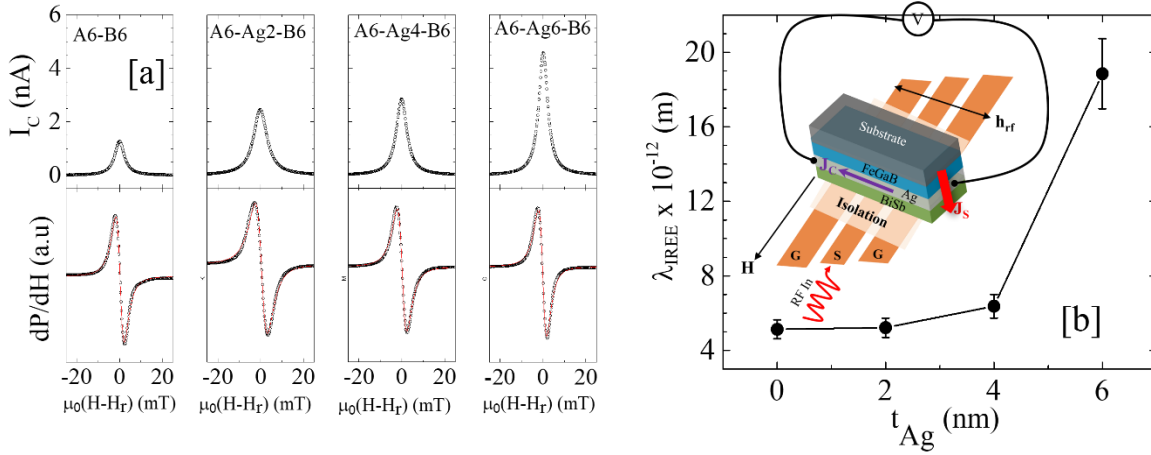


Fig. 39 (a) (top) Field dependence current $I_C = V_{symm}/R$ derived from the voltage measured at FMR driven by 10 GHz excitation, (bottom) FMR spectrum measured at 10 GHz. The red line shows the Lorentzian fitting. (b) Variation of λ_{IREE} as a function of the Ag layer thickness. Inset shows the experimental setup used for the measurements.

As seen in Fig. 39(a), the charge current at Ag/BiSb interface increases with the Ag layer thickness. The 3-D spin current density generated at the interface of Ag/BiSb is of the same order of 10^5 A/m² with the change in Ag thickness, while the 2-D charge current density ($j_C^{2D} = \frac{V_{symm}}{R.l}$) increases from 5.6×10^{-7} A/m for A6-B6 sample to approximately 2×10^{-6} A/m for A6-Ag6-B6 sample. The parameter characterizing this spin to charge conversion due to IREE effect at Ag/BiSb interface has been evaluated as $\lambda_{IREE} = j_C^{2D}/j_S^{3D}$ which increases from 5.13×10^{-12} m for A6-B6 sample to 18.85×10^{-12} m for A6-Ag6-B6 sample shown in Fig. 39(b). Here we would like to point out that the unchanged j_S^{3D} with the increase in Ag layer thickness shows that Ag act as a quasi-perfect spin transmitter because of its weak SOC and high spin diffusion length (λ_{SD}) ~ 300 nm. Secondly, the calculated θ_{SH} from the spin diffusion model ($J_{C-3D} \approx \frac{1}{2} \theta_{SH} t_{Ag} j_S^{3D}$) for Ag layer comes out to be completely unphysical ($\theta_{SH} > 1$) which shows that the spin to charge conversion due to Ag layer is negligible. The low values of IREE length have been reported before [73] where the quality of the interfaces and self-induced dc voltage in metallic ferromagnets were the main reasons. The most likely reason of low IREE length in this case would be the polycrystalline nature of BiSb because it has been reported that the single crystals of BiSb alloy exhibit high spin orbit coupling and topological surface states. The positive sign of λ_{IREE} shows that the helical spin configuration of electron-type Fermi contours in Ag/BiSb is counterclockwise which can be proved in further studies using angular resolved photoemission spectroscopy measurements.

B5. Sputter deposition of $Fe_{60}Co_{20}B_{20}$, $Fe_{75}Co_{25}$ and $Ni_{81}Fe_{19}$ thin films

$Fe_{60}Co_{20}B_{20}$ (FeCoB) is a well-known soft ferromagnetic metallic glass. Thin films of FeCoB are used extensively in magnetic tunnel junction and spin orbit torque (SOT) devices because of their amorphous structure which leads to magnetic softness as well as isotropic in-plane magnetization. Ultra-thin films of FeCoB can be tailored for perpendicular magnetic anisotropy by interfacing them with thin layers of MgO followed by selective crystallization. With the goal of making FeCoB/Bi₂Te₃ and FeCoB/BiSb SOT heterostructures, we have optimized the growth of very thin films of FeCoB with dc magnetron sputtering. These studies have also contributed to the M.S. thesis of our student Sean Davis.

Thin films of varying thickness (4 nm – 20 nm) were deposited at room temperature onto silicon wafers. The samples were

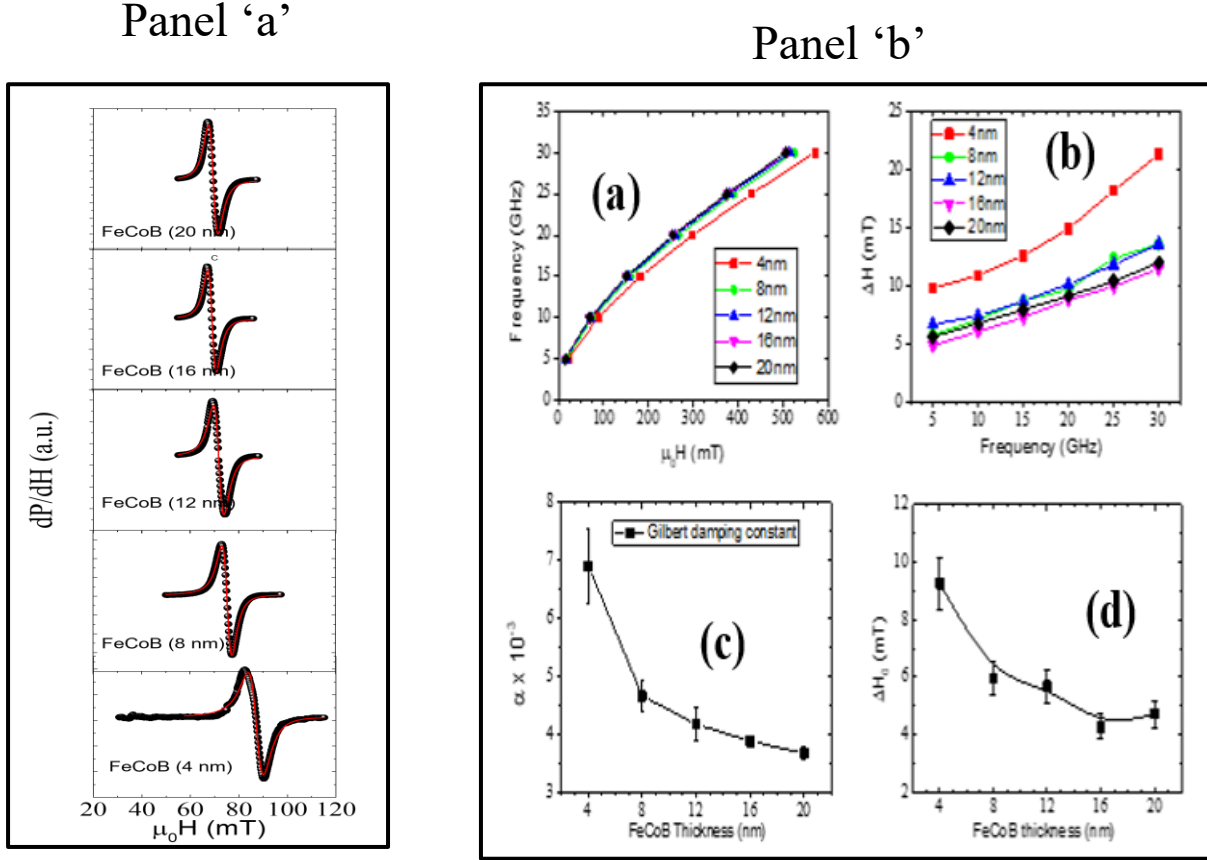


Fig. 40: Panel 'a' - FMR spectra at 10 GHz of FeCoB films of several thickness. Panel 'b' – variation of critical FMR parameters of the FeCoB films.

characterized using both DC and AC magnetic measurements. M-H hysteresis loops were measured as a function of film thickness (d) using a vibrating sample magnetometer (VSM) at room temperature. Ferromagnetic resonance spectroscopy was conducted with frequencies ranging from 5 to 30 GHz and analyzed using a two component Lorentzian function. Fig. 40(a) shows a set of FMR scans taken at 10 GHz on films of different thickness. A systematic evolution of FMR linewidth is observed with the increasing film thickness. The panels of Fig. 40(b) show the results of FMR measurements over a broad frequency range for films of different thickness. We note that the Gilbert damping parameter and the zero-frequency linewidth of FMR, both, decrease initially on increasing the films thickness and then approach an asymptotic value when d reaches ≈ 12 nm. We plan to use this experience of depositing FeCoB films from an alloy target to grow bilayers of FeCoB with MgO and multilayers of FeCoB/Pd to achieve perpendicular magnetic anisotropy of a varying degree of robustness. Such films will be integrated with the large spin Hall angle materials like β -W and Bi_2Te_3 to study SOT switching of magnetization.

B5.1. Further Studies of structure and magnetization of $\text{Fe}_{75}\text{Co}_{25}$ epitaxial films:

The $\text{Fe}_{75}\text{Co}_{25}$ (FeCo) films were deposited on (001) MgO and (001) MgAl_2O_4 (MAO) crystals by magnetron sputtering of an alloy target. The structural quality of these films has been established by X-ray diffraction measurements. The equilibrium crystallographic structure of the $\text{Fe}_{75}\text{Co}_{25}$ alloy is a body centered cubic (BCC) cell of lattice constant ≈ 2.85 Å. Figure 41(a) shows the θ - 2θ

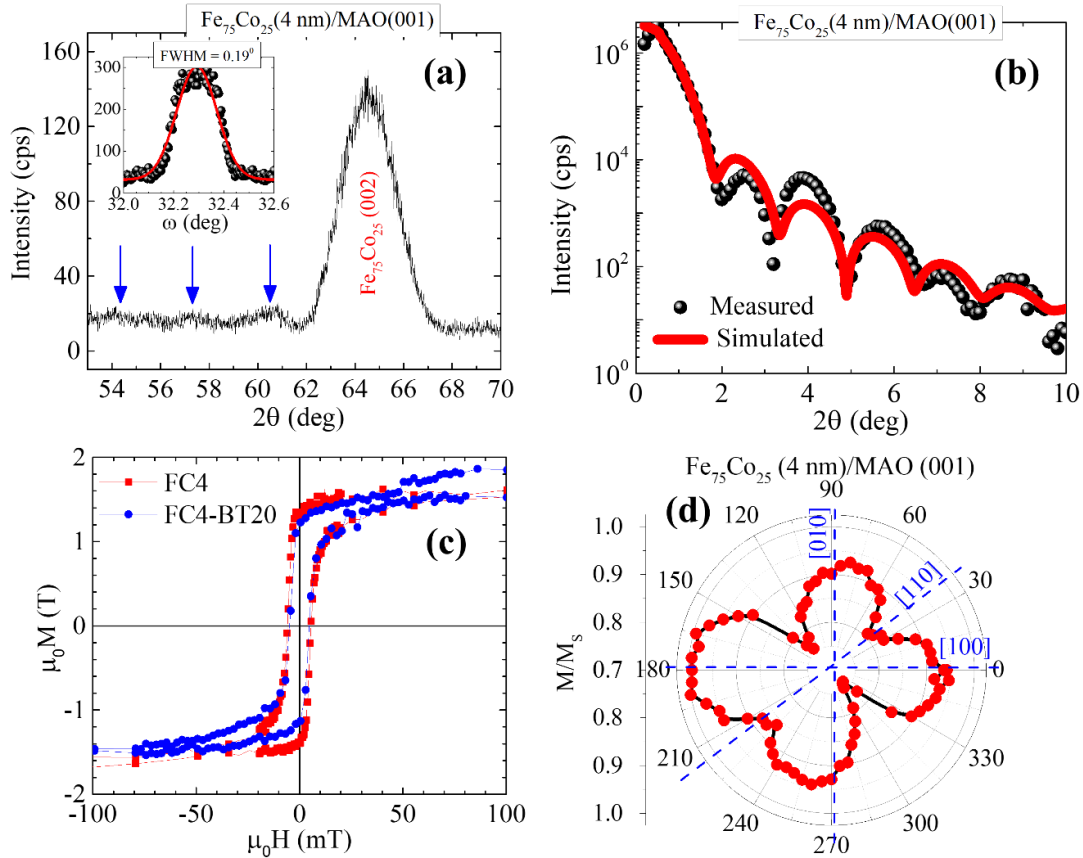


Figure 41 (a) θ - 2θ X-ray diffraction measurements of FC4 sample on MAO (001) substrate. Inset shows the XRD rocking curve of $\text{Fe}_{75}\text{Co}_{25}$ (002) peak which has a full width at half maxima (FWHM) of 0.19° . Arrows in the figure indicate the positions of Laue oscillations. (b) X-ray reflectivity scan of FC4 sample gives a surface roughness of 0.3 nm. (c) Magnetic hysteresis loops of FC4 and FC4-BT10 samples measured at room temperature with in-plane magnetic field. (d) The in-plane angular dependence of M/M_s for FC4 sample when the magnetic field is rotated from 0 to 360 degrees.

XRD pattern of a 4 nm thick sample grown on MAO (001) substrate. The prominent (002) peak at 64.5° yields an out-of-plane lattice constant of ≈ 2.90 Å. The diffraction pattern also shows weak Laue oscillations which have been indicated by arrows in the figure. The inset of Fig.41(a) is the rocking curve with a full width at half maximum of $\approx 0.19^\circ$. The clear Laue oscillations and narrow rocking curve demonstrate epitaxial growth of $\text{Fe}_{75}\text{Co}_{25}$ on MAO ($a = 8.083$ Å) when we consider a 45° rotation of the basal plane of the BCC $\text{Fe}_{75}\text{Co}_{25}$ with respect to the basal plane of face centered cubic MAO. Figure 41(b) shows the X-ray reflectivity (XRR) profile of FC4 sample. The modeling of these XRR data yield a film roughness of ≈ 0.3 nm.

Figure 41(c) shows the in-plane magnetization of a 4 nm thick film of FeCo, together with the magnetization of a FeCo-Bi₂Te₃ bilayer, which we will discuss in the later sections. The saturation magnetization of the FeCo film is $\mu_0 M_S \approx 1.65 \pm 0.2$ T with coercivity of 20 mT, which is lower by $\approx 20\%$ compared to the calculated value for bcc Fe₇₅Co₂₅ [74,75]. This discrepancy is presumably due to thin film size effects [76] as the thickness of this film is only 4 nm. Figure 4(d) shows the variation of magnetization of this film as the in-plane magnetic field is rotated from 0 to 360°. The four-fold magnetic anisotropy seen in Fig. 4(d) confirms that the easy and hard axes within the framework of in-plane cubic magnetic anisotropy are along the [100] and [110] directions respectively.

B.5.2 Ni₈₁Fe₁₉ (Permalloy) films:

We have also carried the deposition of Py thin films by dc sputtering of a Ni₈₁Fe₁₉ arc melted target. The purpose of growing these films was to integrate them with the chiral magnet Mn₃Sn thin films as well as support the research of our collaborators at AFRL-WPAFB. In Fig. 42(a and b) we show the magnetic

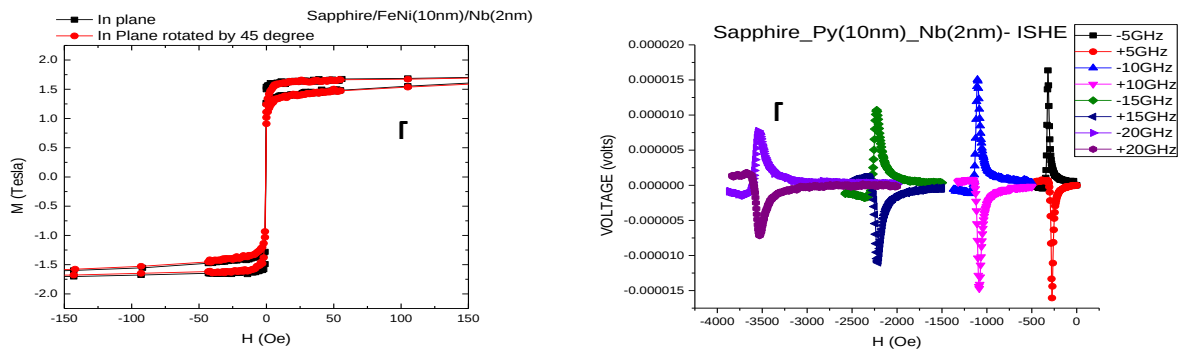


Fig. 42(a,b): Magnetization loop of a 10 nm thick Py film deposited on sapphire. The in-plane magnetic field was applied at two different orientations to check for in-plane magnetic anisotropy. (b) ISHE voltage at different frequencies. The data for positive and negative values of magnetic field have been overlapped for clarity.

hysteresis loop and inverse spin Hall voltage of a 10 nm thick Py film covered with 2 nm niobium, which was deposited to protect the film from oxidation. The values of saturation magnetization and coercivity of these films are comparable to what has been reported in the literature. We have also deposited 25 and 40 nm thick Py films on prepatterned 2-inch diameter wafers of SiO₂/Si. The AFRL team is conducting acoustically driven FMR experiments on these films.

B6. Studies of spin pumping across Bi₂Te₃ - FeCo interface:

Thin films with stack structure of (100) MgAl₂O₄/ Fe₇₅Co₂₅ (4nm)/ Bi₂Te₃ (0, 2, 4, 6, 8, 10 and 20nm)/Al(2nm) were deposited by DC magnetron sputtering of Fe₇₅Co₂₅ (99.99%) and Bi₂Te₃ (99.99%) targets in a load-lock multi-target high vacuum system with a base pressure of 8×10^{-9} Torr. The (100) cut MgAl₂O₄ (MAO) substrates were cleaned sequentially using acetone and isopropyl alcohol in an ultrasonic bath for 15 minutes before loading into the sputtering chamber. First the Fe₇₅Co₂₅ film was deposited at the rate of 0.07 nm/s at 350 °C in 5mTorr pressure of ultrahigh purity (99.9999%) argon gas. The substrates were then cooled down to room temperature and Bi₂Te₃ layers were grown on top of the Fe₇₅Co₂₅ surface at the rate of 0.16 nm/s. The sputtering power for Bi₂Te₃ was set to a lower value (≈ 20 W) to avoid surface melting of the target. Finally, to protect the Fe₇₅Co₂₅/Bi₂Te₃ heterostructure from oxidation, a thin (~ 2 nm) layer of Al was deposited at room temperature at the rate of 0.02nm/s. Unless otherwise stated, the labelling FC4 and FC4-BT2 to FC4-BT20 will be used for identification of the samples with Fe₇₅Co₂₅ (FC) thickness of 4 nm and Bi₂Te₃ (BT) ranging from 2 to 20 nm. For FMR-ISHE measurements rectangular shapes

samples with lateral dimensions of $7.0 \times 2.0 \text{ mm}^2$ were used while square shape samples ($5.0 \times 5.0 \text{ mm}^2$) were measured for THz measurements.

The spin pumping and spin to charge conversion processes at the interface of these bilayers have been studied by exciting the magnetization of the FM layer at two different time scales, namely: (1) through Ferromagnetic resonance (FMR) at GHz frequency and measurements of FMR induced ISHE dc voltage, and (2) femtosecond laser-induced spin injection followed by an ultrafast spin-to-charge current conversion by means of ISHE, which leads to emission of THz radiation. For FMR-ISHE measurements rectangular shapes samples with lateral dimensions of $7.0 \times 2.0 \text{ mm}^2$ were used while square shape samples ($5.0 \times 5.0 \text{ mm}^2$) were selected for THz measurements.

Figure 43(a) shows the output voltage (V_{mix}) of the sample FC4-BT2. We have extracted the overall ISHE plus rectification response by anti-symmetrizing the signals obtained at the plus and minus polarity of the dc field. This procedure eliminates the possible contributions of thermal voltages to the dc signal. The field symmetrized V_{mix} is derived from the superposition of symmetric and antisymmetric Lorentzian functions. This symmetrized data is fitted to Eq. 10. The symmetric dc signal (V_{symm}) produced in the FC4-BT2 sample at 10 GHz excitation with different levels of excitation power is shown Fig. 43(b). The response remains linear up to a power of +15 dBm. The V_{symm} for all the samples of the FC4-BTx family for in-plane magnetic field is shown in Fig. 43(c). However, attributing this voltage entirely to ISHE may be erroneous because a dc voltage is also produced by rectification of the eddy currents in the metallic FM layer of a FM/NM bilayer. The source of this rectification is the anisotropic electronic transport in the FM layer due to s-d scattering. This effect is clearly seen in the data of Fig. 43(c) for a plain film of $\text{Fe}_{75}\text{Co}_{25}$ (sample FC4). This dc signal is symmetric about the resonance field and changes its sign on reversing the direction of the dc field. The noteworthy feature of Fig. 43(c) is the drop in this dc voltage as Bi_2Te_3 is added on top of the $\text{Fe}_{75}\text{Co}_{25}$ layer. This suggests that the polarity of the true ISHE signal produced in the TI layer is opposite to that due to rectification of eddy currents in the FM layer. From this decreasing trend in the signal and the geometry of our experiment vis-à-vis the relation $\mathbf{j}_c = \theta_{\text{SH}}(\mathbf{j}_s \times \boldsymbol{\sigma})$, we conclude that the spin Hall angle of Bi_2Te_3 has a positive sign. This observation is consistent with the reported data on Bi_2Te_3 exfoliated crystals [77].

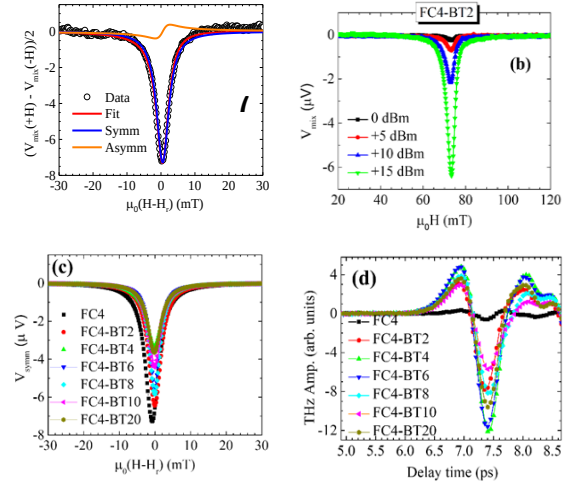


Fig. 43: (a) ISHE voltage of FC4-BT2 sample, (b) Symmetrized voltage of the same sample at different excitation power, (c) Symmetric voltage of all sample and (d) THz amplitude of the same set of samples as shown in (c).

Spin pumping across these FM-TI interfaces has also been carried out by exciting the samples with femtosecond laser pulses, which launches an ultrafast spin current $\mathbf{j}_s$ from FeCo into the Bi_2Te_3 , which subsequently converts into a charge current $\mathbf{j}_c$ by means of the ISHE, and the relaxation of this charge current results in the emission of THz radiation [40]. The time-domain traces from different samples are shown in Fig. 14(d). As is obvious from the figure, there is a clear change of the THz peak amplitude for varying Bi_2Te_3 thickness.

To quantify the spin diffusion length of Bi_2Te_3 , we have followed a strategy in which the charge current is calculated by subtracting the signal of bare FC4 from the net signal of FC4-BTx samples. The spin diffusion length of Bi_2Te_3 is estimated by the fitting of charge current ($I_c = V_{\text{peak}}/R_{\text{TI}}$) deduced from the symmetric voltage corresponds to the ISHE signal to the following equation [78],

$$I_c = \frac{V_{peak}}{R_{TI}} \propto \lambda_{SD} \cdot \tanh\left(\frac{t_{Bi_2Te_3}}{2\lambda_{SD}}\right) \quad (13)$$

Where V_{peak} , R_{TI} and $t_{Bi_2Te_3}$ are peak symmetric voltage, sample resistance, and thickness of the Bi_2Te_3 bilayer, and λ_{SD} is the spin diffusion length. The result of this analysis, which yields $\lambda_{SD} = 1.82 \pm 0.76$ nm, is displayed in Fig. 44(a).

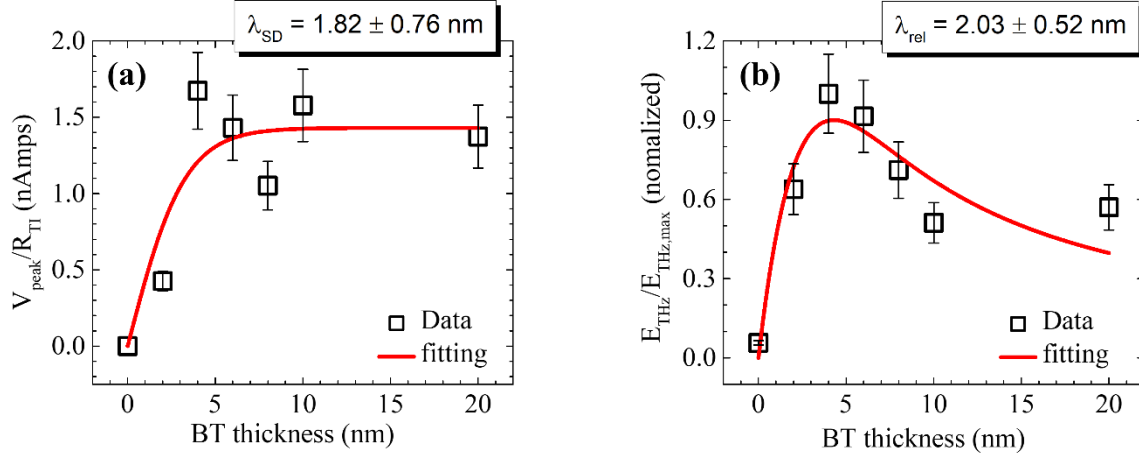


Figure 44(a) ISHE induced charge current normalized by sample width plotted as a function of Bi_2Te_3 thickness. Solid line is a fit of these data to Eq. 2, which yields the spin diffusion length $\lambda_{SD} = 1.82 \pm 0.76$ nm in Bi_2Te_3 . (b) Normalized THz electric field as a function of Bi_2Te_3 thickness, data is fitted with Eq. 5 to yield $\lambda_{rel} = 2.03 \pm 0.52$ nm.

By using the same analogy in THz measurements, we have extracted the hot electron velocity relaxation length (λ_{rel}) for Bi_2Te_3 . Fig. 44(b) shows the normalized THz peak-to-peak amplitude extracted from the time-domain traces shown in Fig. 44(c) as a function of the Bi_2Te_3 thickness. This thickness-dependent THz emission characteristic is in stark contrast to the results from ISHE measurements. Frequency dependent electric field is given by the relation [79],

$$E_{THz}(\omega) = \frac{eZ_0}{n_{air} + n_{MAO} + Z_0 G(\omega, t_{Bi_2Te_3})} J_c(\omega), \quad (5) \text{ and}$$

$$J_c(\omega) = \frac{AF_{inc}}{t_{FeCo} + t_{Bi_2Te_3}} j_s^0 \cdot \lambda_{rel} \tanh\left(\frac{t_{Bi_2Te_3}}{2\lambda_{rel}}\right) \cdot \gamma \quad (6)$$

Where, G = conductance of bilayer

AF_{inc} = fraction of absorbed pump power

j_s^0 = injected spin current

λ_{rel} = spin relaxation length (hot electron velocity relaxation length)

γ = spin hall angle

We assume that G , AF_{inc} , j_s^0 , λ_{rel} and γ to be independent of TI thickness and hence the above model can be normalized to the following equation,

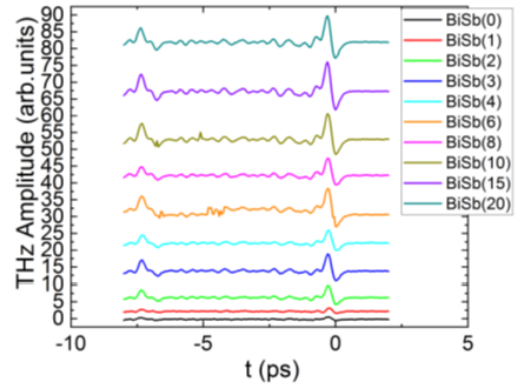


Fig. 44(c): THz emission spectra of the $FeGaB/BiSb$ heterostructures. The $BiSb$ layer thickness increases from zero to 20 nm.

$$\frac{E_{THz}}{E_{THz,max}} \approx \max \left(\frac{t_{FeCo} + t_{Bi2Te3}}{\tanh\left(\frac{t_{Bi2Te3}}{2\lambda_{rel}}\right)} \right) \frac{\tanh\left(\frac{t_{Bi2Te3}}{2\lambda_{rel}}\right)}{t_{FeCo} + t_{Bi2Te3}} \quad (14)$$

The fitting shown in Fig 44(b) using equation 14 and gives the values of $\lambda_{rel} = 2.03 \pm 0.52$ nm, which is in stark contrast to the results obtained from ISHE.

The effects of spin rectification due to AMR/PHE are not pronounced in THz measurements since AMR rolls off with the frequency and may be suppressed at higher frequency. It has been shown in earlier studies that AMR consists of both extrinsic and intrinsic contributions in which former is ascribed to the electron scattering which rolls off rapidly with frequency while the latter is independent of frequency but can be higher in complex structures like hexagonal Co [80]. However, in the present case the concentration of Co is only 25 % and the structure of the alloy is cubic.

B7. Anomalous Hall effect in epitaxial films of chiral antiferromagnet Mn_3Sn

Antiferromagnets (AFMs) are considered as the next generation spintronic materials over conventional ferromagnets (FMs) owing to their fast-switching speed, zero stray fields and stability against field-induced distortions of spin arrangements [81]. A particularly fascinating class of AFMs is presented by the compounds that lack structural inversion symmetry and possess a non-collinear arrangement of spins. Such systems host topologically non-trivial spin textures such as skyrmions [81]. The Heusler compounds of the type Mn_3Z ($Z = Sn$ and Ge) stabilized in DO_{19} crystal symmetry are examples of such materials. Mn_3Sn is an AFM with the Mn spins arranged in a Kagome type inverse triangular structure stabilized due to geometric frustration and the Dzyaloshinskii-Moriya interaction (DMI) [82]. The triangular 120° arrangement of Mn spins on the [0001] plane, set at the Néel Temperature of 420 K, yields a net uncompensated in-plane magnetic moment of the order ≈ 20 m_B /f.u. at 300 K [83,84]. A weak interplanar spin interaction stabilizes a helical phase at ≈ 250 K which changes to a spin-glass state at $T < 100$ K [85]. For utilizing the full functionality of Mn_3Sn magnetic phases, epitaxial films of this compound are needed. In this work, we present an extended study in c- axis textured Mn_3Sn thin films grown on [111] surface of MgO. The temperature dependent magneto-transport measurements in a Hall bar geometry reveal the appearance of three magnetic phases over a temperature regime of 2 to 300 K with their distinctive signatures in the AHE.

Thin film with stack structure of [111] MgO/ Mn_3Sn (100 nm)/Nb (2 nm) were deposited by DC magnetron sputtering of $Mn_{75}Sn_{25}$ (99.99%) and Nb (99.995%) targets in a load-lock multi-target high vacuum system with a base pressure of 8×10^{-9} Torr. First, the MgO substrates were preheated to 700 °C for 20 mins in high vacuum to remove any organic contaminants adsorbed on the surface. Since MgO is prone to adsorption of moisture, this heat treatment also allows removal of OH^- radicals from its surface. After this step, the substrates were cooled to ambient temperature and Mn_3Sn films were deposited at the rate of 0.073 nm/s in 5 milli-Torr of ultrahigh purity (99.9999%) argon gas. The deposited films were annealed *insitu* at 500 °C for 20 minutes. Finally, a thin (≈ 2 nm thick) layer of niobium was deposited on top of the Mn_3Sn films at room temperature for protection against oxidation. The crystallographic structure and mosaicity of the films were measured with X-ray diffraction (XRD) using a Bruker D-8 Advance X-ray diffractometer with $Cu-K_{\alpha 1}$ radiation. The Hall resistivity and anisotropic magnetoresistance of these films were measured down to 2 K in a physical property measurement system equipped with a 9-tesla superconducting magnet.

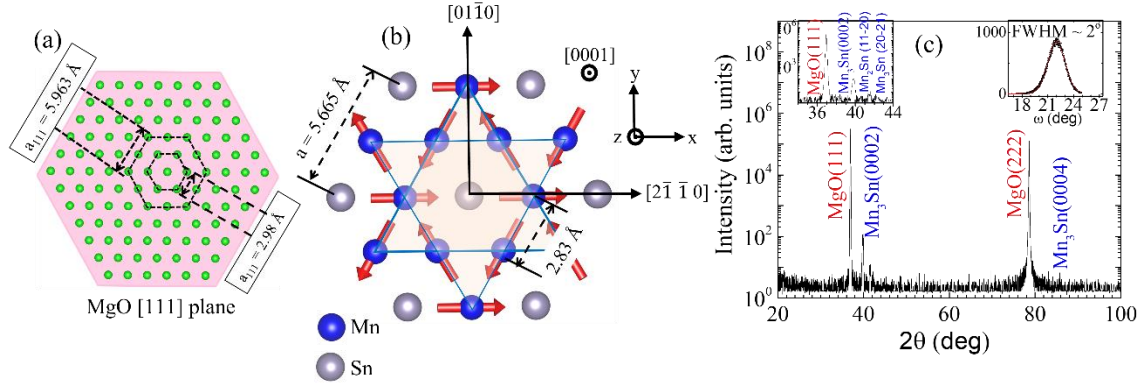


Figure 45: (a) A sketch of atomic (Mg) positions on the $\langle 111 \rangle$ plane of MgO, the Mg atoms are shown as green circles which form a hexagonal plane of lattice spacing 5.963 Å. (b) Atomic positions on the c-plane of the Mn_3Sn lattice with in-plane $[01\bar{1}0]$, $[2\bar{1}\bar{1}0]$ and out of plane $[0001]$ directions marked. The orientations of Mn spins are shown in the Kagome plane. Crystal structures are drawn using VESTA software. [30] (c) $\theta - 2\theta$ X-ray diffraction pattern of a 100 nm thick Mn_3Sn film grown on MgO $[111]$ substrate. C-plane crystallographic directions are marked. Left inset shows the zoomed XRD near (0002) peak where the small peaks near 41.8° and 42.2° correspond to the Mn_2Sn (11 $\bar{2}$ 0) and Mn_3Sn (20 $\bar{2}$ 1) respectively. Right inset shows the rocking curve measurement near (0002) which gives full width at half maxima value of $\sim 2^\circ$.

Mn_3Sn crystallizes in a hexagonal DO_{19} type structure with space group $P6_3/\text{mmc}$ and lattice parameters $a = 5.665$ Å and $c = 4.531$ Å. The Mn atoms are arranged in an inverse triangular manner forming the Kagome layers stacked along the c-axis, in which the Heisenberg interaction strength on up and down triangles is different, giving rise to a ‘breathing’ anisotropy. The epitaxial relationship between $[111]$ MgO and Mn_3Sn is illustrated in Fig. 45(a) and (b). Figure 45(c) shows the $\theta - 2\theta$ XRD pattern of a 100 nm Mn_3Sn thin film where the presence of $[0002]$ and $[0004]$ reflections of the hexagonal Mn_3Sn confirms that the $[000n]$ reflections appear next to the cubic $[111]$ reflections of MgO. Mn_3Sn lattice constant c of 4.530 Å, determined from the $[0002]$ reflection is close to the bulk value. The peaks near $2\theta = 41.8^\circ$ and 42.2° in Fig. 45(c) are attributed to the Mn_2Sn (11 $\bar{2}$ 0)

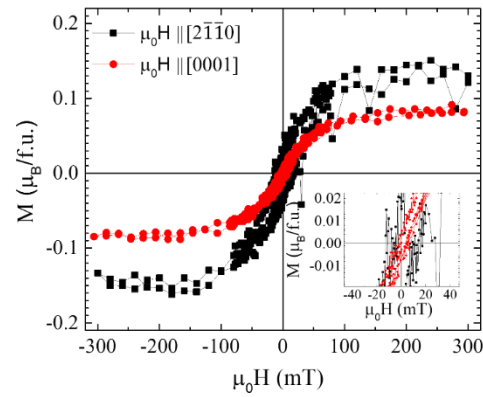


Figure 46: Magnetization of a 100 nm thick Mn_3Sn film at two orientations of the external field; in-plane parallel to $[2\bar{1}\bar{1}0]$ and out of plane, parallel to $[0001]$ directions. Inset shows a magnified view of data near zero field to differentiate the coercive field in the in-plane and out of plane directions.

and Mn_3Sn (20 $\bar{2}$ 1) respectively arising presumably due to the Mn deficiency [86]. The Mn deficient samples show a helical magnetic phase transition near 250 K due to the formation of Mn_2Sn , which has a Curie temperature of ≈ 250 K [87]. The significance of this transition will be addressed further in the following sections.

Figure 46 shows the room temperature magnetization loops of a Mn_3Sn film measured using a vibrating sample magnetometer (VSM). These data have been corrected for the diamagnetic response of the MgO substrate. The sample shows a weak ferromagnetic response with a saturation moment of $\approx 0.12 \mu_B/\text{f.u.}$ for the in-plane field. For the out-of-plane field, the moment drops to $\approx 0.08 \mu_B/\text{f.u.}$ The weak moment seen for the in-plane field indicates a small canting of the Mn spins in the a-b plane. Whereas the minuscule moment in the out-of-plane direction suggests some canting along the c-axis as well. The small in-plane and out of plane moments seen here are an order of magnitude higher than the reported values for Mn_3Sn crystals ($4\text{--}7 \text{ m}\mu_B/\text{f.u.}$) [83]. This discrepancy has been discussed previously in the context of disorder and Mn deficiency in thin films, which in turn generate the out of plane tilting of Mn moments [88]. The in-plane uncompensated moment in these epitaxial Mn_3Sn films were found to be $\sim 0.21 \mu_B/\text{f.u.}$, which is approximately double of the moments found in the present case. In these reports [88], the larger magnetization is attributed to the uncompensated moments arising due to the disorder and defects caused by excess of Sn atoms randomly occupying Mn sites as well as Mn vacancies [88]. We therefore argue that the Mn_3Sn films in present case possess lower disorder and antisite defects. We also note an anisotropy in the coercive field (H_c) of these films as shown in the inset of Fig. 2. The coercive field when the field is orientation along the $[2\bar{1}\bar{1}0]$ direction ($\mu_0 H_c \approx 10 \text{ mT}$) is higher by a factor of ≈ 3 , as compared to the H_c for the field orientation along $[0001]$. This is presumably due to the mosaicity of epitaxy in the ab-plane of the film and the measured magnetization reflects an average over many crystallites.

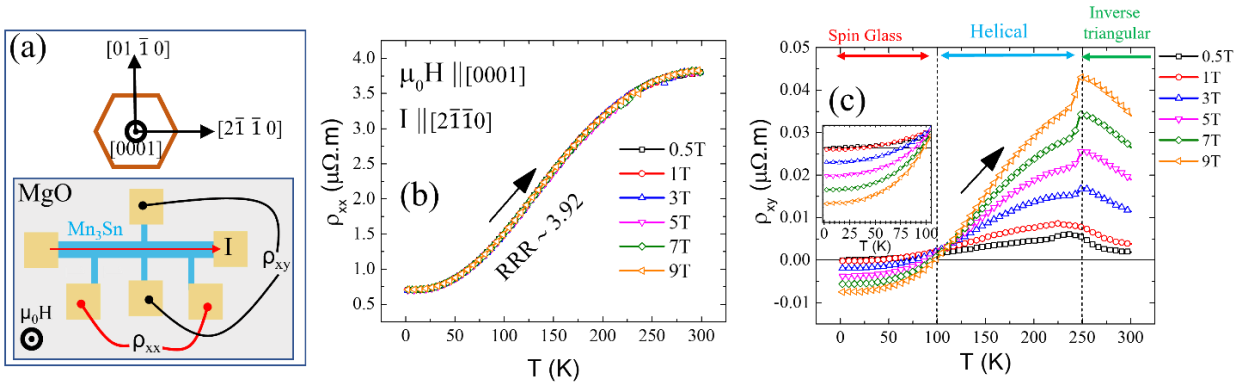


Figure 47 (a) Schematic representation of magneto-transport measurements in a Hall bar geometry. (b) Longitudinal resistivity (ρ_{xx}) measured as a function of temperature at several values of magnetic field applied perpendicular to the plane of the film ($\langle 0001 \rangle$) during field warming. (c) Hall resistivity (ρ_{xy}) measured as a function of temperature at different values of the out-of-plane magnetic field. The Hall resistivity clearly shows transitions from spin glass to helical and from helical order to inverse triangular antiferromagnetic order on warming from 2 K to 300 K. These transitions are marked by dotted vertical lines. Inset shows a magnified view of the Hall data at $T < 100 \text{ K}$.

The temperature dependent magneto-transport measurements offer a powerful means to probe magnetic phase transitions in Mn_3Sn thin films. Here we measure the longitudinal (ρ_{xx}) and Hall (ρ_{xy}) resistivity of a 100 nm thick Mn_3Sn film patterned in a Hall bar geometry, as shown in Fig. 47(a). The value of ρ_{xx} is extracted after symmetrizing the $+H$ and $-H$ data i.e., $\rho_{xx} = (\rho_{xx}(+H) + \rho_{xx}(-H))/2$ whereas the true ρ_{xy} is obtained on antisymmetrization of the transverse resistivities i.e., $\rho_{xy} = (\rho_{xy}(+H) - \rho_{xy}(-H))/2$. Figure 47(b) shows the $\rho_{xx}(T)$ data, which confirms the metallic nature of Mn_3Sn as reported in works on bulk crystals [83]. The room temperature ρ_{xx} of $3.8 \mu\Omega\cdot\text{m}$ is close to the reported value ($3.1 \mu\Omega\cdot\text{m}$) in epitaxial thin films [89]. The residual resistivity ratio (RRR ~ 3.92) marked in Fig. 47(b) is higher by a factor of ≈ 3 as

compared to the result of Taylor et.al. [89], suggesting a better-quality film. Fig. 47(b) also shows that the ρ_{xx} curves measured at different magnetic fields overlap, indicating a very low ($\approx 0.1 - 0.3\%$) magnetoresistance for the out of film plane oriented magnetic field. This value is comparable to the previously measured MR in out of plane field [89] and indicates the presence of weak magnetization in these AFM films.

B8. Magneto-transport and magnetic textures in Ho/FeCoGd/ β -W multilayers with additive chiral exchange.

The formation, stability, and dynamics of spin textures in thin films of ferromagnetic transition metals (TM) alloyed with rare-earth (RE) elements have attracted much attention in recent years [90-93]. Although the nucleation of magnetic bubble domains in TM-RE alloys was addressed extensively half a century ago, the current interest is driven by spin-orbit-torque (SOT) physics in thin films of these alloys interfaced with a non-magnetic heavy metal. The TM-RE alloys such as Fe-Gd are ferrimagnetic (FiM), where the 3d and 4f electron spin lattices are coupled antiferromagnetically. The relative strength of magnetic order parameters in these two-sublattice systems drives compensation effects where the net magnetization goes to zero, coercive field diverges, and the magnetization acquires out-of-plane anisotropy. The weak, out-of-plane magnetization of TM-RE films in the vicinity of the compensation point is perturbed easily by magnetic torques of the spin currents produced by a strongly spin-orbit-coupled non-magnetic (SOC-NM) layer proximate to the TM-RE film. The broken inversion symmetry at the TM-RE alloy – SOC-NM interface also produces a chiral exchange interaction that twists the magnetization locally to produce topologically non-trivial spin textures called skyrmions [90-93]. The dynamics of magnetic skyrmions in TM-RE thin films interfaced with 5d metals like platinum have been imaged with x-ray absorption and scattering techniques [91,92].

The current-driven dynamics of skyrmions and the twisted magnetization seen by moving electrons in the interior of a skyrmion leave their signatures in the Hall resistivity in a field regime where the magnetization is not fully saturated. While the skyrmion Hall effect has been studied in 3d ferromagnetic multilayers and inversion symmetry broken alloys, such studies on TM-RE systems are limited [93]. This issue is important since it provides a powerful tool to electronically detect the presence and motion of skyrmion in thin-film systems.

Ferrimagnets like TM-RE alloys offer a unique advantage over other ferromagnets in controlling the nucleation threshold, size, and speed of skyrmions due to their highly suppressed net magnetization near the compensation temperature (T_{com}), which can be tuned by changing the alloy composition. These advantages of FiMs can be exploited effectively if individual skyrmions could be nucleated easily in these systems. In the case of 3d FMs, the nucleation threshold for topological spin textures (TST) has been optimized by enhancing the interfacial chiral exchange. For example, periodic multilayers of (ABC)_n type where B is a metallic ferromagnet and A and C are SOC-NMs that produce the Dzyaloshinskii – Moriya interaction (DMI) of opposite sign host TSTs at ambient temperature [94,95]. Here we use a new approach to enhance DMI in (ABC)_n multilayers of ferrimagnetic TM-RE alloys. It is based on the recent finding of a large positive Hall angle (Θ_{SH}) of approximately 0.14 in elemental holmium [96]. The more-than-half filled 4f shell of Ho results in the largest Θ_{SH} amongst all 4f elements. The other SOC NM of the (ABC)_n stack is β -tungsten with a reported Θ_{SH} of approximately - 0.3. The opposite signs of Θ_{SH} in Ho and β -W allows for additive DMI in (Ho/FeCoGd/ β -W)₁₀ multilayers.

The β -W (1 nm)/[Ho(2 nm)/FeCoGd(4.5 nm)]₁₀/ β -W (4 nm) multilayers were deposited by sputtering of Ho, W, Fe₇₅Co₂₅ and Gd targets in a load-lock sputtering station of base pressure of about 1×10^{-6} Pa. The sputtering rates of Fe₇₅Co₂₅ and Gd were optimized to produce the Fe₅₅Co₁₉Gd₂₆ films by co-sputtering. We have used polarized neutron reflectivity measurements at ambient temperature to address the periodicity, both structural and magnetic, of these multilayers along with X-ray photoelectron microscopy to image magnetic textures.

The compensation temperature of TM_{1-x}RE_x alloys in thin film and bulk forms reveal that the T_{com} increases steadily on increasing the RE concentration in the alloy. In most cases, the T_{com} exceeds 300 K when x becomes more than approximately 24 %. The active magnetic layer in the periodic multilayer investigated here has the stoichiometry Fe₅₅Co₁₉Gd₂₆ with a T_{com} close to 350 K. However, a direct comparison between the response of the single-layer FeCoGd and the multilayer would need caution because one of the spin Hall elements of the multilayer is Ho with its strong magnetic signatures. The 4f moments of metallic Ho order antiferromagnetically at approximately 130 K with signatures of spin chirality. The antiferromagnetic (AFM) phase switches to a ferromagnetic (FM) phase at $T < 30$ K [97].

We have addressed the elemental and magnetic periodicity of the multilayer with polarized neutron reflectivity (PNR) measurements. Figure 48(a) shows the model-fitted reflectivity data plotted as a function of wavevector transfer Q , taken at ambient temperature in 3 T in-plane field. Data and fits are scaled by Q^4 to better highlight features across the entire measured Q range. PNR is used to determine the depth profiles of the nuclear scattering length density (ρ_N , indicative of the nuclear composition) and the magnetic scattering length density (ρ_M , proportional to the in-plane magnetization) of thin films and multilayers. Non-spin-flip reflectivities ($++$ and $--$) were measured using 0.475 nm wavelength neutrons on the PBR instrument at the NIST Center for Neutron Research. Fig. 48(a) shows a large spin splitting between R^{++} and R^{--} , demonstrating sensitivity to the in-plane sample magnetization parallel to H while the pronounced Bragg peak at $Q = 0.8 \text{ nm}^{-1}$ corresponds to the average periodicity of the superlattice. Model fitting was performed using the Refl1D software package [98]. The data could not be well fit by a perfect superlattice, but it is well fit with a model that allows for significant thickness variations in the bottom-most and top-most FeCoGd layers, as shown in Fig. 48(b). The average superlattice unit cell thickness is 7.7 nm, thinner than the expected 8.5 nm. The complex nuclear profile shows ρ_N close to expected values for the constituent materials. Nonzero imaginary ρ_N is indicative of Gd, as it is the only element in the system with a significant imaginary component at the neutron wavelength used. The magnetic profile is shown in green, and when converted to units of magnetization, corresponds to 165 kA m^{-1} for the FeCoGd layers, and a net magnetization of 136 kA m^{-1} for the [Ho/FeCoGd/W]₁₀ superlattice as a whole. It is interesting to note that the model shown corresponds to a thin (0.45 nm) and strongly negative Ho layer magnetization (-344 kA m^{-1}), although that strongly negative feature is smeared out by apparent interlayer roughness. However, while this profile is *consistent with* antiparallel alignment of the FeCoGd and Ho layer magnetizations, we cannot conclusively make this specific claim. Since the sensitivity here is to the depth profile averaged over a very large area in-plane, it is impossible to strictly distinguish large length scale intermixing from local interlayer roughness. As such, different choices in model parameterization could give similar or identical depth profiles that each fit the data to an indistinguishable degree but lead to different conclusions.

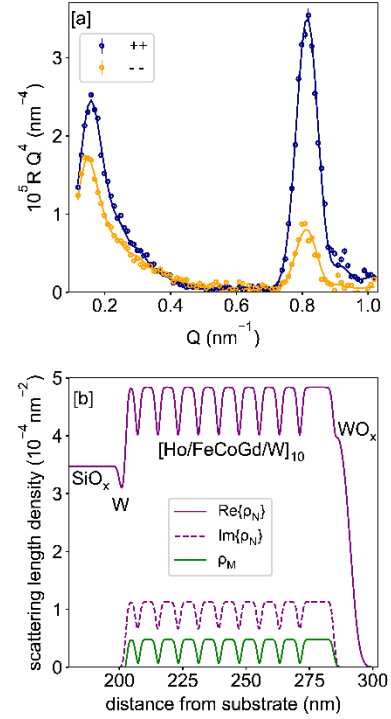


Fig. 48: (a) Spin splitting between R^{++} and R^{--} demonstrating sensitivity to in-plane sample magnetization parallel to external field. (b) Model fits to depth profile data for nuclear and magnetic scattering.

The robust conclusion that we can draw from the PNR results is that there is indeed a strongly modulated magnetic structure with FeCoGd contributing much more to the overall magnetization than does the Ho.

The magnetic domain texture of the multilayers has been imaged by means of spatially resolved x-ray magnetic circular dichroism (XMCD) at the XPEEM/LEEM endstation of the Electron Spectro-Microscopy beamline (21-ID-2) of NSLS-II. The x-ray absorption spectroscopy was performed at room temperature in a partial yield mode, with the analyzer energy window of approximately 15 eV centered on the maximum of the secondary emission peak, with the circular-left and circular-right polarization of the light, respectively, tuned to the Gd-M5 absorption edge (approximately 1173 eV). The incidence angle of the x-ray beam on the sample was about 73° to surface normal. A representative XMCD image is shown in Fig. 49. The chiral magnetic texture with equal area coverage by domains of opposite contrast, as seen in Fig. 49, suggests an out-of-plane magnetic state of a net-zero remanence. Since the contrast in the figure is related to M5 edge of Gd, its uniformity further indicates that there is no segregation of this 4f element.

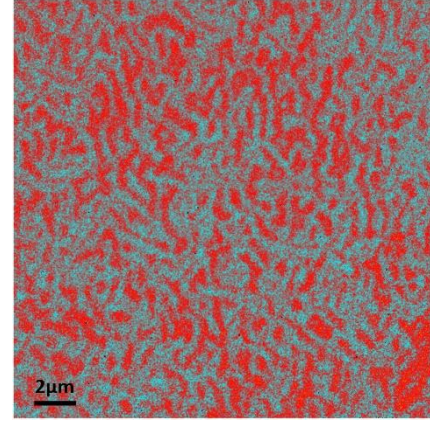


Fig. 49: X-ray magnetic circular dichroism (XMCD) image taken at the Gd-M5 absorption edge (~ 1173 eV) and showing stripe-like ferromagnetic domains.

Our electron transport and dc magnetization studies focus on the temperature regime of $2 \text{ K} \leq T \leq 350 \text{ K}$ where the net magnetization of the $\text{Fe}_{55}\text{Co}_{19}\text{Gd}_{26}$ film is dominated by the contribution of 4f sublattice. The magnetic hysteresis loops of the multilayer measured at 10, 100, 300, and 350 K with external magnetic field oriented perpendicular to the plane of the film ($\square_0 H^\perp$) are shown in Fig. 50 where, for clarity, the data for 100, 300 and 350 K have been given a rigid shift along the field axis with respect to the $M(H)$ at 10 K. We note that the saturation field, H_s , decreases rapidly following the behavior of the saturation magnetization (M_s) as we raise the measurement temperature to 300 K. In fact, parallel field ($\mu_0 H^\parallel$) measurements on the multilayer revealed that at $T < 300 \text{ K}$ the magnetization has in-plane anisotropy. At 300 K however, the saturation fields for $\mu_0 H^\perp$ and $\mu_0 H^\parallel$ configurations are identical. The $M(H)$ loop at 300 K whose zero-remnance indicates a fine multidomain structure with an equal number of up and down magnetic domains is like that of a TM-RE alloy film that hosts bubble domains. The H_s drops further along with the M_s on raising the temperature to 350 K. The temperature dependence of M_s per unit area of the multilayer film over the temperature range of 100 to 350 K is shown in the inset of Fig. 50. The rapid drop in M_s on raising the temperature is suggestive of approaching T_{com} .

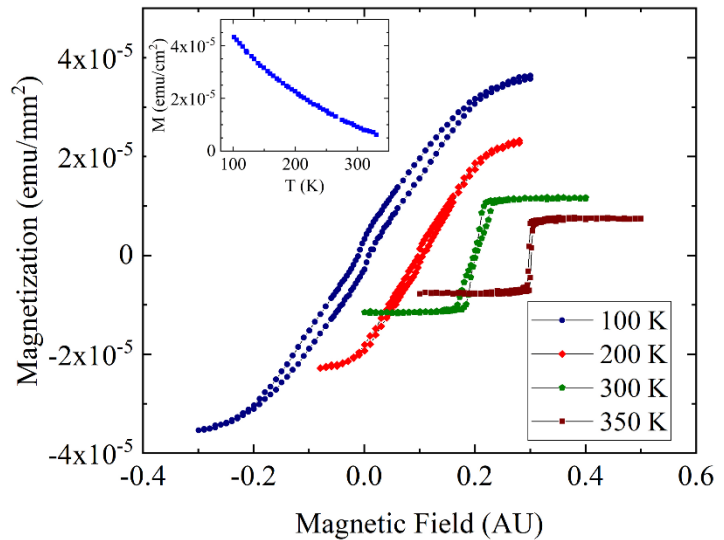


Fig.50: Out-of-plane magnetization loops of $[\text{Ho}/\text{FeCoGd}/\text{W}]_n$ multilayer measured at $T = 100, 200, 300$ and 350 K . The 200, 300 and 350 K loops have been given a rigid shift 0.1, 0.2 and 0.3 tesla, respectively for clarity. Inset shows variation of saturation magnetization with temperature ($1 \text{ emu}/\text{cm}^2 = 10 \text{ A}$).

Since the Hall resistivity (ρ_{xy}) of a magnetically ordered, material is connected to the state of its magnetization [99], we have measured the ρ_{xy} of the $[\text{Ho}/\text{FeCoGd}/\beta\text{-W}]_{\times 10}$ multilayers over a broad range of temperature and strength of the external magnetic field. The measurements of $\rho_{xy}(H)$ loops, which will be discussed in the subsequent section show that the Hall resistivity reaches saturation at fields ≤ 2 T at all temperatures between 2 to 300 K. Fig. 51(a) shows the magnitude ($|\rho_{xy}|$) of the Hall resistivity at 2.0, 4.0 and 8.0 T over the temperature range of 2 to 300 K. While the $|\rho_{xy}|$ decreases on raising the temperature, the sign of ρ_{xy} remains negative over the entire temperature range. Here the sign of ρ_{xy} needs to be viewed in the light of the Hall resistivities of the constituent RE and TM elements, which is negative for Gd and Ho and positive for Fe and Co over the temperature range investigated here. Since the magnetization of the TM sublattice is antiparallel to the direction of the external field, the Hall voltage due to the TM sublattice will also have a negative sign. Therefore, with the data in hand, it is not possible to say which sublattice contributes the most to the value of ρ_{xy} . Clearly, any deviation from collinearity of two sublattices, as happens at a spin – flop transition, or due to the formation of nontrivial spin textures, will be picked up in the measurement of ρ_{xy} . An interesting feature of the data shown in Fig. 51(a) is the appearance of a kink in $\rho_{xy}(T)$ in the vicinity of 130 K. This feature is sharply defined in the 2 T data but becomes broader on increasing the field. While discussing the $\rho_{xy}(T)$ data of the multilayers we also need to consider the contributions of the W and Ho layers imbedded in the multilayer to the Hall coefficient. Tungsten is non-magnetic and hence its Hall coefficient is set only by the Lorentz force of magnetic field on free carriers. Since the carrier density in tungsten is high ($10^{22}/\text{cm}^3$), the Hall resistivity due to free electrons is negligible compared to the AHE of Fe and Co. However, the 4f moments of holmium order into a chiral antiferromagnetic state below 130 K through a conduction electron mediated interaction. The AFM order changes into an FM state at $T = 20$ K. We believe that the kink in $\rho_{xy}(T)$ in the vicinity of 130 K is due to the AFM ordering of the Ho layers. Interestingly, the longitudinal resistivity $\rho_{xx}(T)$ shown in Fig. 51(b), is insensitive to this AFM ordering. We also note that while the ρ_{xy} change by about 50 % on raising the temperature from 2 to 300 K, the corresponding change in ρ_{xx} is only 1.5%. Further, Fig 51(b) shown that the out-of-plane magnetic field has virtually no effect on the ρ_{xx} . From these observations, it is easy to understand why the AFM order in Ho layers does not reflect in the measurements of ρ_{xx} . We note from Fig. 51(a) that the saturation value of ρ_{xy} drops monotonically with the increasing temperature for $T > T_N$. While the out-of-plane $M(T)$ also shows a similar behavior (Fig. 50(inset)), the slopes of the two curves are different, implying that the ρ_{xy} does not scale with the magnetization.

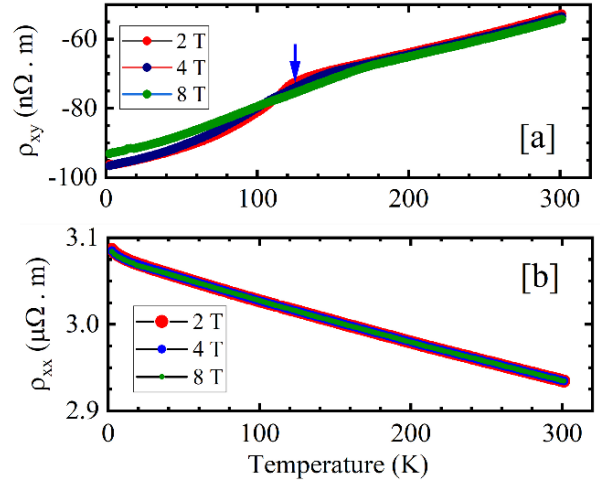


Fig. 51: (a) The Hall resistivity of the multilayer as a function of temperature from 2 to 300 K measured at 2, 4 and 8 T magnetic applied perpendicular to film plane. (b) The longitudinal resistivity as a function of temperature.

We address the possible correlation between $\rho_{xx}(H)$ and $\rho_{xy}(H)$ by measuring these resistivities at several temperatures in the field range of 0 to ± 1 T. Fig. 52 shows the loops of magnetoresistance $\{(\rho_{xx}(H) - \rho_{xx}(0))/\rho_{xx}(0) \times 100\}$ and $\rho_{xy}(H)$ at 10, 100, 200 and 300 K. The magnetoresistance of these high resistivity amorphous alloys is very small ($< 0.1\%$). Further, while the MR in metallic ferromagnets is negative due to suppression of spin-dependent scattering [99], the positive MR seen here is suggestive of interface dominated scattering as seen in multilayers of 3d ferromagnets [100]. At $T < 200$ K, the $\rho_{xy}(H)$ loops show distinct irreversibility at the lowest fields but the saturation fields for MR and ρ_{xy} are nearly the same. We also note that at the lowest temperature (10 K), while the ρ_{xy} reaches saturation, the MR starts

dropping beyond the saturation field. This is perhaps due to the suppression of spin-wave excitations at the higher field. At higher temperatures (> 200 K), we also notice that the way the magnetization and ρ_{xy} approach saturation are different. This is clearly seen in the 300 K data plotted in Fig. 52(b). Here the ρ_{xy} first surpasses the saturation value and then flattens off at high fields. The magnetization on the other hand remains well below saturation in the regime of field where ρ_{xy} displays the cusp-like behavior.

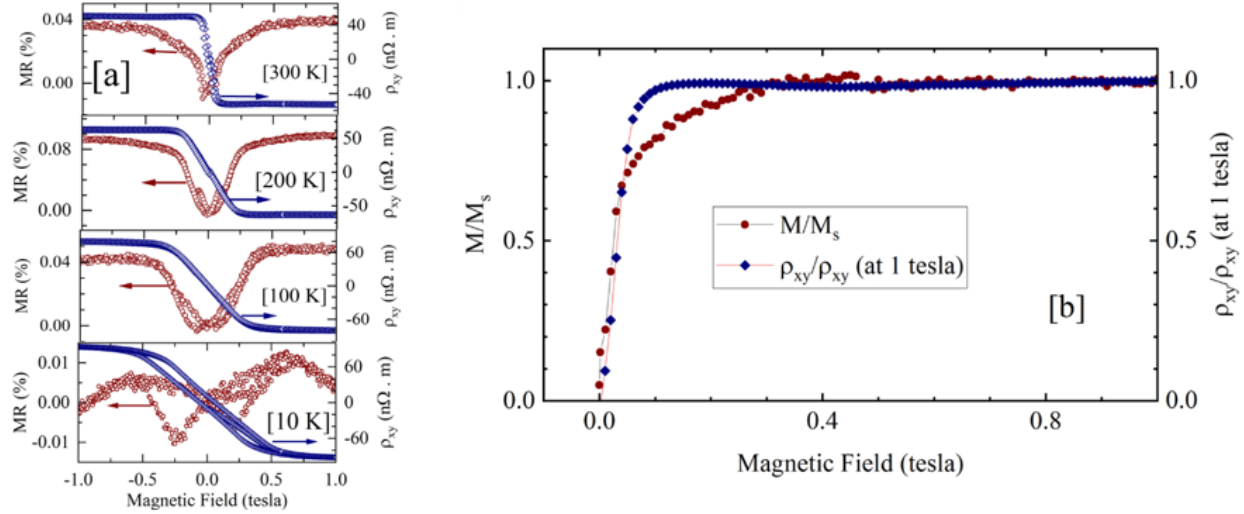


Fig. 52: (a) Loops of the out-of-plane field magnetoresistance (MR %) and anomalous Hall resistivity (ρ_{xy}) at 10, 100, 200 and 300 K. (b) The variations of magnetization and Hall resistivity normalized to their value at 1 T are plotted as a function of magnetic field. These measurements were performed at 300 K.

We address the field dependence of MR and ρ_{xx} further by measuring virgin curves in the field range of zero to 8 T. Fig. 53 shows these data taken at 300 K. The MR rises quickly from zero to 0.35% as the field is increased to 0.7 T, and beyond this field, the MR still increases but at a much slower rate (0.0025%/T). The field dependence of ρ_{xy} has a distinctive cusp (indicated by a dark arrow) in the regime where the MR is approaching the state of a constant slope. To address the origin of this cusp, we first show, in the inset of Fig. 53, the variation of the saturation value of MR and ρ_{xy} as a function of temperature. We note that while both MR and ρ_{xy} change nearly linearly with temperature in the range of 10 to 200 K, this correlation between MR and ρ_{xy} is lost at higher temperatures.

For ferromagnetic elements or alloys of a single collinear magnetic sublattice, the ρ_{xy} is expressed as [101]

$$\rho_{xy} = R_0 \mu_0 H + \mu_0 R_s M_s \quad (15),$$

where R_0 is the ordinary Hall coefficient arising from the Lorentz force on mobile charges. In the Drude model of free carrier conduction, $R_0 = 1/ne$ where e is the electronic charge and n is the carrier density. The second term in Eq. 15 contains the effect of spontaneous magnetization on scattering and group velocity of charge carriers. The anomalous Hall coefficient R_s is related to the longitudinal resistivity as $R_s = \alpha \rho_{xx} + \beta \rho_{xx}^2$, where the coefficient α and β correspond to skew scattering and the effects of band structure,

respectively. Further, it is also argued that the coefficient β draws some weightage from the side-jump scattering of charge carriers [66]. Eq. 15 suggests that the ρ_{xy}^{AHE} ($= \mu_0 R_s M_s$) should scale with the magnetization and eventually reach saturation when the system is fully magnetized. Furthermore, in the saturated state of magnetization, the ρ_{xy}^{AHE} is expected to scale with the longitudinal resistivity with a power of one, or two or some intermediate value depending on the strengths of α and β . However, this simple relation between ρ_{xy} , ρ_{xx} , and M_s of a collinear ferromagnet may not apply in the present case where two antiferromagnetically coupled magnetic sublattices compete. Moreover, the DMI at the interfaces of FeCoGd layers with Ho and β -W may make a topological contribution to the Hall resistivity as seen in the case of Pt/Co/Ir and related multilayers of 3d ferromagnets with 5d metals of large spin-orbit coupling [102]. We rewrite Eq.15 for the multilayer of W/FeCoGd/Ho as

$$\rho_{xy} = R_0 \mu_0 H + \mu_0 R_s^{\text{TM}} M_s^{\text{TM}} + \mu_0 R_s^{\text{RE}} M_s^{\text{RE}} + \rho_{xy}^{\text{THE}} \quad \text{--- (16),}$$

where the second and third terms respectively are the contributions of the TM and RE sublattices to anomalous Hall. The first term in Eq. 16 is linear in the field and the second two terms are constant above magnetic saturation due to the negligible (≤ 0.0025 %/Tesla) magnetoresistance above H_s . The last term in Eq. 14 is the Hall resistivity arising from the DMI driven spin textures.

Based on our measurements and the data reported in the literature we conclude that the sign of the anomalous Hall coefficient of Gd is negative and for FeCo it is positive. The negative sign of R_s^{RE} in the temperature range investigated here (2 to 350 K) then indicates that the magnetization of the RE sublattice is parallel to the external field. The oppositely oriented TM sublattice also makes a negative contribution to R_s . From this argument, it is also clear that any deviations from the collinearity of two sublattices would always reduce ρ_{xy} as maximum in the magnitude of ρ_{xy} occurs only when the TM and RE sublattices are antiparallel. However, as clear from the ρ_{xy} (H) data of Fig. 53, there is a robust excess ρ_{xy} which appears as a cusp just before the saturation field is reached. We could, in principle, extract this excess Hall resistivity by noting that the first term in Eq. 16 is linear in the field and the second two terms are constant above magnetic saturation due to the negligible (≤ 0.0025 %/Tesla) magnetoresistance above H_s . For a single sublattice, the AHE below H_s has been calculated from the scaling relation between ρ_{xy} and ρ_{xx} [101]. It can be expressed as

$$\rho_{xy}^{\text{AHE}} = [\alpha \rho_{xx} + \beta \rho_{xx}^2] \cdot M, \quad \text{--- (17)}$$

where α and β are scaling coefficients [101].

However, the extraction of anomalous Hall in the field range 0 to H_s is rather challenging for a two-sublattice system. Moreover, for this AFM-coupled two-sublattice system no scaling between ρ_{xy} and ρ_{xx} above H_s is seen at $T > 200$ K where the cups in ρ_{xy} appear. Furthermore, the correct analysis of the data in terms of Eq. 16 would need two such equations involving ρ_{xy} , ρ_{xx} and M of each sublattice. Here we use a simple extrapolation approach to estimate the excess ρ_{xy} . In the inset of Fig. 54, the field-linear ρ_{xy} above H_s has been extrapolated to zero field. We extract the excess ρ_{xy} by subtracting the measured ρ_{xy} from its

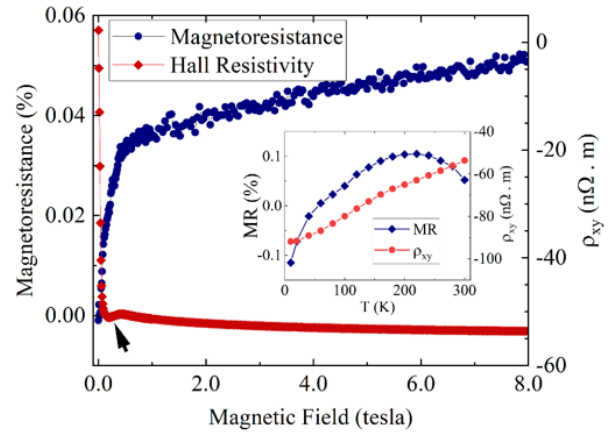


Fig. 53: The out-of-plane Magnetoresistance and anomalous Hall resistivity of the multilayer at 300 K plotted as a function of magnetic field. The black arrow at the bottom left corner of the figure highlights the cusp in ρ_{xy} . Inset shows variation of MR (%) and ρ_{xy} at 9 Tesla as function of temperature.

extrapolated value. The results of this extraction for the data taken from 200 to 300 K are shown in the main panel of Fig. 54. As we note in the figure, the excess ρ_{xy} increases at higher temperatures where the system is approaching magnetic compensation.

The excess Hall resistivity below magnetic saturation in these multilayers is likely to have a topological origin. The XMCD image of Fig. 49 shows a stripe domain pattern at zero field for Gd at room temperature. The literature on thin films of TM-RE alloys reveals that such stripes convert into bubble-like excitations on approaching magnetic saturation [103]. The opposite signs of the spin Hall angle in β -W and Ho layers, which periodically sandwich the FeCoGd layers are expected to induce an additive DMI which stabilizes topological spin excitations like skyrmions [102]. There is ample evidence of skyrmion-like spin textures in TM-RE alloys multilayered with non-magnetic heavy metals like Pt and Ta [102]. The excess Hall resistivity that we see here may arise as the conduction electrons traversing the magnetic textures hosting spin chirality acquire a Berry phase which creates a fictitious magnetic field. The Lorentz force experience by the conduction electrons under this magnetic field of Berry phase origin would make a distinct contribution to ρ_{xy} .

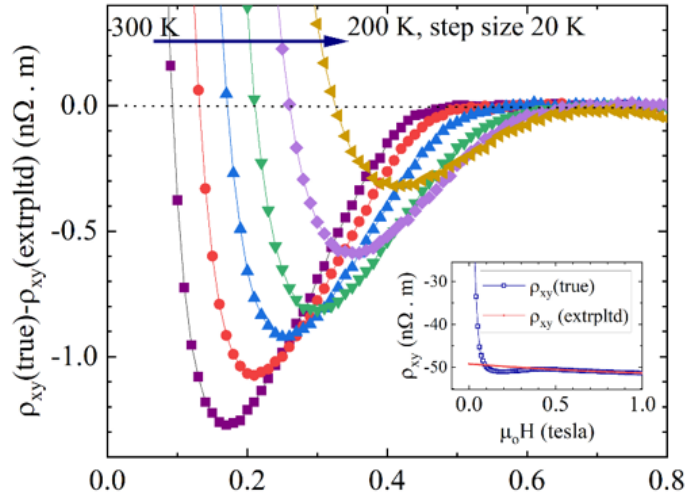


Fig. 54: The evolution of the topological Hall resistivity with magnetic field at several temperatures from 200 to 300 K. Inset shows the ρ_{xy} data at 300 K over a limited range of field to highlight the procedure used to extract the data shown in the main figure.

The DMI driven spin chirality has been addressed earlier in Pt/Gd₄₄Co₅₆/TaO₂ heterostructures [104] and Pt/Gd₂₅Fe_{65.6}Co_{9.4}/MgO multilayers [105]. In the former case, skyrmion of diameter ~ 10 nm are nucleated by a charge current of high density (10^7 A/cm²) as it flows down the narrow channels of the trilayer. In the multilayer [90], skyrmions of a diameter of 180 nm are seen close to magnetic saturation where the spiral domain textures present in the fully demagnetized state disappear completely. Although to the best of our knowledge there is no Hall data available for DMI stabilized skyrmions in RE-TM alloys, we believe the robust ρ_{xy}^{THE} seen here is the result of the additive DMI produced by two heavy metals of opposite Hall angle. It is important to point out here that skyrmions also make an additional contribution to the Hall voltage as they move under the action of a large transport current. However, the current density required for depinning skyrmions is much larger (10^7 A/m²) compared to the J_c used in our experiments.

In summary, in this section we have addressed the magnetic state, robustness of atomic periodicity, and magneto-transport in a magnetic multilayer where each constituent magnetic layer consists of antiferromagnetically coupled 3d and 4f spin sublattices. This elementary magnetic unit of the multilayer is sandwiched between thin films of β -tungsten and holmium, which possess spin Hall angles of opposite polarity and thus subject the magnetic layer to additive DMI. The zero-field ambient-temperature XPEEM images of magnetic domains reveal a sub-micron scale filamentary texture of opposite polarity indicating the zero-remanence state of an out-of-plane magnetic anisotropy film. Although the 4f spins in bulk holmium are not ordered at ambient temperature, the PNR data suggest an antiferromagnetic coupling between the 4f moments of the holmium layers and the net magnetization of the elementary magnetic unit. The measurements of longitudinal and Hall resistivities together with static magnetization over a wide range of temperature and magnetic field strength reveal some interesting aspects of the competition between 3d and 4f sublattices and in the influence of the additive interfacial DMI on this competition. The prominent feature of this competition is a cusp in $\rho_{xy}(H)$ which appears just below magnetic saturation and grows in

strength on approaching the compensation temperature of the 3d-4f magnetic unit. A simple analysis of the anomalous Hall resistivity of our two-sublattice magnetic films where each sublattice has opposite Hall angle together and a topological contribution resulting from interfacial DMI sheds light on the origin of this cusp.

C. Collaborations

C.1. Collaboration with AFRL – WPAFB Materials Directorate

We continue our collaboration with the magnetism group at AFRL-WPAFB led by Dr. Michael Page. Recently we supplied a set of sputter-deposited thin films of permalloy to support their research on acoustically driven ferromagnetic resonance devices. We have also launched a new initiative with them to study magnetism in thin films of chiral antiferromagnets like Mn_3Sn . A set of Mn_3Sn and $\text{Mn}_3\text{Sn}/\text{FeCoB}/\text{Nb}$ heterostructures was sent to them for high resolution X-ray diffraction and magnetic force microscopy studies. A new approach has been used these films which involves co-sputtering of Mn and Sn instead of sputtering an alloy target. Further, to get better uniformity, these films are on (111) SrTiO_3 and (100) MgO with a 5 nm thick ruthenium seeding layer. We are currently measuring electron transport and ISHE/FMR in these films at different temperatures. It is hoped that this work will lead to an excellent joint publication. We plan to continue this collaboration to address bias-field-free switching of magnetization in perpendicular magnetic anisotropy multilayers deposited on Mn_3Sn and imaging of magnetic textures in such heterostructures with Lorentz microscopy and magnetic force microscopy measurements.

C.2 Collaboration with Johns Hopkins University

We are collaborating with Prof. Peter Armitage and his group at Johns Hopkins physics department to address terahertz electrodynamics of $\text{Py}/\text{Nb}/\text{Py}$ and $\text{Mn}_3\text{Sn}/\text{Py}/\text{Nb}/\text{Py}$ superconducting spin valves, in addition to studies of spin orbit torque terahertz emitters. We have completed the studies of complex conductivity of superconducting beta-tungsten films. These studies have yielded a wealth of information on the origin of superconductivity in this non-equilibrium phase of tungsten.

C.3 Collaboration with University of Delaware

We have been working closely with Prof. Benjamin Jungfleisch at the Physics Department - University of Delaware on the studies spin orbit torque driven terahertz emission in FeGaB/BiSb and $\text{FeGaB}/\text{Ag}/\text{BiSb}$ heterostructures. This collaboration has yielded two publications. We are currently finalizing one more manuscript on terahertz emission from $\text{FeCo}/\text{Bi}_2\text{Te}_3$ bilayers.

C.4 Collaboration with NIST Neutron Research Center

We have conducted polarized neutron reflectivity experiments in collaboration with Dr. Brian Kirby of NIST on multilayers of $\text{Ho}/\text{FeCoGd}/\text{B-W}$ where the Ho and b-W layers impart additive DMI exchange on the ferrimagnet FeCoGd . The results of this study have been published in Physical Review B. We will continue this collaboration in future to address magnetic interactions in chiral antiferromagnets like Mn_3Sn and Mn_3Ge .

C.5 Collaboration with Center for Functional Nanomaterials (CFN) – Brookhaven Laboratory

We have successfully completed two sets of X-ray photoelectron microscopy experiments at the National Synchrotron Light Source in collaboration with Dr. Jerzy Sadowski of CFN-BNL. The first set of experiments was performed on $\text{Ho}/\text{FeCoGd}/\text{B-W}$ multilayers and the second one is on thin films of $\text{Co}_x\text{Ho}_{1-x}$ thin films.

D. STUDENT PARTICIPATION AND TRAINING

Engagement of graduate and undergraduate students in research is an important component of teaching and training. Three undergraduate students, Deandre McAlmont, Dorrian Garry-Stokes and Naeem Brown have received training in pulsed laser ablation, sputtering and on FMR studies of magnetic materials. A Master's student, Sean Davis has completed his M.S. thesis on synthesis and characterization of FeCoB thin films. The department of Physics at Morgan State also participated in an interdisciplinary Ph. D. program on bioenvironmental sciences. Two graduate students of this program, Prabesh Bajracharya and Rajeev Nepal, are working in the lab for their Ph D thesis. While Prabesh is engaged in FeGaB/BiSb interfaces using the composite PLD/sputtering system, Rajeev uses the multitarget sputtering machine to deposit FeCo films and FeCo/ β -W heterointerfaces. Recently we have inducted Harshirat Kaur, an MS student in this project. She is studying room – temperature FMR and ISHE in FeCoB/Bi₂Te₃ and FeCoB/Mn₃Sn bilayer systems.

ENGAGEMENT OF POST DOCTORAL RESEARCHER

Dr. Vinay Sharma has worked as a postdoctoral associate on this project. He has gained extensive experience in setting up of cryogenic FMR systems along with studies of spin pumping in a variety of ferromagnet/topological insulator bilayers.

E. PUBLICATIONS, PRESENTATIONS, and INVITED TALKS

E1. Journal Articles

1. Vinay Sharma, Rajeev Nepal, Weipeng Wu, E. A. Pogue, Ravinder Kumar, Rajeswari Kologani, Lars Gundlach, M. B. Jungfleisch and Ramesh C Budhani, Comparing spin injection in Fe₇₅Co₂₅/Bi₂Te₃ at GHz and optical frequencies, **Appl. Phys. Lett.** **122**, 072403 (2023). <https://doi.org/10.1063/5.0132617>
2. Sharma, Vinay, et al. Planar Hall effect and Anisotropic Magnetoresistance in Thin Films of Chiral Antiferromagnet Mn₃Sn, arXiv:2307.07795v1, [khhwllkhttps://doi.org/10.48550/arXiv.2307.07795](https://doi.org/10.48550/arXiv.2307.07795)
3. Vinay Sharma, Prabesh Bajracharya, Anthony Johnson and Ramesh C Budhani, “Interface-driven spin pumping and inverse Rashba-Edelstein effect in FeGaB/Ag/BiSb Multilayers”, **AIP Advances** **12**, 035028 (2022). <https://doi.org/10.1063/9.0000311>
4. Rajeev Nepal, Vinay Sharma, Lisa Pogue, Natalia Drichko, Ramesh C. Budhani; Disorder driven variations in magnetoresistance and planar Hall effect in Bi₂Te₃ thin films, **Thin Solid Films** **761**, 139520(2022). DOI: [10.1016/j.tsf.2022.139520](https://doi.org/10.1016/j.tsf.2022.139520)
5. Prashant Chauhan, Ramesh Budhani, and N. P. Armitage, “Anomalously small superconducting gap in a strong spin-orbit coupled superconductor: β -tungsten”, **Phys. Rev. B** **105**, L060503 (2022). <https://doi.org/10.1103/PhysRevB.105.L060503>
6. Ramesh C. Budhani, Vinay Sharma, Ezana Negusse, Jacob Casey, Arjun K. Pathak, Jerzy T. Sadowski, and Brian Kirby, “Magnetotransport and magnetic textures in Ho/FeCoGd/ β -W multilayers”, **Phys. Rev. B.** **105**, 024412 (2022). <https://doi.org/10.1103/PhysRevB.105.024412>
7. P. Bajracharya, V. Sharma, A. Johnson and R. C. Budhani, “Resonant precession of magnetization and precession – induced DC voltages in FeGaB thin films”, **J. Phys. D Appl. Phys.** **55**, 75303 (2021). [https://DOI 10.1088/1361-6463/ac34ab](https://doi.org/10.1088/1361-6463/ac34ab)
8. Vinay Sharma, Weipeng Wu, Prabesh Bajracharya, Duy Quang To, Anthony Johnson, Anderson Janotti, Garnett W. Bryant, Lars Gundlach, M. Benjamin Jungfleisch, and Ramesh C. Budhani,

- “Light and microwave driven spin pumping across FeGaB–BiSb interface”, **Phys. Rev. Mater.** **5**, **124410** (2021). <https://doi.org/10.1103/PhysRevMaterials.5.124410>
9. Ramesh C. Budhani, Joshua S. Higgins, Deandre McAlmont, and Johnpierre Paglione; *Planar Hall effect in c-axis textured films of Bi₈₅Sb₁₅ topological insulator*, **AIP Advances** **11**, **055020** (2021). <https://doi.org/10.1063/5.0049577>
 10. Anthony Johnson, Ezana Negusse, Vinay Sharma, Dan Anyumba, Deandre McAlmont, and Ramesh C. Budhani, “Sublattice magnetization driven anomalous Hall resistance of FeCoGd amorphous films” **AIP Advances** **10**, **115214** (2020); <https://doi.org/10.1063/5.0026614> (Appeared on Cover Page of AIP advanced).
 11. Derek A. Bas, Piyush J. Shah, Alexei Matyushov, Maksym Popov, Viktor Schell, Ramesh C. Budhani, Gopalan Srinivasan, Eckhard Quandt, Nian Sun, and Michael R. Page, “Acoustically Driven Ferromagnetic Resonance in Diverse Ferromagnetic Thin Films”. **IEEE Trans. Mag.** DOI: [10.1109/TMAG.2020.3019214](https://doi.org/10.1109/TMAG.2020.3019214)
 12. Dereje Seifu, Asha J. Hall, David Elbert, Patricia McGuiggan, Oliver J. Myers, and Ramesh C. Budhani “Magnetostrictive particulates of Tb_{0.3}Dy_{0.7}Fe₂ integrated into carbon fiber reinforced polymer for structural damage monitoring” **AIP Advances** **2020** <https://doi.org/10.1063/5.0013282>

E3. Conference Presentations

1. Ralph Romero, Ramesh C Budhani and Norman P Armitage. Abstract: P38.00011: Terahertz Emission from Inverse Spin Hall Effect in β -W 5:00 PM–5:12 PM Live March Meeting APS 2021.
2. Vinay Sharma, Weipeng Wu, Prabesh Bajracharya, Quang To, Anthony Johnson, Anderson Janotti, Garnett Bryant, Lars Gundlach, Matthias Benjamin Jungfleisch, Ramesh Budhani; “Investigation of spin pumping across FeGaB–BiSb interface at two different time scales” APS March Meeting 2022 Chicago-Oral presentation # Y55.00004.
3. Prabesh Bajracharya, Vinay Sharma, Weipeng Wu, Anthony Johnson, Matthias Benjamin Jungfleisch, Ramesh Budhani; “Light and microwave driven spin current detection in FeGaB thin films”, APS March Meeting 2022 Chicago- Poster Presentation # T00.00208).
4. Vinay Sharma, Ramesh Budhani, Ezana Negusse, Jacob Casey, Arjun Pathak, Jerzy Sadowski, Brian Kirby; “Magneto-transport and magnetic textures in Ho/FeCoGd/ β -W multilayers with additive chiral exchange”, APS March Meeting 2022 Chicago Oral Presentation #Z54.00014.
5. Rajeev Nepal, Vinay Sharma, Weipeng Wu, Anthony Johnson, Matthias Benjamin Jungfleisch, Ramesh Budhani; “Concurrent observation of robust anti-damping torque and inverse spin Hall effect in Fe₇₅Co₂₅/Bi₂Te₃ multilayers”, APS March Meeting 2022 Chicago, Oral Presentation #T52.00005.
6. Vinay Sharma, Rajeev Nepal, Harsimrat Kaur, Ramesh C. Budhani “Spin-to-charge conversion at cryogenic temperatures in the chiral antiferromagnet Mn₃Sn interfaced with a metallic ferromagnet”. To be presented at the IEEE Magnetism Conference 2022.

E4. Invited talks

1. Ramesh C Budhani – Winter school on quantum materials at the University of Maryland College Park on 01-17-2023. Title – “Thin film growth of chalcogenide-based 2d compounds”.
2. Ramesh C Budhani – CEC/ICMC-23 International conference Honolulu, Hawaii on 07-10-2023. Title “Spin Chirality Driven Magneto - Transport in Mn₃Sn and Co₃Sn₂S₂ Kagome Magnets”

F. Acknowledgments

We thank Dr. Kenneth Goretta, Program manager AFOSR for his prompt support on the operational aspects of this grant. Our thanks are also due to our collaborators Dr. Michael Page and his team at AFRL, Dr. Peter Armitage at JHU, Dr. Paglione at UMD, Dr. Jungfleisch at UD, Dr. Brian Kirby at NIST, and Dr. Jerzy Sadowski at BNL for providing their laboratory facilities and intellectual input whenever needed for this project. We also thank the administrative staff of Morgan State for the timely processing of the required paperwork for successful implementation of this project.

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