Enhanced spin-orbit torque (SOT) and spin accumulation quantification in perpendicularly magnetised systems

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2020

Wong, Q. Y. (2020). Enhanced spin-orbit torque (SOT) and spin accumulation quantification in perpendicularly magnetised systems. Doctoral thesis, Nanyang Technological University, Singapore.

<https://hdl.handle.net/10356/143337>

<https://doi.org/10.32657/10356/143337>

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Enhanced Spin-Orbit Torque (SOT) and Spin
accumulation quantification in perpendicularly
magnetised systems

WONG QI YING

SCHOOL OF PHYSICAL AND MATHEMATICAL SCIENCES

2020

Enhanced Spin-Orbit Torque (SOT) and Spin
accumulation quantification in perpendicularly
magnetised systems

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School of Physical and Mathematical Sciences

A thesis submitted to the Nanyang Technological University in
partial fulfilment of the requirement of the degree of
Doctor of Philosophy

2020

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Authorship Attribution Statement

This thesis contains material from two papers published and one ongoing work in the following peer-reviewed journals where I was the first and/or corresponding author.

Chapter 3 is published as Q. Y. Wong, W.L. Gan, F.L. Luo, G.J. Lim, C.C.I. Ang, F.N. Tan, W.C. Law, W.S. Lew*, "*In-Situ* Kerr harmonic measurement in determining current-induced effective fields in MgO/CoFeB/Ta", *Journal of Physics D: Applied Physics*, 51, 115004 (2018). DOI: [10.1088/1361-6463/aaaedb](https://doi.org/10.1088/1361-6463/aaaedb)

The contributions of the co-authors are as follows:

- I performed all the harmonic Hall measurements with the Kerr microscope and characterised the film structure. I did all analysis, and the manuscript was prepared by me, with inputs from W.L. Gan and F.L. Luo.
- W.L. Gan and C.C.I. Ang assisted in the Kerr microscopy measurements and discussed the characterisation of DMI.
- G.J. Lim, F.N. Tan, and W.C. Law assisted in the deposition and VSM characterisation of the film stacks.
- F.L. Luo discussed the derivations of the analytical fitting equation
- Prof. Lew supervised and engaged in discussion for the entire paper.

Chapter 4 is published as Q. Y. Wong, C. Murapaka, W. C. Law, W. L. Gan, G. J. Lim, and W. S. Lew, Enhanced Spin-orbit Torques in Rare Earth Pt/[CoNi]₂/Co/Tb systems, *Physical Review Applied* 11, 024057 (2019) DOI: [10.1103/PhysRevApplied.11.024057](https://doi.org/10.1103/PhysRevApplied.11.024057)

The contributions of the co-authors are as follows:

- I performed all the harmonic Hall and current-induced switching measurements and characterised the film structure using VSM. I did all analysis, derived the analytical expressions for a modified H_z biased harmonic Hall measurement, and the manuscript was prepared by me and results were thoroughly discussed with C. Murapaka and W. C. Law.
- W.C Law and G.J. Lim assisted in the thin film deposition.
- W. L. Gan assisted in the usage of the Kerr microscopy images.
- Prof. Lew supervised and engaged in discussion for the entire paper.

Chapter 6 consists of ongoing work that will be presented as Q. Y. Wong, F. L. Luo, W. C. Law, W. L. Gan, H.Y. Poh, G.J. Lim and W. S. Lew, Direct current biased harmonic probing of Spin Accumulation Magnetoresistance in Rare Earth Systems.

The contributions of the co-authors are as follows:

- I derived an analytical expression for the experimental quantification of spin accumulation using a dc biased harmonic Hall technique, performed all the harmonic Hall measurements. The analysis was discussed with F. L Luo, and I prepared the manuscript, and the results were discussed with the rest.
- W.C Law and G. J. Lim assisted in the thin film deposition.
- W. L. Gan and H. Y. Poh assisted with the MagVision Kerr microscope and macrospin simulations.
- Prof. Lew supervised and engaged in discussion for the work.

10 January 2020

Date

Wong Qi Ying

Abstract

The ever increasing demand for high-speed data transfer and data processing have pushed the field of electronics toward spintronics, making the utilisation of electron spin in devices a reality. Spin-orbit torque (SOT) is a current-induced phenomenon which manipulates magnetisation via momentum transfer from accumulated spins at the heavy metal (HM)/ferromagnet (FM) interface. SOT strength is related to the anti-damping like (DL) and fieldlike (FL) effective fields. Due to its potential for faster switching and lower energy consumption, the pursuit of SOT physics becomes imperative. In this thesis, systematic investigations of SOT physics were carried out by evaluating the SOT effective fields, *i.e.* DL term and FL term, through a harmonic Hall measurement technique, in perpendicularly magnetised single FM (CoFeB) and multilayer (Co/Ni) structures. A combination of *in-situ* Kerr and harmonic measurement enabled the evaluation of the co-existing SOT fields while imaging the magnetisation behaviour during the measurement. Observation of dendritic-like domains indicates the influence of the interfacial Dzyaloshinskii–Moriya interaction (DMI) at the CoFeB/Ta interface as affirmed by micromagnetic simulations. In the pursuit of alternative spin Hall materials, rare-earth metal Tb in Pt/[Co/Ni]₂/Co/Tb multilayer structures with perpendicular magnetic anisotropy (PMA) have demonstrated enhanced SOT. Due to the large spin-orbit coupling in Tb, the effective damping like efficiency, ξ_{DL}^{eff} , is determined to be 0.55 for 9-nm Tb as compared to $\xi_{DL}^{eff} = 0.18$ for the reference Pt/[Co/Ni]₂/Co/Ta stack. Enhanced magnetisation switching efficiency is observed with increasing Tb thickness affirming the sizable anti-damping torque responsible for deterministic switching. Furthermore, due to a more substantial interfacial coupling in Co/Tb interface than in Co/Ta interface, strong angular dependence of the damping-like and field-like terms is present but diminishes with increasing Tb thickness. These results affirm that Tb plays a significant role in tuning the SOT efficiency in these structures as the increased Tb concentration leads to an enhancement of the effective spin Hall angle and switching efficiency. In a further investigation, an anomalous second harmonic Hall resistance was induced when a biasing current was applied to the structures. The analytical derivations and experimental results reveal an anomalous second Hall resistance originating from the spin accumulation. This measured anomalous second harmonic Hall resistance enables the adiabatic quantification of the spin accumulation in the structures. Spin accumulation ranges from ~3.0% in $t_{Tb} = 5\text{nm}$ to ~6.6% in $t_{Tb} = 8\text{ nm}$, and ~1.3% in the reference stack of $t_{Ta} = 5\text{nm}$ of the local magnetisation recorded when the total applied current density is 10^{11} A/m^2 .

Acknowledgements

This thesis is a testament to God's grace and favour upon my life. It would not have been possible to complete my doctoral studies, if not for the God-sent people who aided me in my journey throughout my studies.

Firstly, I would like to thank Professor Lew Wen Siang and my previous co-supervisor, Dr Danny Shum, for providing me with the opportunity for the scholarship which funded the duration of my doctoral studies. Prof Lew provided guidance and support throughout my research, especially so in the area of academic writing, which have helped me in presenting my work more concisely and scientifically. At the same time, allowing flexibility and freedom in the pursuit of the research topic and is always approachable for in-depth manuscript and research discussions.

Secondly, I am thankful to the NTU Spintronics Device group members for availing yourselves sacrificially at times to teach me the crux of experimental and analytical research. During the start of my candidature, Dr Kwon Jae Suk taught me how to use the electron beam lithography equipment and some experimental work on the harmonic Hall measurement. Gerard Joseph Lim taught me all the intricate details of device fabrication from UV lithography, to sputtering and ion milling, and LABVIEW programming at the latter stages, as well as discuss research ideas. Dr Tan Funan and Dr Law Wai Cheung helped me significantly with the deposition of thin films so that I could move on more quickly to device patterning and measurements. Dr Luo Feilong and Dr Li Sihua availed themselves to teach me on the essential things to note when conducting SOT characterisation measurements which benefited me a lot during my work. Dr Gan Wei Liang has helped me immensely with his extensive knowledge on magnetism and magnetic materials as well by proposing possible solutions and brainstormed with me when I am stuck and together with Calvin Ang Ching Ian taught me to use the MagVision Kerr microscope which allowed us to conduct *in-situ* harmonic measurements as well as to characterise DMI on a tricky CoFeB based sample. Dr

Chandrasekhar Murapaka for his expertise on spintronics and magnetism, which has greatly benefited me. Putu Andhita has been of great help even though his main research area is not in spintronics, with his strong mathematical and physics background, he has not only provided fresh perspectives on my problems but also challenged me to find consistency in my analytical derivations for my work. As for the rest of the group, though they are not mentioned here, I am grateful for their invaluable support and friendship throughout this period which are even more valuable than the skills and knowledge learnt during the four years in the group.

Thirdly, I would like to present my heartfelt gratitude to my cell group for their unwavering support, understanding and care through during my entire candidature, which made the bitter journey very much sweeter. Also, I would like to express gratitude to my parents for their support all these years.

Lastly, and certainly not the least, I would like to thank my missus for standing by me all these years, from the first day when I told her that I intend to pursue my doctoral studies, she has always encouraged me whenever I view things bleakly and without hope. She embodied what Love is; she is patient, kind, does not envy, does not boast, is not proud, does not dishonour others, is not self-seeking, is not easily angered, keeps no record of wrongs. She always protects, always trusts, always hopes, and always perseveres even though how nasty that I can be when I crumble. As this part of the journey ends, a new journey awaits together with her and God. My heart is full.

List of Publications

International Refereed Journal

1. Q. Y. Wong, C. Murapaka, W.C Law, W.L. Gan, G.J. Lim, W.S. Lew*, "Enhanced Spin-orbit Torques in Rare Earth Pt/[CoNi]₂/Co/Tb systems", Physical Review Applied 11, 024057 (2019).
2. F. L. Luo, Q.Y. Wong, S.H. Li, F. N. Tan, G. J. Lim, X. Wang, and W.S. Lew*, "Dependence of spin-orbit torque effective fields on magnetization uniformity in Ta/Co/Pt structure", Scientific Reports, 9 (1), 10776 (2019)
3. F.N. Tan, Q.Y. Wong, W.L. Gan, S.H. Li, H.X. Liu, F. Poh, and W.S. Lew*, "Electric field control for energy efficient domain wall injection", Journal of Magnetism and Magnetic Materials, 485, 174 (2019).
4. S. Krishnia, P. Sethi, W. L. Gan, Q. Y. Wong, G.J. Lim, and W.S. Lew*, "Spin-orbit torque induced effective field modulation in synthetic antiferromagnetic structure," Journal of Magnetism and Magnetic Materials, 475, 327 (2019).
5. Q. Y. Wong, W.L. Gan, F.L Luo, G.J. Lim, C.C.I. Ang, F.N. Tan, W.C Law, W.S. Lew*, "*In-Situ* Kerr harmonic measurement in determining current-induced effective fields in MgO/CoFeB/Ta," Journal of Physics D: Applied Physics, 51, 115004 (2018).
6. Qi Ying Wong, Ning Liu, Cheng-Gee Koh, Hoi-Yeung Li, and Wen Siang Lew*, "Isolation of magnetically tagged cancer cell through an integrated magnetofluidic device," Microfluidics and Nanofluidics, 20:139 (2016).

Conference Presentations

1. Enhanced Spin Orbit Torques in Rare-earth based Pt/[Co/Ni]₂/Co/Tb systems
Q.Y. Wong, C. Murapaka, W.C Law, W.L. Gan, G.J. Lim, W.S. Lew *64th Annual Conference on Magnetism and Magnetic Materials (MMM 2019), Las Vegas, USA*
2. H_z Biased Field Modulation of The Harmonic Measurement in The Determination of The Effective Fields in MgO/CoFeB/W
Q.Y. Wong, G.J. Lim, W.L. Gan, W.S. Lew *62nd Annual Conference on Magnetism and Magnetic Materials (MMM 2017), Pittsburgh, USA*
3. Observation of Large Planar Hall Effect on the Current Induced Spin-orbit Effective Fields in Ta/MgO/CoFeB/Ta
Q. Y. Wong, W.L. Gan, F.L Luo, and W.S. Lew*
INTERMAG 2017, Dublin, Ireland
4. “Skyrmion in-line injection and driving by spin Hall torque.”
Weiliang Gan, Sachin Krishnia, Qi Ying Wong, Wai Cheung Law, Gerard Joseph Lim, and Wen Siang Lew
INTERMAG 2017, Dublin, Ireland
5. “Angular dependence of spin-orbital torque induced magnetisation switching in synthetic antiferromagnetic structures.”
S. Krishnia, P. Sethi, W. L. Gan, Q. Y. Wong, and W. S. Lew
INTERMAG 2017, Dublin, Ireland
6. “Spin orbital torque induced effective field modulation in synthetic antiferromagnetic structures.”
S. Krishnia, P. Sethi, W. L. Gan, Q. Y. Wong, and W. S. Lew
INTERMAG 2017, Dublin, Ireland

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Acronyms

AC	alternating current
AHE	anomalous Hall effect
AMR	anisotropic magnetoresistance
ANE	anomalous Nernst effect
CIP	current-in-plane
CMOS	complementary metal-oxide-semiconductor
CPP	current perpendicular to the plane
CPU	central processing unit
DC	direct current
DL	damping-like
DMI	Dzyaloshinskii-Moriya interaction
DRAM	dynamic random-access memory
DSP	digital signal processor
DW	domain wall
DWM	domain wall motion
EBL	electron beam lithography
FL	field-like
FM	Ferromagnetic Metal
GMR	giant magnetoresistance
HDD	hard disk drive
HM	heavy transition non-magnetic metal
IL	insulating layer
IMA	in-plane magnetic anisotropy
LLG	Landau-Lifshitz-Gilbert
MOKE	magneto-optical Kerr effect
MRAM	Magnetoresistive random access memory
mr-Rem	micro resist (technology) Remover
MTJ	magnetic tunnel junction
NM	non-magnetic metal
OHE	ordinary Hall effect
PHE	planar Hall effect
PMA	perpendicular magnetic anisotropy
PMMA	4% 950k molecular weight of polymethyl methacrylate in anisole
PVD	physical vapour deposition
RAM	random access memory
RE	rare-earth metal
RF	radio frequency

ROM	read-only memory
SL	Slonczewski-like
SNR	sound-to-noise ratio
SOC	spin-orbit coupling
SHE	spin Hall effect
SOT	spin-orbit torque
SOT-MRAM	spin-orbit torque MRAM
SRAM	static random-access memory
SSD	solid state drive
STT	spin-transfer torque
STT-MRAM	spin-transfer torque MRAM
TB	tunnel barrier
TMR	tunnelling magnetoresistance

Chapter 1: Introduction

The evolution of technology over the past decades have revolutionised modern day electronics. In particular, the invention of transistors has led to significant progress in computer memory architecture since the idea of a floating gate non-volatile memory device in 1967¹. From the first 64-bit cache memory chip in 1969² to elaborate semiconductor-based data storage systems used for handheld devices and massive supercomputers³; these advances have whetted the appetite for even better devices. In such an environment, the research direction is to push for multi-terabyte data storage with low operating power, low cost, and high density in small confinements.

The ever-increasing demand for higher computing speeds has resulted in limits of conventional memories. Conventional memories such as FLASH, Dynamic Random-Access Memories (DRAMs) and Static Random-Access Memories (SRAMs) can no longer support the high-speed data transfer and highly-efficient processing at the turn of the century, causing a latency gap quandary as exhibited in the memory hierarchy pyramid shown in Figure. 1.1

Figure 1.1 Computer Memory Hierarchy Pyramid

In the memory hierarchy shown in Figure 1.1, the top of the pyramid corresponds to the fastest, more expensive, and lower capacity memories, i.e. central processing units (CPU) register and caches memories such as the SRAMs and DRAMs. The bottom of the pyramid consists of the lower cost, slower and higher density storage memories such as the magnetic Hard Disk Drives (HDDs) and Solid-State Drives (SSDs). Nonetheless, the middle of the pyramid lies the latency gap resulting in a bottleneck in data transfer processes. Since higher memory speeds are essential in modern devices, the latency limitation leads to a pressing need for replacement memory. In general, RAMs have superiority over Read-Only Memories (ROMs) because RAMs allow for information to be retrieved randomly while the ROM retrieves it in an orderly fashion. In doing so, RAMs have significantly lower latency, smaller size, higher compactability for applications in modern-day devices.

With the increase in demand for handheld or wearable technology, the demand for fast storage and non-volatility has increased likewise⁴, leading to the scaling down of cell sizes, increasing single cell memory density and lowering the power consumption for read and write operations in RAMs. Static Random-Access Memory (SRAM) and the Dynamic Random-Access Memory (DRAM) often utilised as cache memories are volatile memories that consume more power even though they have quicker read and write access times as compared to non-volatile Flash memories. Electron and hole transport mobility are the fundamental operating principle of these devices. As such, when they are scaled-down, quantum effects such as electron tunnelling occur through the gate, and Joule heating causes device breakdown, marking the impending end to Moore's law⁵. This imminent end has sparked the scientific community to search for potential alternatives to semiconductor technology^{6,7}. In the search for suitable alternatives, another intrinsic property of the electron, known as the spin was utilised as a means for data storage through binary bits, '0' and '1'. This discovery led to a new branch of research on spin-based electronics, also known as 'spintronics'. Non-volatility, low power, high density and speed of spintronic devices have significant advantages over conventional electronics, making them suitable candidates for next-generation storage devices⁸⁻¹⁰.

1.1 Spintronics: The Way Forward

Spintronics connects the spin degree of freedom of electrons with its transport to achieve the active manipulation of magnetic moments^{11–13}. Interpretation of the spintronics field existed early through Mott's spin-dependent scattering of electrons in 1929¹⁴ and experimental magnetic tunnelling by Julliere in 1975¹⁵ and the spin injection through a normal metal by Johnson and Silsbee in 1985¹⁶. A groundbreaking discovery occurred in 1988 that changed the worldview of spintronics. Peter Grünberg¹⁷ and Albert Fert¹⁸ discovered the giant magnetoresistance (GMR) which also won them the Nobel Laureate prize for physics in 2007¹¹. GMR discovery led to the modern conception of read/write heads of hard disk drives (HDD) in 1990 by IBM based on the GMR replacing the anisotropic Magnetoresistive effect (AMR) read/write heads, leading to a catalytic spark for future spintronic research opportunities.

Figure 1.2 *Giant Magnetoresistive (GMR) effect on the spin-valve structure. Parallel alignment of two ferromagnetic layers leads to low resistance, while the anti-parallel configuration of the magnetic layers leads to high resistance. (a) Current in-plane (CIP) configuration (b) Current perpendicular to plane direction.*

The GMR effect is due to electron scattering as current passes through the spin valve structure causing changes in resistance states due to the relative magnetisation orientation in the spin valve; parallel configuration (low resistance) and anti-parallel configuration (high resistance). The read current is passed through the device, and a magnetoresistance is read, which provides information on the relative orientation of the magnetisations of the FM layers. Resistance can be read in two configurations: namely,

the current-in-plane (CIP) and the current perpendicular to plane (CPP) as shown in Figure 1.2. The GMR was soon replaced by the tunnel Magnetoresistive (TMR) effect where the non-magnetic metal (NM) was replaced with an insulating layer (IL)¹⁹⁻²¹. The trilayer structure consisting of the FM/IL/FM was then known as the magnetic tunnel junction (MTJ) are characterised by large magnetoresistances. This property was later used as a recording unit for Magnetic Random-Access Memories (MRAM)

1.2 Spin Torque-MRAM (ST-MRAM)

ST-MRAM comprises of nano-magnetic structures that can potentially replace CMOS-based memories^{8,13}. The non-volatility of the ST-MRAM enables minimisation of static power consumption and leading towards a normal-off/instant-on computing system¹⁰. The MTJ has a low resistance state when the magnetisation orientations for the two magnetic layers are parallel and has high resistance when the layers are antiparallel due to TMR effect^{18,19}.

Berger and Slonczewski first postulated that spin-polarised current could manipulate magnetisation in ferromagnets via the spin-transfer torque (STT) mechanism. As such, a sufficiently large current passing through the MTJ would enable the change in the relative magnetisation. In this STT mechanism, the storage free layer magnetisation can effectively be changed without an applied field through the utilisation of a spin-polarised current as illustrated in Figure 1.3.

Figure 1.3 *Schematic illustration of the STT mechanism*

The physics of the STT are based on the spin interaction between $4s$ and $3d$ electrons in ferromagnetic materials such as Co, Fe and Ni. $4s$ free electrons being the outermost electrons forms the spin-polarised current while the itinerant $3d$ electrons form the $3d$ band which gives rise to the local magnetisation as well as contribute to the current flow^{22–25}. From spin-dependent scattering between the $3d$ and $4s$ electrons, the $4s$ electrons are spin-polarised along the orientation of the local magnetisation. As such, when a current is incident on the fixed layer of the MTJ, the $4s$ electron spins align themselves parallel and antiparallel to the local magnetisation^{12,23,26–28}. This mechanism proposed by Slonczewski *et al.* describes the adiabatic reorientation of spins along the local magnetisation through angular momentum transfer is also known as the adiabatic component of the STT^{29,30}. In a reverse orientation, spin polarised electrons entering from the fixed layer are reflected by the free layer. This reflection gives rise to non-adiabatic STT through a linear momentum transfer. The magnetisation reversal is dependent on the critical current density, which affects the adiabatic and non-adiabatic STT. Till date, STT has been implemented in MRAMs, and the STT-MRAM have drawn great interest and research due to its non-volatility, scalability and low-read access times^{31,32}. Due to scalability issues, the earlier in-plane magnetic anisotropy (IMA) MRAMs shifted towards MRAMs with perpendicular magnetic anisotropy (PMA), which can further minimise cancellation of demagnetisation fields and increase TMR^{33–35}.

Figure 1.4. Schematic of ST-MRAM memory array with bit and word lines. (a) Two terminal STT-MRAM cell and array, (b) proposed SOT-MRAM architecture and three-terminal single-cell MTJ.

1.3 Motivation

Although the STT-MRAM works as fast as a conventional SRAM, this writing method faces several issues. Such issues include higher writing currents in the order of 10^{12} Am^{-2} , time-dependent degradation of the tunnel barrier and joule heating leading TMR reduction. The longer write latency of the STT-MRAM leads to its unsuitability for L_1 caches. The degradation of the tunnel barrier can be reduced drastically through an alternative mechanism known as the Spin-Orbit Torque (SOT). In 2012, Liu *et al.*³⁶ proposed a three-terminal MTJ that utilised SOT for switching, similar to the single SOT-MTJ shown in Figure 1.4. In this SOT-MTJ design architecture, the read and write lines are separated, and the writing current will not be required to pass through the insulating MgO tunnel barrier. This significantly overcomes the time-dependent degradation faced by its STT predecessor and thereby avoids the other TMR-related problems of the traditional tunnel junctions. Furthermore, further investigations by other groups reveal that the SOT mechanism induces rapid magnetisation switching with lower current densities^{37–43}, as well as move domain walls with high velocities in ferromagnetic nanowires sandwiched by non-magnetic heavy metals (HMs) and oxide^{36,37,44,45}.

The origins of SOT are attributed to the large spin-orbit coupling in the conduction electrons of the ferromagnetic and HM leads to two well-known phenomena, namely, the Spin Hall Effect^{36,39,46,47} and the Rashba effect^{37,48–50}, which will be further discussed in Chapter 2. These two effects lead to the SOT on the local magnetisation. SOT can be generically described in the revised Landau-Lifshitz-Gilbert (LLG) equation by the term $-\gamma \mathbf{M} \times \mathbf{J}_s$, where γ is the gyromagnetic ratio, $\mathbf{M}$ is the ferromagnetic layer magnetisation, $\mathbf{s}$ is the spin accumulation, and $\mathbf{s}$ is a spin diffusion coefficient in the ferromagnetic layer. $-\gamma \mathbf{M} \times \mathbf{J}_s$ can be further decomposed into field-like torque, $\tau_{FL} = -H_{FL} \mathbf{M} \times \mathbf{p}$ and damping like torque, $\tau_{DL} = -H_{DL} \mathbf{M} \times (\mathbf{m} \times \mathbf{p})$, where $\mathbf{p}$ represents the electrons spin orientation diffusing into the ferromagnetic layer, $\mathbf{m}$ is the unit vector of $\mathbf{M}$. These torques are similar to the adiabatic and non-adiabatic STT respectively^{46,51,52}. The corresponding fields associated with the SOT are called the field-like term, $\mathbf{H}_F = H_F \mathbf{p}$ and damping-like term $\mathbf{H}_D = H_D (\mathbf{m} \times \mathbf{p})$ ^{37,46,49,52–57}.

Given potential advantages in SOT, several characterisation techniques such as alternating current (AC) Hall voltage modulation^{50,54,55,57–62}, spin-torque ferromagnetic resonance (ST-FMR)^{63–70}, magneto-optical Kerr effect (MOKE)⁷¹, SOT-assisted magnetisation switching^{36,37,39} and domain wall motion (DWM)^{50,72–74}, were developed and explored. Hayashi *et al.*⁵⁷ expanded on the AC harmonic Hall measurement by including both the Anomalous Hall Effect (AHE) and the planar Hall effect (PHE) signals in analytical expressions quantifying the spin-orbit (SO) effective fields. Greater understanding is needed for implementation of SOT in devices because SOT requires the cancellation of the dipole field and the creation of asymmetry to the switching field. As such, the implementation of devices will be a technological integration problem. Therefore, further understanding of the SOT is necessary for the progression towards technological applications, as such the focus of this thesis will be on the fundamental investigation of SOT effective fields in both single ferromagnetic and multilayer structures.

This thesis is arranged as follows:

- Chapter 1 provides the introduction to spintronics and MRAMs and the objectives of this thesis
- Chapter 2 provides the necessary theoretical information and basis for understanding this thesis
- Chapter 3 explains the detailed fabrication processes and experimental setup for the SOT characterisation using AC harmonic Hall measurement and the current-induced magnetisation switching
- Chapter 4 presents quantification of the effective fields by utilising a combination of the *in-situ* Kerr microscopy and harmonic measurement of a free layer consisting of MgO/CoFeB/Ta
- Chapter 5 presents the enhancement of SO effective fields due to the presence of rare-earth Tb in a perpendicularly magnetised multi-layer Pt/(CoNi)₂/Co/Tb structure. Illustrating rare-earth Tb as an alternative spin Hall material.
- Chapter 6 presents the probing of spin accumulation magnetoresistance utilising a mixed harmonic Hall technique to quantify the amount of spin accumulation in perpendicularly magnetised rare earth systems.
- Chapter 7 provides the conclusion of the thesis and a generic overview of the potential future work

Chapter 2: Theoretical Framework

2.1 Energy in a Magnetic System

2.1.1 Exchange Energy

Magnetism in solids originates from the electrons and the intrinsic spin of the atoms. The exchange interactions between neighbouring atoms can be expressed as the Dirac-Heisenberg Hamiltonian given by:

$$H_{ex} = \sum_{i,j} -J_{ex} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (2.1)$$

where J_{ex} is the exchange integral, $\mathbf{S}_i$ and $\mathbf{S}_j$ are the spin angular momentum of two neighbouring atoms i and j respectively. The J_{ex} can be explained further using the Heitler-London model for a two-electron wave function in a hydrogen molecule.

Figure 2.1. Distances between the nucleus, electrons of the H atom.

The Hamiltonian of the system can be expressed by the sum of the potential and kinetic energies given by:

$$\hat{H} = \frac{p_1^2}{2m_1} + \frac{p_2^2}{2m_2} + \frac{e^2}{4\pi\epsilon_0} \left(\frac{1}{r_{12}} + \frac{1}{r_{ab}} \right) - \frac{e^2}{4\pi\epsilon_0} \left(\frac{1}{r_{a1}} + \frac{1}{r_{b1}} + \frac{1}{r_{a2}} + \frac{1}{r_{b2}} \right), \quad (2.2)$$

The energy eigenvalues in the presence of the perturbation caused by the interaction are found by solving the secular determinant:

$$\begin{vmatrix} H'_{11} - E & H'_{12} \\ H'_{21} & H'_{22} - E \end{vmatrix} = 0, \quad (2.3)$$

$$(H'_{11} - E)(H'_{22} - E) - H'_{12}H'_{21} = 0 \quad (2.4)$$

$$H'_{11}H'_{22} - H'_{11}E - H'_{22}E + E^2 - H'_{12}H'_{21} = 0 \quad (2.5)$$

Since it is known that, $H'_{12} = H'_{21}$ and $H'_{11} = H'_{22}$, and the integral overlap such that,

$$S = \int \psi_1^* \psi_2 = \langle \psi_1 | \psi_2 \rangle = 1 \quad (2.6)$$

The energy of the system can be represented as:

$$E = \hat{H}'_{11} + \hat{H}'_{12} = \langle \psi_1 | \hat{H}' | \psi_1 \rangle \pm \langle \psi_1 | \hat{H}' | \psi_2 \rangle, \quad (2.7)$$

where $C = \langle \psi_1 | \hat{H}' | \psi_1 \rangle$ is the coulombic integral and $J_{ex} = \langle \psi_1 | \hat{H}' | \psi_2 \rangle$ is the exchange integral, rewriting (2.7) we get,

$$E_{\pm} = C \pm J_{ex} \quad (2.8)$$

where if $J_{ex} > 0$, the state E_- lies lower and it can be readily verified that this is the state which corresponds to an asymmetric linear combination of the two-electron spatial orbital wave functions; and if $J_{ex} < 0$, the state E_+ corresponds to a symmetric linear combination of the two-electron wave function.

As such, the total wavefunction of the system can be represented as the form:

$$\psi(1,2) = \psi(\vec{r}_1, \vec{r}_2) \cdot \phi(\vec{\sigma}_1, \vec{\sigma}_2) \quad (2.9)$$

where $\psi(\vec{r}_1, \vec{r}_2)$ and $\phi(\vec{\sigma}_1, \vec{\sigma}_2)$ corresponds to the orbital part and spin part, respectively.

$$\psi(\vec{r}_1, \vec{r}_2) = \frac{1}{\sqrt{2}} [\psi_a(\vec{r}_1) \psi_b(\vec{r}_2) \pm \psi_a(\vec{r}_2) \psi_b(\vec{r}_1)] \quad (2.10)$$

where $[+]$ corresponds to the symmetric combination of the wavefunction, associated with $S = 0$; $[-]$ corresponds to the asymmetric combination of the wave function, associated with $S = 1$. Based on Pauli's Exclusion principle, two identical fermions cannot occupy the same quantum state simultaneously. As such, the total electron wavefunction $\psi(1,2)$ should be antisymmetric, $\psi(1,2) = -\psi(2,1)$. This implies that when the spatial part of the wavefunction is symmetric, the spin component is antisymmetric, and vice-versa.

In the early fundamental understanding⁷⁵⁻⁷⁷ of magnetisation and magnetic ordering of the 3d transition metal elements, the Bethe-Slater curve shown in Figure 2.1 has successfully explained the antiferromagnetic ordering of bcc Cr, the ferromagnetic ground state of bcc Fe, hcp Co and fcc Ni^{78,79}, as well as the metastable polymorphs of these elements, such as bcc Co⁸⁰ and bcc Ni⁸¹. The microscopic origin of the Bethe-Slater curve is related to the direct-exchange, in which the relationship can be derived based on the relationship between magnetic ordering and the nearest-neighbour atomic distance^{78,79}. When the 3d electrons are relatively close to each other, the exchange energy, $J_{ex} < 0$ but when it moves further away, $J_{ex} > 0$, before dropping off. The Bethe-Slater curve provides a simple method of estimating the J_{ex} based on the average interatomic spacing. The value of J_{ex} enables the determination of the type of magnetic ordering present in the material, such that $J_{ex} > 0$ (ferromagnetic ordering) and $J_{ex} < 0$ (antiferromagnetic ordering). Recent evaluations of the exchange interaction show more complex physics involved, such that the Beth-Slater becomes too simple for evaluating the pressure dependence the Curie temperature of bcc Fe⁸² or the AFM order of Fe spins on W(001) surface⁸³ or different crystal lattice structures⁷⁷. That being said, the Beth-Slater curve relating to magnetic ordering remains sufficiently relevant for the understanding of the materials used in this thesis.

Figure 2.2 *Bethe-Slater Curve illustrating the function of the exchange integral with respect to the interatomic distances, where Mn and Cr have negative exchange energy which corresponds to an antiferromagnetic arrangement, while α Co and γ Fe, Ni and Gd have ferromagnetic arrangements.*

2.1.2 Magnetocrystalline Anisotropy Energy

The magnetocrystalline anisotropy energy originates from the spin-orbit interaction (SOI) of electrons and depends on the direction of the spontaneous magnetisation with respect to the lattice axes. As a result of this dependence, the electron spins tend to align in the direction of a distinct crystallographic axis, also known as the easy axis. Assuming that the energy has crystal symmetry, let $\{\alpha_i\}$ ($i = x, y, z$) represent the direction of the cosines of the magnetisation polarisation vector, $\vec{J}_s$ in the crystal orthogonal coordinate axis in which [100] and [001] are the x and z-axis respectively. The phenomenological function of the anisotropy, f_k as a function of $\{\alpha_i\}$, can be expressed as the expansion of the power series of α_x , α_y and α_z , and using the relation of $\alpha_x^2 + \alpha_y^2 + \alpha_z^2 = 1$ and neglecting the constant term to obtain an expression as:

$$\begin{aligned} f_k &= B_{21}(\alpha_x^2 + \alpha_y^2 + \alpha_z^2) + B_{41}(\alpha_x^4 + \alpha_y^4 + \alpha_z^4) + B_{42}(\alpha_x^2 \alpha_y^2 + \alpha_y^2 \alpha_z^2 + \alpha_z^2 \alpha_x^2) \\ &\quad + B_{61}(\alpha_x^6 + \alpha_y^6 + \alpha_z^6) + B_{62}(\alpha_x^2 \alpha_y^4 + \alpha_y^2 \alpha_z^4 + \alpha_z^2 \alpha_x^4) + B_{63} \alpha_x^2 \alpha_y^2 \alpha_z^2 + \dots \\ &= K_1(\alpha_x^2 \alpha_y^2 + \alpha_y^2 \alpha_z^2 + \alpha_z^2 \alpha_x^2) + K_2 \alpha_x^2 \alpha_y^2 \alpha_z^2 + \dots \end{aligned} \quad (2.11)$$

where B_{mn} and K_i are expansion constants.

Similarly, the expressions for cubic crystals can be deduced as:

$$\text{Tetragonal:} \quad f_k = K_1 \sin^2 \theta + K_2 \sin^4 \theta + K_3 \sin^4 \theta \cos 4\phi + \dots \quad (2.12)$$

$$\text{Hexagonal:} \quad f_k = K_1 \sin^2 \theta + K_2 \sin^4 \theta + K_3 \sin^6 \theta \cos 4\phi + K_4 \sin^6 \theta \cos 6\phi + \dots \quad (2.13)$$

where θ and ϕ are the zenith and azimuthal angles of the magnetisation polarisation vector. The expansions in (10) to (12) are infinite series but the first few terms of the magnetocrystalline anisotropy constants, K_i can be experimentally determined.⁸⁴ The direction or crystal axis in which the function f_k is minimum or maximum, it corresponds to the easy direction (easy axis) or hard direction (hard axis) respectively. If there exists no anisotropy in the c plane, the anisotropy is known to be uniaxial. Since the magnetocrystalline anisotropy is typically smaller than the exchange energy and $K_1 \gg K_2$, the function, f_k can be simplified to:

$$f_k = K_1 \sin^2 \theta \quad (2.14)$$

where θ denotes the angle between the easy axis and the magnetisation vector.

2.1.3 Zeeman Energy

The Zeeman energy exists when an external magnetic field, $\vec{H}$ is exerted on a magnetic system. Assuming a static magnetic field is distributed infinite space into which a magnetic moment is brought to the position $\vec{r}$ in a vacuum from the point of infinity. The magnetic polarisation at the position $\vec{p}(\vec{r})$. The work done by an external force to bring the change in energy of the system consisted of the field and the magnetic moment is⁸⁴:

$$E = - \int_{\infty}^{\vec{r}} \frac{\vec{p} \cdot \partial \vec{H}}{\partial \vec{r}} = - \int_{\infty}^{\vec{H}(\vec{r})} \vec{p} \cdot d\vec{H} , \quad (2.15)$$

And can be re-expressed as⁸⁵,

$$E = -\mu_0 \int \vec{H} \cdot d\vec{M} dV , \quad (2.16)$$

2.1.4 Demagnetisation Energy

Demagnetisation occurs due to the uncompensated magnetic moments at the surface of the magnetic elements or the spatial shape changes of the magnetisation. In order for the minimisation of energy from the magnetic field lines, the magnetisation prefers to align itself to the boundaries of the element. Thus, the demagnetisation is heavily dependent on the shape of the element. From Maxwell's equation,

$$\nabla \cdot \vec{B} = 0 , \quad (2.17)$$

where $\vec{B} = \mu_0 (\vec{H}_{demag} + \vec{M})$, such that $\vec{H}_{demag}$ is the demagnetisation field. As such, for a volume of charges, the relation between $\vec{H}_{demag}$ and $\vec{M}$ can be expressed as:

$$\nabla \cdot \vec{H}_{demag} = -\nabla \cdot \vec{M} , \quad (2.18)$$

Thus, simplifying the above expression to attain the demagnetisation energy as:

$$E_{demag} = -\frac{\mu_0}{2} \int \vec{H}_{demag} \cdot \vec{M} dV , \quad (2.19)$$

2.1.5 Magnetic domains and domain walls

The magnetisation of a system is described by magnetic domains of collinear magnetic moments which are also called Weiss domains⁸⁶. Dominated by the exchange energy of the system, the magnetisation aligns in a parallel configuration, as shown in Figure 2.3 (a). However, this is the least energy favourable configuration. The magnetic flux can be reduced by introducing two domains with oppositely aligned magnetic moments separated by a sharp boundary as shown in Figure 2.3 (b). In order to minimise the energy configuration further; the magnetic moments can be aligned parallel to the structural edges, as shown in Figure 2.3 (c). The sharp transitions along the boundaries of the magnetic domains are also known as the domain walls (DWs). Fundamentally, the formation of domains occurs due to the minimisation of the total free energy, E_{tot} , and in particular, caused by the reduction of the demagnetising and anisotropic field energy.

Figure 2.3 Schematic illustration of the magnetic flux reduction in a system (a) with no DWs and a large area with magnetic flux, (b) that has a reduction of the magnetic area through the reduction in the magnetic area by introducing a DW, (c) which exhibits no magnetic flux with multiple DWs.

DW can be classified by its energy per unit area and the DW width. The DW width occurs due to the competition between the exchange energy and the anisotropic energy in the system. There are mainly two types of DWs in a bulk system where the magnetisation depends on the position perpendicular to the wall only, as shown in Figure 2.4 (a) and (b) Néel walls and Bloch walls, respectively. Néel walls are characterised by spin components that rotate normal to the plane of the DW, whereas the Bloch walls rotate magnetisation in the plane of the DW, shown in Figure 2.4. The energy density of the DW can be approximated to the exchange and anisotropic energy given by⁸⁵:

$$\sigma_B \approx 4\sqrt{A.K} \quad (2.20)$$

where A is the exchange stiffness constant and K is the magnetocrystalline anisotropy constant. The DW thickness of a Bloch wall is given by⁸⁵:

$$\delta_B \approx 2\sqrt{\frac{A}{K}} \quad (2.21)$$

For the case for a Neel wall within a magnetic system with thickness, t much smaller than the DW width, δ_N the energy density will be similar to that of the Bloch wall. As such for ultrathin magnetic layers, both Néel and Bloch walls can be favoured in ultrathin films, but fundamentally, in the absence of the stabilisation by the Dzyaloshinskii-Moriya interaction (DMI), Bloch walls are the lower energy state configuration. DMI will be further discussed in later sections of the thesis.

Figure 2.4 Schematic of (a) Néel and (b) Bloch walls in a ferromagnetic system.

2.2 Spin-Orbit Coupling

Spin-orbit coupling (SOC) can be seen as a correction term to the electrostatic energy term arising from the relativistic Dirac equation. This phenomenon is commonly described by an electron with intrinsic spin, orbiting around a nucleus. This closed-loop path of the electron gives rise to an orbital angular momentum, $\vec{l}$ which couples with the spin angular momentum, $\vec{s}$ as shown in Figure 2.5. However, SOC is also found in systems with a potential gradient or $\vec{E}$ -field which gives rise to SOC as shown in Figure 2.5(a) while in Figure 2.5 (b) the emergent $\vec{B}$ -field couples with the electron spin momentum $\vec{s}$.

Figure 2.5 Schematic illustration of spin-orbit coupling (a) ls -coupling of the spin momentum of the electron and the orbital momentum due to the electron orbital motion around the nucleus (b) Bs -coupling where the $\vec{B}$ -field is generated by an $\vec{E}$ -field of a moving electron from the reference frame of the electron.

The coupling phenomenon can be better illustrated through the consideration of two inertial systems α and β , as shown in Figure 2.6. Generally, the electric and magnetic fields are given by the Lorentz transformations where the systems are moving with velocity, $\vec{v}$ towards each other. The Lorentz transformation of the electric and magnetic fields that are either parallel ($\parallel$) or perpendicular ($\perp$) is written as:

$$\vec{E}_{\parallel}^{\beta} = \vec{E}_{\parallel}^{\alpha}, \quad (2.22)$$

$$\vec{B}_{\parallel}^{\beta} = \vec{B}_{\parallel}^{\alpha}, \quad (2.23)$$

$$\vec{E}_{\perp}^{\beta} = \gamma \left(\vec{E}_{\perp}^{\alpha} + \vec{v} \times \vec{B}^{\alpha} \right) \quad (2.24)$$

$$\vec{B}_{\perp}^{\beta} = \gamma \left(\vec{B}_{\perp}^{\alpha} - \frac{\vec{v}}{c^2} \times \vec{E}^{\alpha} \right) \quad (2.25)$$

$\gamma = 1/\sqrt{1 - v^2/c^2}$ is the Lorentz factor with c the speed of light in vacuum. Using (2.19) such that $v \ll c$, the emerging $\vec{B}$ field can be expressed as shown in Figure 2.6(b) as:

$$\vec{B}^{\beta}(\vec{v}) = \frac{1}{2c^2} (\vec{E}^{\alpha} \times \vec{v}) \quad (2.26)$$

where the $1/2$ is the relativistic Thomas factor due to the non-inertial frame. For larger velocities, the factor follows the expression, $\gamma^2/(\gamma + 1)$. Thus, the Zeeman Hamiltonian due to SOC-field can be written as:

$$H_{SOC} = \frac{\mu_B}{2c^2} \vec{\sigma} \cdot (\vec{E} \times \vec{v}) = \mu_B \vec{\sigma} \cdot \vec{B} \quad (2.27)$$

where $\vec{\sigma}$ are the Pauli matrices.

Figure 2.6 Schematic illustration of the spin-orbit coupling in different inertial systems. (a) An electron is moving in a laboratory fixed reference frame with a fixed $\vec{E}$ field. (b) Rest frame of the electron, with moving $\vec{E}$ field towards the electron, leading to a $\vec{B}$ field.

SOC is generically categorised into two terms in a crystal system; with one of the terms being symmetry independent, i.e. Spin Hall effect (SHE) which exists in all crystal systems. The other term depends on symmetry in crystals without inversion symmetry such as the Dresselhaus (bulk) and Rashba (interfacial) effect. SOC is made manifest in several kinds of effects such as the magnetocrystalline anisotropy, Dzyaloshinskii-Moriya interaction (DMI), damping, anisotropic magnetoresistance (AMR), anomalous Hall effect (AHE), SHE, Rashba and Dresselhaus effect. The following section discusses the SHE and Rashba effect in greater detail because of their crucial role in contributing to the SOT effect fields.

2.3 Spin Hall Effect

In the previous section, spin-orbit coupling had been briefly discussed and attributed for the rise in spin-dependent scattering from impurity potentials. Spin-dependent scattering was first observed in solids when measuring the anomalous Hall effect (AHE) of ferromagnetic metals (FM). Although the AHE was discovered in the mid-1800s, its microscopic origins are not well understood until the mid-1900s. In 1971, D'yakonov and Perel⁸⁷ predicted that spin-orbit coupling (SOC) leads to a new family of Hall effects which is commonly known as the spin Hall effect (SHE). In their model, extrinsic Mott scattering (Skew scattering in materials) is the cause of the SHE and is still the most prominent approach in understanding SHE.

In an NM with large SOC, unpolarised electrons experience a spin-dependent velocity directing at the edges, as shown in the schematic of the SHE in Figure 2.7. A transverse spin current with respect to the charge current is formed with no net charge current distribution. This leads to spin accumulation at the edges, which transfers itself as a spin current when the edges are connected to another material, i.e. When an FM is adjacent to the conducting NM with large SOC, spin-polarised currents diffuse into the FM at the NM/FM interface.

Figure 2.7. A spin Hall effect schematic showing spin accumulation caused by spin-dependent scattering in the NM.

As the SHE phenomena consist of several effects, the following three effects will be discussed, namely, Mott scattering, anomalous velocity, and side-jump effects.

2.3.1 Mott Scattering

Mott scattering is a model that illustrates the elastic scattering of spin $\frac{1}{2}$ fermionic particles which can be visualised in Figure 2.8, which is extrinsically dependent on the presence of impurities.

Figure 2.8 *Illustration of Mott Scattering, where an electron with a particular velocity experiences a spin-dependent scattering along the B-field gradient.*

The electric field from the nucleus within the electron shell is dominant. Thus an incoming electron experiences a B -field that is equivalent to the Oersted field given by:

$$\vec{B}^{(b)}(\vec{v}) = \frac{\vec{E}^{(a)}}{2c^2} \times \vec{v} \quad (2.28)$$

where $\frac{1}{2}$ is the relativistic Thomas factor due to the non-inertial frame.

As the electric field is radial, the magnitude of the electric field from the nucleus diminishes with increasing distance from the nucleus, as shown in Figure 2.8. This B -field gradient gives rise to an additional spin-dependent force to the dominant Coulombic force acting on the incoming electron. Thus, the SOC force can be written in the form:

$$\vec{F} = \nabla(\vec{\mu} \cdot \vec{B}) \quad (2.29)$$

where $\vec{\mu} = (-eg/2m_e)\vec{s}$ is the magnetic moment of an electron, $-e$ is the electron charge, m_e is the mass of the electron and g is the g -factor from the Dirac equation.

The force can be re-expressed as,

$$\vec{F} = (\vec{\mu} \cdot \nabla) \vec{B} + (\vec{B} \cdot \nabla) \vec{\mu} + \vec{\mu} \times (\nabla \times \vec{B}) + \vec{B} \times (\nabla \times \vec{\mu}) \quad (2.30)$$

With the consideration that the spin-up (+ s_z) and spin down ($-s_z$) have similar trajectory towards the electron shell with velocity, v_x , (2.30) can be reduced to:

$$\vec{F} = \vec{\mu} \times (\nabla \times \vec{B}) = \pm \mu_z \frac{\partial B_z}{\partial y} \hat{e}_y \quad (2.31)$$

Electrons of opposite spins have a force acting in opposite directions as illustrated in Figure 2.8. Mott scattering is often characterised by the asymmetric motion of spin-polarised electrons in opposite directions, which relates closely to spin-polarisation. The symmetry is expressed by:

$$A_{\uparrow} = \frac{N_{\uparrow} - N_{\downarrow}}{N_{\uparrow} + N_{\downarrow}} = SP_{\uparrow} \quad (2.32)$$

where N is the number of electrons with spin orientation, S , also known as the Sherman factor and P is the polarisation.

2.3.2 Anomalous Velocity

Karplus and Luttinger⁸⁸ proposed the idea of the anomalous velocity in 1954 based on the theoretical formulation of the motion of electrons. Smit introduced the anomalous velocity based on wave packets formed from Bloch band states in 1955⁸⁹ and 1958⁹⁰, and in 1984, Berry⁹¹ introduced the most common theory till date, the geometrical phase. The anomalous velocity is also known as the intrinsic deflection that is independent of the impurity density and solely dependent on the electronic band structure of the periodic Bloch states^{92,93}, which can be expressed by:

$$\vec{v} = e\vec{E} \times \vec{\Omega} \quad (2.33)$$

where $\vec{\Omega}$ is known as the Berry phase curvature. The Berry curvature can be interpreted as an effective magnetic field in k -space of the Rashba spin-split band structure. In 2004, Sinova et al. reported that the anomalous velocity could be understood as a spin carrier in an applied electric field possessing an additional momentum, p_x as shown in Figure

2.9. The generation of the $\vec{B}$ -field causes spin precession. Assuming low damping, the emerging out-of-plane spin component is expressed by:

$$s_z = \frac{s\hbar^2}{2p^2} eE_x \sin \delta_{\vec{E}-\vec{p}}, \quad (2.34)$$

where $\delta_{\vec{E}-\vec{p}}$ is the angle between $\vec{E}$ field and the carrier momentum, $\vec{p}$. The sign of the out-of-phase spin, s_z will then depend on the direction of the momentum transverse to the E_y field, as shown in Figure 2.9, giving rise to an additional velocity, v_y independent of impurities.

Figure 2.9. Schematic illustration of the intrinsic spin Hall effect. The Fermi-surface in an effective $\vec{E}$ -field experiencing a torque such that the spin tilts out-of-plane in opposite directions.

2.3.3 Side-jump Effects

Berger introduced an extrinsic effect which ‘side-jumps’ off an impurity. The scattering extrinsically causes the side-jump against impurities. However, it is non-dependent on the concentration of impurity and contributes intrinsically to the spin Hall effect. A free-electron wave packet with constant velocity getting deflected by impurities does not side jump more than $1\text{\AA}$. However, the side-jump effect is spin-dependent, leading to the deflection of the wave packet in opposite directions for opposite spin polarity. Upon leaving the impurity, the constant velocity and directionality remain, as shown in Figure 2.10. The perturbation field caused by the impurity causes the electrons to change band, which results in the anomalous velocity.

Figure 2.10 Schematic of side jump mechanism when a wave packet with velocity, $\vec{v}$ causes the wave packet to be displaced by Δy during the perturbation impurity.

2.4 Rashba effect

The Rashba effect is an interfacial effect that arises from the interface of two crystals with different crystal potentials, V_1 and V_2 respectively, as shown in Figure 2.11. Due to the broken crystal symmetry, an electric field becomes the result of a potential gradient which exists between the crystals within screening length of a few nanometers^{94,95}. The conduction electron with velocity, $\vec{v}$ experiences an electric field as depicted by the green arrow in Figure 2.11. With the electron moving in the x -direction and the electric field pointing along the normal z -direction, the Rashba Hamiltonian can be expressed as⁹³:

$$H_R = \frac{\alpha_R m}{\hbar} \vec{\sigma} \cdot (\vec{z} \times \vec{v}) \quad (2.35)$$

where α_R is the Rashba coefficient. In a simplified model, the Rashba coefficient is equivalent to $\alpha_R = g\mu_B E / 2mc^2$ with g being the g -factor. In reality, due to the distortion of the electron wavefunction near the nuclei caused by the lack of inversion symmetry at the interface⁹⁶, the α_R was measured to be two orders of magnitude larger. The potential gradient which exists is strongly dependent on the band structure of the interfacial atoms which induces a non-uniform Rashba field.

Figure 2.11 *Schematic illustration of the Rashba effect. The interface of two crystals with different potentials, V_1 and V_2 , which causes a potential gradient. In this potential gradient, the electron travelling with velocity, $\vec{v}$ experiences an electric field which causes an emerging magnetic field.*

2.5 Dzyaloshinskii-Moriya Interaction

The Dzyaloshinskii-Moriya Interaction (DMI) is an antisymmetric exchange interaction, in which, the presence of SOC, two magnetic ions minimise their total magnetic energy by canting their magnetic moments. The Hamiltonian for the DMI between magnetic moments can be expressed as:

$$H_{DMI} = -\vec{D} \cdot (\vec{S}_1 \times \vec{S}_2), \quad (2.36)$$

where $\vec{D}$ is the DM exchange vector corresponding to the DMI strength. The neighbouring $\vec{S}_1$ and $\vec{S}_2$ spins align themselves in a minimum energy state such that the plane of the spins are always orthogonal to the DM vector, $\vec{D}$, as seen in Figure 2.12 (a)

Figure 2.12 Schematic illustration for the (a) bulk Dzyaloshinskii-Moriya interaction and, (b) interfacial Dzyaloshinskii-Moriya interaction.

In the interfacial DMI which occurs in HM/FM stacks that were investigated for SOT. The canting of spins is caused by the SOI from the HM, as shown in Figure 2.12 (b). The DMI energy can be written as:

$$E_{DMI} = \int f_{DMI} dV, \quad (2.37)$$

where f_{DMI} is:

$$f_{DMI} = D \left(m_z \frac{\partial m_x}{\partial x} - m_x \frac{\partial m_z}{\partial x} + m_z \frac{\partial m_y}{\partial y} - m_y \frac{\partial m_z}{\partial y} \right), \quad (2.38)$$

D is the area per unit energy density in J/m² associated with the DM vector, $\vec{D}$ through the proportionate factor $1/a_0 t$. a_0 is the atomic lattice constant and t is the FM film thickness.

2.6 Magnetisation Dynamics: Landau-Lifshitz-Gilbert (LLG) equation

In this section, the theoretical description of the magnetisation dynamics for magnetic field induced and current-induced magnetisation dynamics will be discussed and explored. Landau and Lifshitz first described the magnetisation dynamics in 1935 through the equation, $d\vec{M}/dt = -\gamma(\vec{M} \times \vec{H}_{eff})$, where γ is the gyromagnetic ratio given by $\gamma = g\mu_0\mu_B/\hbar$. This implies that the magnetisation is precessing around the effective applied field, as shown in Figure 2.13. The angle, θ between the magnetisation vector and effective field vector, does not change due to the precession. Thus, in order to obtain an equilibrium state in which magnetisation and effective field are parallel, a secondary torque is added to the equation. This is also known as the damping term, as shown in Figure 2.13 and can be expressed as the original Landau-Lifshitz equation given by⁹⁷:

$$\frac{d\vec{M}}{dt} = -\gamma \cdot (\vec{M} \times \vec{H}_{eff}) - \frac{\alpha}{M_s} \vec{M} \times (\vec{M} \times \vec{H}_{eff}), \quad (2.39)$$

where the damping constant, α must be experimentally determined. The damping term causes the magnetisation to be torqued towards the direction of the effective field until collinearity occurs. Subsequently, Gilbert added an alternative damping term to the equation and the Landau-Lifshitz-Gilbert (LLG) equation can be expressed as:

$$\frac{d\vec{M}}{dt} = -\gamma_G \cdot (\vec{M} \times \vec{H}_{eff}) - \frac{\alpha_G}{M_s} \vec{M} \times (\vec{M} \times \vec{H}_{eff}), \quad (2.40)$$

Figure 2.13 *Magnetisation precession around the effective field and the precession with additional damping torque towards the effective field.*

By comparing (45) and (46), the coefficients of the LLG equation are related in the following:

$$\gamma = \frac{\gamma_G}{(1 + \alpha_G^2)} \quad ; \quad \alpha = \frac{\alpha_G \gamma_G}{(1 + \alpha_G^2)}, \quad (2.41)$$

2.7 Current Induced Spin Torques

The current which passes through the magnetic material influences the dynamics of the magnetisation. Berger predicted⁵¹ that the s-d exchange between the spin-polarised conduction electrons and the DW causes a torque on the conduction electrons as they travel across the boundary. In return, the magnetisation experiences a torque which leads to magnetisation switching, precession or DW motion. This phenomenon is the spin-transfer torque (STT) and was added into the LLG equation by Slonczewski⁹⁸. Ever since there were a variety of formalisms for the description of the STT, and two terms consider for the STT are discussed in the following sections. Figure 2.14 below illustrates the interaction between a spin-polarised current and the magnetisation of the ferromagnet, in which their difference in polarisations lead to spin-torque acting on the ferromagnet.

Figure 2.14. When a spin polarised current enters a ferromagnet, the interaction between the spin-polarised current and the magnetisation causes a change in the spin direction of the outgoing electron in contrast to the incoming electron. The difference in the polarisation causes a torque on the ferromagnet. The purplish arrow represents the magnetisation of the ferromagnet.

2.7.1 Adiabatic Spin-Transfer Torque

The adiabatic STT arises due to current injection. The spin-polarised current traverses through DWs, such that the spin orientation of the conduction electrons aligns itself to match the direction of the magnetisation. Thus, an adiabatic STT is a gradual change of the spin of the electron as it travels through the magnetisation texture and the reactive torque from that back onto the magnetisation. This rotational motion of the electrons can be expressed as⁹⁹:

$$\left(\frac{\partial \vec{M}}{\partial t} \right)_{\text{Adiabatic}}^{\text{STT}} = -(\vec{\mu} \cdot \nabla) \vec{M}, \quad (2.42)$$

where $\vec{\mu}$ is the effective velocity given by [92]:

$$\vec{\mu} = \frac{gP\mu_B}{2eM_s} \vec{J}, \quad (2.43)$$

where P is the spin polarisation of the magnetic material and $\vec{J}$ is the charge current density.

2.7.2 Non-Adiabatic Spin-Transfer Torque

The non-adiabatic STT is due to the non-adiabaticity of the electron spins in, causing a change in magnetisation. This torque exerts on the local magnetisation due to the misalignment of the electron spin and the local magnetisation. The non-adiabatic STT can be expressed as²⁹:

$$\left(\frac{\partial \vec{M}}{\partial t} \right)_{\text{Non-Adiabatic}}^{\text{STT}} = \beta \vec{M} \times ((\vec{\mu} \cdot \nabla) \vec{M}), \quad (2.44)$$

where β is the non-adiabaticity constant. Both STT terms can be added into the LLG equation in (46) to describe the magnetisation dynamics of a system with applied fields and charge currents:²⁹

$$\frac{d\vec{M}}{dt} = -\gamma_G (\vec{M} \times \vec{H}_{\text{eff}}) - \frac{\alpha_G}{M_s} \vec{M} \times (\vec{M} \times \vec{H}_{\text{eff}}) - (\vec{\mu} \cdot \nabla) \vec{M} + \beta \vec{M} \times ((\vec{\mu} \cdot \nabla) \vec{M}), \quad (2.45)$$

Both the adiabatic and non-adiabatic STTs are inherent in the measurement as current also flows in the FM layer. However, the current density required for magnetisation manipulation is an order of magnitude larger than that compared to the SOT. ($\sim 10^{12}$ A/m²)⁷³.

2.7.3 Spin-Orbit Torque

Figure 2.15 Schematic illustration of the damping-like and field-like SOT effective fields on a perpendicularly magnetised FM.

The SOT can be perceived as a current-induced effective field, $\Delta\vec{H}$, and expressed in the LLG equation as:

$$\frac{d\vec{M}}{dt} = -\gamma \left(\vec{M} \times \left(\vec{H}_{eff} + \Delta\vec{H} \right) \right) - \frac{\alpha}{M_s} \left(\vec{M} \times \frac{d\vec{M}}{dt} \right), \quad (2.46)$$

In a paper by Zhang *et al.*⁴⁶, they showed that spin accumulation transverse to the local magnetisation occurs, which acts as a polariser layer similar to the reference layer in spin valves or MTJs which is given by:

$$\frac{d\vec{M}}{dt} = -\gamma_G \cdot \vec{M} \times \vec{H}_{eff} - a\vec{M} \times \hat{p} - b\vec{M} \times (\hat{p} \times \vec{M}) \left((+\Delta\vec{H}) \right) + \frac{\alpha_G}{M_s} \left(\vec{M} \times \frac{d\vec{M}}{dt} \right), \quad (2.47)$$

$\hat{p}$ is the unit vector of the spin current traversing into the FM layer which results in spin accumulation acting on the localised magnetisation. a and b are constants with respect to $\vec{M}$, which are related to the material parameters and current as⁴⁶:

$$a = \frac{hJa_0^3\gamma}{\sqrt{2e\mu_B}\lambda} \left(\frac{1 - \cos De^{-D}}{D} \right), \quad (2.54) \quad b = \frac{hJa_0^3\gamma}{\sqrt{2e\mu_B}\lambda} \left(\frac{\sin De^{-D}}{D} \right), \quad (2.48)$$

where h is Planck's constant, J is the current density, $D = t_F / \sqrt{2}\lambda$, a_0 is the lattice constant and λ is the spin diffusion length. The two torques are related to a and b are known as the field-like torques⁴⁶ and damping-like torques^{46,51,98}, respectively. As such, the corresponding effective fields can be expressed as:

$$\vec{H}_{FL} = \frac{a}{\gamma} \hat{p}, \quad (2.49) \quad \vec{H}_{DL} = \frac{b}{\gamma} \hat{p} \times \vec{M}, \quad (2.50)$$

2.8 Electrical Detection of Magnetisation

2.8.1 Ordinary Hall Effect (OHE)

Figure 2.16 Schematic of Ordinary Hall effect

The OHE occurs when a charge current in a conductor experiences a Lorentz force due to the presence of an external magnetic field, $\vec{B}$ acting perpendicular to the charge current density with velocity, $\vec{v}$. The Lorentz force can be expressed as:

$$\vec{F} = -e\vec{v} \times \vec{B}, \quad (2.51)$$

This Lorentz force can be directly detected as a Hall voltage given by the equation:

$$V_H = vB\Delta y \cos \alpha, \quad (2.52)$$

with α being the angle between $\vec{v}$ and $\vec{B}$. In the case when the magnetic field is applied out-of-plane, the ordinary Hall voltage makes a relatively small contribution to the overall Hall voltage as compared to the AHE at room temperature in microstructures.

2.8.2 Anomalous Hall Effect (AHE)

Figure 2.17 Schematic of Anomalous Hall effect

The AHE is firmly rooted in the ideas of SOC which is also of similar origins to the SHE. AHE phenomenologically describes the induced transverse charge-spin current due to the spin-polarised charge current, as shown in Figure 2.17. In the AHE case, when there is no $\vec{B}$ field applied, the effect still exists. This transverse Hall voltage can be written as:

$$R_H = \Delta R_A \cos \theta, \quad (2.53)$$

where $\Delta R_A = (C/t_f)M_s$ is the AHE resistance, C is a proportionate constant which relates itself to the AHE, t_f is the thickness of the magnetic material and θ is the angle between the magnetisation and the surface normal.

2.8.3 Planar Hall Effect (PHE)

Figure 2.18 Schematic of Planar Hall effect

The PHE is related to the AMR of a system which can be described by an electric field given by:

$$\vec{E} = \rho_{\perp} \vec{J} + (\rho_{\parallel} - \rho_{\perp}) \hat{m} (\vec{J} \cdot \hat{m}), \quad (2.54)$$

where $\vec{E}$ is the effective electric field in the magnetic structure, $\vec{J}$ is the current density applied to the magnetic structure, $\rho_{\perp}$ is the resistivity when the magnetisation is orthogonal to the current density, $\rho_{\parallel}$ is the resistivity when the magnetisation is aligned with the current density and $\hat{m}$ is the unit magnetisation vector. Fixating the magnetisation geometry as Figure 2.17, and current density in the x-direction, the electric field components, E_x and E_y can be expressed as^{100,101}:

$$E_x = \rho_{\perp} J + (\rho_{\parallel} - \rho_{\perp}) J \sin^2 \theta \cos^2 \varphi, \quad (2.55)$$

$$E_y = (\rho_{\parallel} - \rho_{\perp}) J \sin^2 \theta \cos \varphi \sin \varphi, \quad (2.56)$$

E_x corresponds to the AMR whereas the E_y corresponds to the PHE. This transverse electric field E_y is reformulated to express in terms of the Hall voltage given as:

$$R_H = \Delta R_p \sin^2 \theta \sin 2\varphi, \quad (2.57)$$

where $\Delta R_p = k M_s^2 / t_F$ is the PHE resistance change and k is a proportionality constant.

2.8.4 Spin Hall Magnetoresistance (SMR)

The SMR is most prominently focused on a SHE-induced proximity effect which explained some results for a ferromagnetic insulator(FI)/NM bilayer structure in yttrium iron garnet (YIG)/Pt interface^{102–104}. The induced resistance is measured through the inverse SHE where a pure spin current induces a transverse charge current¹⁰⁵. Several other explanations of the SMR phenomena have related it to edge spin accumulation¹⁰⁶, Hanle effect¹⁰⁷ or interfacial spin-orbit scattering¹⁰⁸. Hence, the magnetoresistance correlation with x-direction current is related by¹⁰³:

$$R_x = R_0 - \Delta R_S m_y^2 = R_0 - \Delta R_S \sin^2 \theta \sin^2 \varphi, \quad (2.58)$$

$$R_y = R_H = \Delta R_S m_x m_y = \Delta R_S \sin^2 \theta \sin 2\varphi, \quad (2.59)$$

R_0 is the electrical resistance of the wire and ΔR_S is the resistance change due to SMR. Since the effect exhibit similar geometry to the PHE, the longitudinal resistance is required to distinguish their effects. The SMR has been observed in NM/FM layer heterostructures recently¹⁰⁹.

Chapter 3: Fabrication & Measurement Methods

In this chapter, thin film deposition, magnetic thin film characterisation, sample fabrication process steps, and the electrical measurement methods will be explained in greater detail.

3.1 Physical Vapor Deposition

All thin films and contact electrodes investigated in this thesis are deposited using ultra-high vacuum magnetron sputtering using the AJA 8-gun sputtering system. Perpendicularly magnetised thin-film structures were deposited onto 300nm thermally oxidised silicon substrate at base pressures of low 10^{-8} Torr and working pressure of 3mTorr at 50W. The schematic of the magnetron sputtering and the confocal AJA magnetron sputtering system is shown in Figure 3.1.

Figure 3.1 *Schematic of the confocal magnetron sputtering and the AJA sputtering deposition system.*

Sputtering is a physical vapour deposition (PVD) technique which utilises high voltage to ionise inert noble gases, i.e. Ar, Xe, as well as reactive gases such as N₂ and O₂ and generates plasma, in an ultra-high vacuum system^{110–113}. The ultra-high vacuum of the system is attained through the concurrent employment of a turbomolecular pump and a cryopump which enables the achievement of $\sim 10^{-8}$ Torr base pressure. This base

pressure ensures that contamination of process gases is kept to the minimum while enhancing the mean free paths of the ion for plasma sustenance. Within the 10^{-8} Torr base pressure, the working pressure of 2-3mTorr and Ar/Xe flow rate of approximately 20 sccm, the sputter rate can be determined through calibration samples using atomic force microscope (AFM). The sputtered thickness for a fixed time of 1000s was then used to calculate the sputter rate (nm/s) for each target material.

When sputtering, the positively charged ions are then accelerated towards the negatively biased material target. This process involves the striking of the positive gas ions onto the target dislodging and ejecting target material in a typical line of sight cosine distribution. The ejected target atoms deposit itself onto the wafer surface, which is placed near to the sputtering cathode, as shown in Figure 3.1. Metallic and insulative target materials are deposited using DC power supply and RF power supply, respectively. In magnetron sputtering, the utilisation of closed loops magnetic fields around the target material enables the confinement and entrapment of electrons, which increases the path length of the electrons causing a spiral motion^{114,115}. The efficiency of the initial ionisation is enhanced due to this process enabling plasma sustenance at low pressures, which causes a reduction in both background gas incorporation and energy losses in the sputtered atoms through collisions.

3.2 Vibrating Sample Magnetometer (VSM)

Vibrating sample magnetometer (VSM) is an essential tool in determining the fundamental magnetic properties of the material stack. In this thesis, the Lakeshore 8600 series VSM was vastly utilised to obtain the magnetisation details of the stack structures being studied, as shown in Figure 3.2(a) and the working principles of the VSM are shown in Figure 3.2(b).

Figure 3.2. (a) Lakeshore 8600 series VSM that was used to characterise magnetic thin films. The sensitivity of the VSM is approximately $0.1 \mu\text{emu}$ with an electromagnet that is capable of fields up to 2.5 T. (b) Schematic of the working principles and measurement geometry of the VSM

VSM is based on Faraday's law of induction in which a rate of change of magnetic field gives rise to an induced electric current. This electric current is detected from the pick-up coils at the pole-cap of the electromagnet. In the measurement of the magnetic moment, a thin film coated Si wafer is mounted unto the vibrating rod and under the uniform application of a constant magnetic field, the magnetic moments align itself to the applied field. The sample vibrates in the up-down (z) direction, which generates magnetic flux that perturbs the system. This alternating change in flux caused by the alignment of the magnetic moments generates an electromotive force (ε) in the pick-up coils according to Faraday's law of induction, i.e. $\partial\Phi/\partial t = -\varepsilon$, where Φ is the sample

induced change in magnetic flux. As the sample vibrates with a particular frequency, f , this frequency is used as the reference frequency with an in-built lock-in amplifier of the system. This enables the signal from the same frequency to be detected through the reference frequency; as such, this enhances the signal-noise ratio (SNR) of the measurement. For a sample with PMA, its easy axis will be in the z-axis showing a sharp rectangular hysteresis loop and hard axis are as shown in the schematic in Figure 3.3.

Figure 3.3 Schematic of the easy and hard axis loops for a sample with PMA.

3.3 Magneto-Optical-Kerr Effect

The Magneto-Optical-Kerr Effect (MOKE) polarimetry is a technique for the quick characterisation of magnetic thin films based on the polarisation of the reflected light. MOKE may not be able to provide the actual values of magnetisation, but it can rapidly measure the shape of the hysteresis loop and provide brief behaviour of the magnetisation. A simple illustration of the MOKE concept is shown in Figure 3.4(a) and Kerr microscope that was used to capture the behaviour of the magnetisation by observing the change in Kerr contrast when the magnetisation changes are leading to a change in relative polarisation.

Figure 3.4 (a) Schematic illustration of a typical polar-MOKE measurement setup, (b) MagVision Kerr Microscope.

3.4 Electron Beam Lithography (EBL)

EBL is a useful patterning tool that writes nano-scaled patterns unto a resist-coated sample. In this thesis, the Raith e-Line system, as shown in Figure 3.5, was used for the patterning of devices. This process of lithography involves the coating of the sample with a polymeric resist that is sensitive to energetic electron beams. The energetic electron beams either causes the resist to become more soluble (positive resist) or insoluble (negative resist) when immersed in the developer solution.

Figure 3.5 *Raith e-Line EBL system and inset EBL sample stage for the placement of photo-resist coated thin film wafer for patterning.*

Before first layer EBL exposure, thin film samples are diced into $1 \times 1 \text{ cm}^2$ for patterning a number of devices per wafer. The diced samples are then cleaned by ultrasonication in acetone for approximately 15 minutes for the removal of organic contaminants. Subsequently, the samples are sonicated in isopropyl alcohol (IPA) for 15 minutes to remove residual acetone and remaining contaminants. The samples are then blown dry using a N_2 airgun. After cleaning the samples, they are spun-coated with either a positive or negative photoresist (see next sub-section). The coated sample was then placed in a Raith e-line EBL system for pattern transfer.

Figure 3.6 Schematic illustration of the EBL process. The electrons are collimated using an electromagnetic lens and are passed through the pattern generator before exposure unto mounted sample.

In the EBL process, as shown in Figure 3.6, the source of electrons is from thermionic emission of lanthanum hexaboride. The electrons are accelerated from the source and collimated using condenser lenses with beam blanker and deflector controls to direct the electron beam onto the coated sample. The samples are exposed on an ultra-high precision stage, that is controlled precisely using a laser interferometer. The lithographic processes are two-pronged, as the first layer involves a top-down lithography process using a negative resist, while the contact pad layer involves a bottom-up lithography process. The detailed processes are described in the following sections.

3.4.1 First Layer Fabrication: Top-Down Lithography

In this top-down process, a layer of ma-N 2403 Negative Tone resist was coated onto the thin film samples at 6000 rpm for 60s to ensure minimal edge beads and laminar coating of less than 200 nm. The resist was baked over a hotplate at 110°C for approximately 1-2 minutes to evaporate any residual organic solvents. The resist coated film was exposed at a dose of 130 $\mu\text{C}/\text{cm}^2$, with a beam current of 200 pC and constant aperture size and beam voltage of 30 μm and 10 keV, respectively. The beam exposed regions become insoluble due to cross-linking, and the unexposed regions are dissolved away using ma-D 525 developer solution for approximately 90s. The samples were then rinsed in de-ionised (DI) water to remove any residual resist and developer solution. Figure 3.7 illustrates the process flow until the dry-etch and resist stripping process.

Figure 3.7 Schematic of top-down lithography process (i) Thin film deposition (ii) Spin coating of ma-N photoresist (iii) electron beam exposure (iv) development in ma-D 525 (v) dry etch using ion milling technique to etch away deposited film till Si substrate (vi) Lift off/resist stripping in mr-Rem solution.

The stripped samples are placed onto the rotating sample stage of the AJA ATC-2020-IM ion milling system, as shown in Figure 3.8. The sample stage is angled at 80° to the beam direction and rotated azimuthally during the milling process. The samples were milled at 54 W for at 8 minutes. In this process, the Ar^+ ions attack the surface of the sample, ejecting atoms from both the exposed resist as well as the thin film, as shown in Figure 3.7. As the thin film is usually in the order of approximately 20 nm and the underlayer SiO_x are in the orders of $\sim 300\text{nm}$, the samples were over-etched, resulting in a non-uniform SiO_x film layer. However, this is not crucial in our case since the thick resist protects the patterned magnetic structure, and therefore, the surface quality will not be compromised by the dry etch process.

Figure 3.8 Image of the AJA Ion milling system and inset is the schematic of how the Ar^+ plasma is angled towards the rotating sample stage, ensuring that sidewalls are etched appropriately.

3.4.2 Contact Pad Fabrication: Bottom-Up Lithography

Upon completing the first layer fabrication, a bottom-up lithography technique is completed through the utilisation of a positive resist, followed by PVD of the film and subsequent lift-off of the resist. This technique was used for the contact pad fabrication process.

Figure 3.9 Schematic of the bottom-up lithography for contact pad fabrication process (i) Post-ion-milled sample after stripped using mr-Rem solution (ii) spin coating of PMMA 950K positive resist (iii) development in developer solution consisting of MIBK: IPA in 1:3 ratio (iv) Deposition of contact pads and lift-off in acetone

As shown in Figure 3.9, the sample is coated with PMMA 950K resist spun at 4000 rpm for 45s, followed by baking at 150 °C for 2 minutes. The sample is then exposed to an electron beam for the pattern transfer like the previous segment, except that, the exposed regions exposed becomes soluble in the diluted methyl isobutyl ketone (MIBK) in IPA solution. The exposed regions dissolve in the developer after 30s followed by rinsing the sample in IPA for 30s. The contact pads consisting of Ta (5nm)/Cu(100nm)/Ta(5nm) were then sputtered. The Ta capping layer serves as a protection for the Cu layer. Before the deposition, reverse sputtering was done for 60s at 30W RF power to remove impurities at the surface. After the deposition was complete, a gentle lift-off process was done on the samples in acetone at 65°C for 1 hour. After which, acetone was poured away, and second acetone bath was prepared. In the second acetone bath, the samples would then be ultrasonicated to remove the residual resist.

3.5 Measurement Setup

3.5.1 Harmonic Hall Measurement Setup

The alternating current (AC) magneto-transport measurements require the following equipment:

- *Keithley AC-DC 6221* Current Source
- *Signal Recovery 7265* Dual-Phase digital signal processor (DSP) Lock-in amplifier
- Electromagnet with *Kikusui PBZ* bipolar power supply (~8A)
- *National Instrument* LabView software for data acquisition and equipment control
- *TDK Lambda* Power supply for the air-core electromagnet
- *Hirst GM08/Lakeshore* Gaussmeter
- Custom-built manual stage for azimuthal 360° rotation
- Custom-built printed circuit board (PCB) chip
- Wire Bonder

Figure 3.10 Schematic illustration of the harmonic Hall measurement set-up with the detailed connections on the Keithley AC-DC 6221 current source and Signal Recovery 7265 DSP Lock-in amplifier to the electrical break-out box to Hall cross-device.

A full connection of the harmonic Hall setup is, as shown in Figure 3.10. The patterned Hall cross is connected to the AC source, and the Hall voltage connections are connected to the lock-in amplifier in the front channel A, of the lock-in amplifier, as indicated in Figure 3.10. The trigger link of the *Keithley 6221* is connected to the transistor-transistor logic (TTL) terminal of the lock-in amplifier, which provides the reference frequency. The patterned device is mounted unto a custom-built PCB and wire bonded to the chip, as shown in Figure 3.10(a). This device mounted chip is then placed unto a custom-built rotation stage that is capable of 360° azimuthal angular rotation, as shown in Figure 3.11(a). After mounting of the PCB chip unto the stage, the gaussmeter probe is placed in between the pole of the electromagnet and above the device chip. The presence of the air-core electromagnet enables the prevention of domain nucleation during the modified harmonic Hall measurements. All equipment specified earlier is controlled electronically using LabView, and the generic H_z -bias harmonic Hall measurement schematic is shown in Figure 3.10 (b).

Figure 3.11 (a) Wire-bonder, wire bond configuration and device mounted unto the custom-built PCB chip with 8-pins. (b) Experimental set-up of the (i) conventional Harmonic Hall measurement and the (ii) H_z -bias harmonic Hall measurement

The LabView program written for data-acquisition is capable of concurrently measuring the first and second harmonic Hall voltages while being able to control the in-plane magnetic field sweep, AC current sweep, DC offset current and the strength of the air-core electromagnet or permanent magnet, H_z bias field applied used in the measurements. The program overview is shown in Figure 3.12.

Figure 3.12 Program overview of the programmable controls in the LabView program for the harmonic Hall measurements, which evaluates the effective fields using the programmed experimental fittings to attain the effective fields.

Figure 3.13 Measurement geometry in cartesian coordinate system for polar angle (a) reference to plane, and (b) reference to normal

Two measurement geometries are taken into account in this thesis, where chapter 4 is based on Figure 3.13 (a) where the sample experienced a form of switching and fitted using the equation described in Chapter 4 while H_z biased harmonic measurement was used in chapters 5-6, based on the geometry given in Figure 3.13 (b) where the description of the measurement techniques are also described in Chapter 5.

3.5.2 Current-Induced Switching Measurement

The direct current (DC) current induced SOT switching measurements require the following equipment:

- Keithley 24XX series DC source meter
- An electromagnet with Kikusui PBZ bipolar power supply (~8A)
- National Instrument LabView software for data acquisition and equipment control
- Hirst GM08/Lakeshore Gaussmeter
- Custom-built manual stage for azimuthal 360° rotation
- Custom-built printed circuit board (PCB) chip
- Wire Bonder

Figure 3.14 Schematic illustration of the experimental measurement setup for current-induced magnetisation switching, R_H vs I loop measurements.

Figure 3.15 Program overview of the programmable controls in the LabView program for the Hall resistance vs current measurements, which allows the collection of data automatically through the variation of the bias in-plane field and the size of the current sweep.

Figure 3.16 Current induced magnetisation switching curves with external in-plane magnetic field $H_x = \pm 3$ kOe for Pt/[Co/Ni]₂/Co/Ta (5nm) sample.

In the current-induced switching measurement, a bias H_x field was applied in the same direction as the pulsed dc current in order to break symmetry to ensure that SOT switching occurs. The applied current was pulsed with 10ms pulses between positive and negative maximum current to reduce the Joule heating effects on the switching when a constant current was applied. The critical SOT switching current is defined as the point where a change in Hall resistance occurs, as shown in Figure 3.16 for various applied external in-plane field H_x .

Chapter 4: *In-situ* Kerr and Harmonic Measurement of perpendicularly magnetised MgO/CoFeB/Ta

Abstract

In this chapter, the experimental determination of the spin-orbit (SO) effective fields in a MgO/CoFeB/Ta structure utilising a combination of the harmonic measurement and in situ Kerr imaging is reported. Here, an evaluation of the SO effective fields through an analytical energy approach by transforming the anomalous Hall effect and planar Hall effect (PHE) voltage into a field dependency was done while imaging the magnetisation behaviour via differential Kerr microscopy. The analytical fitting to the measurement data indicates the significant coexistence of both a transverse field, H_T , and longitudinal field, H_L , in the longitudinal ($H_L = -12$ Oe, $H_T = 8$ Oe per 10^6 A cm⁻²) and transverse ($H_L = -12$ Oe, $H_T = -17$ Oe per 10^6 A cm⁻²) measurement schemes respectively, due to the PHE. Additionally, the observation of dendritic-like domains which indicate the influence of the interfacial Dzyaloshinskii–Moriya interaction (DMI) at the CoFeB/Ta interface, were observed by *in-situ* Kerr imaging. Micromagnetic simulations confirm that the dendritic domain formation and edge tilting of the magnetisation is due to the DMI.

Q. Y. Wong *et al.*, "*In-Situ* Kerr harmonic measurement in determining current-induced effective fields in MgO/CoFeB/Ta", Journal of Physics D: Applied Physics, 51, 115004 (2018).

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4.1 Introduction

The demand for memory and logic devices^{54,116} has led to significant efforts in spintronic material system investigations. To improve device performance, efforts have been focused on using spin-orbit (SO) effective fields to manipulate magnetisation, *i.e.* in-plane (IP) current-induced switching,^{36,39,117–120} fast DW motion^{53,72,121} and magnetic oscillation^{122–124}. In heavy metal (HM)/ferromagnetic metal (FM)/oxide trilayer systems, the SO fields generate a SO torque (SOT) which induces spin accumulation via *s* and *d* electrons^{46,125}. Several reports have proposed the underlying mechanism of SOT to be the spin Hall effect (SHE)^{36,39,50,53,125} in bulk HM and at the HM/FM interface to be the Rashba effect^{37,48,49,58}. The spin-orbit (SO) effective fields consist of two components^{37,39,49,55,56,126}, *i.e.*, the longitudinal damping-like field (H_L) and the transverse field-like field (H_T) along the $\mathbf{y} \times \mathbf{m}$ direction and $\mathbf{y}$ direction respectively, where $\mathbf{m}$ and $\mathbf{y}$ are directional unit vectors. The signs of H_L and H_T are dependent on the heavy metal and capping layer thicknesses^{56,127}. In MgO/CoFeB/Ta systems, the coexistence of both the H_L and H_T enables a better study of the effective field components⁵⁴ [63] in contrast to Pt-based systems^{39,72}.

In this work, *in-situ* Kerr imaging was conducted during the harmonic Hall measurements to observe the magnetisation behaviour directly. An analytical energy approach was used to fit the harmonic Hall measurements. This was done by transforming the Hall voltage equation to a field-dependent equation to determine the coexisting H_L and H_T in perpendicularly magnetised Ta/MgO/Co₂₀Fe₆₀B₂₀/Ta structure through the second harmonic relation. The dendritic domain width with respect to applied fields was studied, and strength of the Dzyaloshinskii-Moriya interaction (DMI) for the film was determined. Micromagnetic simulations were conducted to compare with the experimentally determined DMI strength.

4.2 Theory

The total magnetic energy in spherical polar coordinates is given by the sum of the contributing energies⁵⁷,

$$E_{total} = (N_z M_s^2 - K_{\perp}) \sin^2 \theta + N_y M_s^2 \cos^2 \theta \sin^2 \phi + N_x M_s^2 \cos^2 \theta \cos^2 \phi - M_s H_z \sin \theta - M_s H_y \cos \theta \sin \phi - M_s H_x \cos \theta \cos \phi, \quad (4.1)$$

where $N_z M_s^2 - K_{\perp} \leq 0$, $N_z M_s^2$ is the perpendicular demagnetising energy density, N_x , N_y , N_z are the demagnetising factors along the z , y and x -direction respectively, H_x , H_y and H_z are the applied field components along the three principal axes. The Hall voltage measured in the harmonic method includes both anomalous (AHE) and planar Hall effects (PHE) when an alternating current with frequency f passes through the nanostructure. The magnetisation oscillation due to the SOT can be characterised by $\Delta\theta$ and $\Delta\phi$. Under conditions where $\phi = 0^\circ$ and $\phi = 90^\circ$, the 2nd harmonic expressions for the longitudinal and transverse scheme can be given by the expressions previously reported in Qiu *et al.*⁴⁵ as,

$$V_{2\omega, //} = -\frac{1}{2} I_{ac} R_{AHE} \Delta\theta \cos \theta - I_{ac} R_{PHE} \Delta\phi \cos^2 \theta \quad (\text{H in } x\text{-}z \text{ plane, } \phi = 0^\circ), \quad (4.2)$$

$$V_{2\omega, \perp} = -\frac{1}{2} I_{ac} R_{AHE} \Delta\theta \cos \theta + I_{ac} R_{PHE} \Delta\phi \cos^2 \theta \quad (\text{H in } y\text{-}z \text{ plane, } \phi = 90^\circ), \quad (4.3)$$

where R_{AHE} and R_{PHE} is the anomalous Hall and planar Hall resistances respectively, $\Delta\theta$ and $\Delta\phi$ are magnetisation oscillations due to the SOT.

The magnetisation oscillations, $\Delta\theta$ and $\Delta\phi$, were determined at the stable state through the partial derivative of the total energy and can be written as,

$$\Delta\theta_L = -\frac{H_L}{\sqrt{H_k^2 - H_x^2}} \quad ; \quad \Delta\phi_L = -\frac{H_T}{H_x} \quad (\text{Longitudinal configuration}), \quad (4.4)$$

$$\Delta\theta_T = -\frac{H_L}{\sqrt{H_k^2 - H_y^2}} \quad ; \quad \Delta\phi_T = -\frac{H_L}{H_y} \quad (\text{Transverse configuration}), \quad (4.5)$$

where H_L is the longitudinal effective field, H_T the transverse effective field, H_x and H_y are the external magnetic fields acting on the system along the x and y -direction respectively, and H_k is the effective anisotropic field.

A relationship between V_H and $H_{x(y)}$ was determined using (4.2) to (4.5). The harmonic voltage relation with H_x and H_y dependence can be re-expressed as,

$$V_{2\omega, //} = -\frac{I_{ac} R_{AHE} H_L}{2H_k} \frac{H_x}{\sqrt{H_k^2 - H_x^2}} + \frac{I_{ac} R_{PHE} H_L H_x}{H_k^2}, \quad (4.6)$$

$$V_{2\omega, \perp} = -\frac{I_{ac} R_{AHE} H_T}{2H_k} \frac{H_y}{\sqrt{H_k^2 - H_y^2}} - \frac{I_{ac} R_{PHE} H_L H_y}{H_k^2}, \quad (4.7)$$

The field-dependent harmonic relation in (4.6) and (4.7) were used to fit the experimental 2nd harmonic signal to determine the effective fields H_L and H_T in both measurement schemes. The above relation assumes that H_L and H_T are constants and independent on magnetisation directions.

4.3 Methods

The film consisting of Ta(2nm)/MgO(2nm)/CoFeB (1.2nm)/Ta(5nm) was deposited on a thermally oxidised SiO_x(300nm) Si substrate via ultra-high vacuum magnetron sputtering. Post-deposition annealing at 200°C was conducted on the film for 2 hours to increase the strength of the perpendicular magnetic anisotropy (PMA). Annealing facilitates the (001) crystalline orientation in the CoFeB layer due to the reorientation of the MgO layer, while Ta provides a sink for the diffusion of B atoms.¹²⁸ The film was measured to have high resistivity (181 $\mu\Omega\text{cm}$), which indicates that Ta largely grows in the β -phase, which is commonly reported to have large spin Hall angle.³⁶ The film was patterned into a symmetrical Hall cross with a width of 5 μm using EBL and ion milling techniques. The magnetic properties of the film were determined using vibrating sample magnetometer with an applied field-oriented out-of-plane (OOP) and in-plane (IP) to the film plane. The film has saturation magnetisation $M_s = 1.4 \times 10^6$ A/m and effective anisotropic field, $H_k = 3.84$ kOe.

Figure 4.1 Schematic of longitudinal and transverse measurement scheme for the symmetrical Hall cross in cartesian coordinate geometry indicating the $H_{x(y)}$ sweep, effective fields, H_L and H_T with respect to the in-plane current for a MgO/CoFeB/Ta nanowire.

Figure 4.2 (a) MagVision Kerr in-situ image of out-of-plane (OOP) magnetic field sweep captured at each individual magnetic field. (b) Anomalous Hall resistance R_{AHE} as a function of H applied in the out-of-plane direction at 300K.

Figure 4.3. (a) Dendritic domain width (up and down domains) variation as a function of applied OOP field, with differential Kerr image of magnetic dendritic domains of the film stack [Ta(2nm)/MgO(2nm)/CoFeB (1.2nm)/Ta(5nm)] in a $5\mu\text{m}$ width Hall cross in demagnetised state and approaching saturation state (inset). The error bars denote the standard deviation from 10 individual domain width determinations. (b) Current-induced switching of the film stack at fixed IP-field of 240 Oe and 270 Oe.

4.4 Results & Discussions

MagVision Kerr imaging system^{129–131} was used to observe the magnetisation behaviour of the thin film and the Hall cross *in-situ*. The measurement schematic is illustrated in Figure 4.1. The magnetisation reversal of the Hall cross was observed to be initiated by nucleation at specific sites, followed by dendritic growth and expansion of domains. The unreversed magnetisation between dendritic branches diminishes until an additional field was applied to overcome the energy barrier, thus leading to a non-sharp switching before reaching saturation.³⁸ The anomalous Hall effect (AHE)¹³² measurement in Figure 4.2 shows that the Hall cross patterned film structure has coercitivity, $H_c = 25$ Oe, and anomalous Hall resistance, $R_{AHE} = 0.68 \Omega$, when the field was swept along the out-of-plane direction with a non-perturbing measurement current density of 7.5×10^4 A/cm², forming dendritic like domains followed by domain expansion till saturation.

The domain width of the demagnetised state of the sample was analysed using the Kerr microscope. An OOP magnetic field of 200 Oe was applied to saturate the sample, and then the applied field was gradually decreased until a demagnetised state showing dendritic domains, were observed as shown in the inset of Figure 4.3(a). Subsequently, the dendritic domain width was measured as a function of the OOP field, as shown in Figure 4.3(a). As the field increased in the positive direction, the width of the up domains (black) expanded until the down domains (white) was annihilated near saturation. In earlier reports, the maximum field in which the domains still exist can be used to determine the Dzyaloshinskii-Moriya interaction (DMI)^{133,134}. The field-induced evolution of the dendritic domains led to a hysteresis like curve in Figure 4.1(b). A hyperbolic tangent function, $w(H) = a \cdot \tanh(\delta \cdot H + \phi) + d$ ^{135,136}, was utilised to determine the terminal width, $w_{term} = |d - a| = 700 \pm 80$ nm, and the average periodicity, $w_{average} = 1200 \pm 80$ nm, just before the annihilation of the dendrites. In a multi-domain state, the domain width provides a means of quantifying the DMI through established models¹³⁷.

Utilising the equations (4.8) to (4.10)^{133,135},

$$\frac{\sigma_{DW}}{\mu_0 M_s^2 t} = \frac{W_{av}^2}{t^2} \sum_{odd\ n=1}^{\infty} \frac{1}{(\pi n)^3} \left[1 - \left(1 - \frac{2\pi n t}{W_{average}} \right) \exp \left(-\frac{2\pi n t}{W_{average}} \right) \right], \quad (4.8)$$

$$\frac{2\pi\sigma_{DW}}{\mu_0 M_s^2 t} = \ln \left[1 + \left(\frac{W_{term}}{t} \right)^2 + \left(\frac{W_{term}}{t} \right)^2 \ln \left[1 + \left(\frac{W_{term}}{t} \right)^{-2} \right] \right], \quad (4.9)$$

$$|D| = \frac{1}{\pi} \left[4\sqrt{AK_{eff}} - \sigma_{DW} \right], \quad (4.10)$$

where σ_{DW} is the DW energy, M_s is the saturation magnetisation, t is the thickness of the film, A is the exchange constant, K_{eff} is the effective anisotropic energy and D is the DMI constant. In the high-field case, when the antiparallel domains approach a terminal width just before annihilation, (4.9) was used to determine the σ_{DW} and subsequently DMI constant, $|D| = 0.65 \pm 0.08$ mJ/m². The presence of DMI enabled the stabilisation of the dendritic domains in the demagnetised state. The $|D|$ was determined throughout the regions with dendritic domains, inclusive of pinning sites within the hall wire which gave rise to the DMI error. The reported DMI was more significant than other reports which observed normal bubble domains at around $D \approx 0.05 - 0.1$ mJ/m², rather than dendritic-like magnetisation texture as observed in our MgO/CoFeB/Ta structure^{138,139}. The larger DMI present in this stack can lead to the stabilisation of the dendritic-like domains. This was affirmed through the comparable domain width in both, the experimental and simulated, cases as elaborated below.

MuMax³ micromagnetic simulation¹⁴⁰ was conducted to verify that the dendritic domains are related to the DMI. The relaxed demagnetised state of the Hall bar was simulated using DMI values ranging from 0 to 2.9 mJ/m² in steps of 0.1. In the numerical simulations, the device has geometrical dimensions of 5 μ m width $\times$ 10 μ m length, K_u of 2.72×10^5 J/m³, M_s of 1.4×10^6 A/m, exchange stiffness, A_{ex} of 2×10^{-11} J/m⁵⁶. The domain width spacing as a function of DMI was determined, as shown in Figure 4.4(a). At $D = 0$ mJ/m², the sample formed larger sized domains, and with increasing

value of D , the domains shrunk exponentially in size. For an experimental domain width of 1130 ± 40 nm, the simulated $|D|$ corresponding to the domain width is 0.6 mJ/m². This simulated result coincides with the experimentally determined DMI, via the method described earlier in this chapter. This affirms that the presence of DMI in MgO/CoFeB/Ta was responsible for stabilising the dendritic domains in the CoFeB layer^{42,141}.

Figure 4.4. (a) Domain width as a function of DMI strength, $|D|$ from numerical simulations with inset snapshots of the dendritic domains at various values of $|D|$ and the in-situ Kerr image of at the centre of the Hall cross in the demagnetised state. Observation of the (b) field-induced nucleation of dendritic domains when a pulsed-field of 1.2 kOe was applied, and (c) domain-creep when $I_{ac} = 1 \times 10^{11}$ A/m² was applied in the (i) x-direction (ii) and y-direction, respectively.

To ensure comparability with spin-transfer torque (STT) critical current densities, current pulses were used to induce magnetisation switching in the Hall cross with an applied IP field of 240 Oe as shown in Figure 4.3(b). The critical switching current was 1.5 mA (1.35×10^6 A/m²) which is comparable to the STT critical switching current¹⁴². The SO effective fields were subsequently quantified through *in-situ* Kerr harmonic measurements, and these schemes of measurement are illustrated in Figure 4.1.

Before the addition of the sinusoidal alternating current, field-induced and current-induced nucleation, were observed separately, as shown in Figure 4.4(b) and (c), respectively. The nucleation of dendritic domains appeared spontaneously at various spots on the Hall cross as the IP-field was swept through zero. The DMI in the sample manifested itself by stabilising the dendritic texture in the Hall cross¹⁴³. Furthermore, the DMI led to the magnetisation tilt at the edges, leading to edge nucleation and expansion in the Hall cross-device. At 1.2 kOe pulsed IP-field, the dendritic domains formed are as shown in Figure 4.4(b) for the sample with film H_k of 3.84kOe. The device H_k behaved similarly to the thin film, which is higher than the domain nucleation occurring near 1.2 kOe. When I_{AC} was applied in the x and y regime without an IP-field for a period of 5 minutes, current-induced domain nucleation occurred and the domain expanded in an elongated manner near the contact pads in the x and y -direction as shown in Figure 4.4(c). This current-induced nucleation was due to the thermally-assisted spin Hall torque on the magnetisation in the Hall cross.

Figure 4.5. *In-situ Kerr observation of the progressive nucleation and expansion of dendritic domains, when an in-plane magnetic field is applied during the longitudinal, and transverse harmonic measurement scheme observed under the MagVision Kerr microscope.*

Figure 4.5 showed the snapshots of the Hall cross magnetisation behaviour at the different applied IP magnetic field when a current was applied to the sample. The current

dependence of the effective field was determined by varying the sinusoidal input current I_{AC} with reference frequency 307.1 Hz. The current densities were modulated with a step size of $1 \times 10^{10} \text{ A/m}^2$, from $1 \times 10^{10} \text{ A/m}^2$ to $1 \times 10^{11} \text{ A/m}^2$. The first and second harmonic voltages (V_{ω} and $V_{2\omega}$ respectively) were measured simultaneously and averaged over 120 readings per data point for better sound to noise ratio (SNR). Two measurement schemes, i.e., longitudinal measurement where applied current and field are in the same direction and transverse measurement where applied current and field, are orthogonal to each other, were performed separately. The observation in Figure 4.5 proves that the hysteresis behaviour observed in the first harmonic voltage was due to the partial switching of the CoFeB layer, and the dendritic domains evolve with respect to the applied in-plane field.

Figure 4.6. *In-situ harmonic Hall Measurement. Current dependence in the longitudinal measurement scheme of the (a) 1st harmonic and the peak shift of the second harmonics as a function of the applied current density (inset) and (b) 2nd harmonic measured at $\theta_H = 4^\circ$ at 300K. Current dependence in the transverse measurement scheme of the (c) 1st harmonic and (d) 2nd harmonic measured at $\theta_H = 4^\circ$ at 300K and the domain width as a function of the applied field before the complete annihilation of the dendritic domains (inset). Domain structure at different field values is shown on the 1st harmonics for both the longitudinal and transverse measurement schemes.*

The current dependence of the harmonic results for both longitudinal and transverse measurements are shown in Figure 4.6 (a) to (d). In the longitudinal measurement, the 1st harmonic signals show hysteresis like loops, indicating the presence of domain nucleation and expansion. At low applied IP-field, a small number of existing domains were randomly moving and were spread throughout the Hall cross. The 2nd harmonic signals reveal asymmetric wave-like patterns about the vertical axes. The rise to the peak, $V_{2\omega, peak}$ is characterised by the formation of dendritic domains as observed in the 1st harmonic signal. The corresponding field, H_{peak} at the asymmetric peaks, $V_{2\omega, peak}$ is related to the effective anisotropy field, H_k of CoFeB and the effective fields acting on the CoFeB. Such that, a higher H_k corresponds to a higher H_{peak} . With the increase of the current density, the H_{peak} decreases monotonically with an applied current density, as shown in Figure 4.6(a) inset. This occurs as the applied current coupled with the external in-plane field, facilitated the depinning of the domains at various sites, thereby favouring domain expansion. As current increases, energy is supplied to overcome the pinning field. Hence an inward shift was observed in the H_{peak} as illustrated by the dotted line in Figure 4.6(b). In the transverse measurement scheme, a symmetrical dip about the vertical axis was observed and flattens out when the magnetisation state changes. Similar nucleation and expansion of the dendritic domains were observed in the 1st harmonic, as shown in Figure 4.6(c). The slope of the transverse curve rises until saturation, indicating the expansion of the domains as the in-plane field increases. In both measurement schemes, the measured domain width for both are shown in Figure 4.6(d) inset. The up magnetised domains (black) increased in width as the field was swept in the positive x -direction and the down magnetised domains (white) increased in width as the field was swept in the opposite direction. The change in domain width for both field sweep direction corresponds to 1100 ± 300 nm.

Figure 4.7. (a) Experimental fitting of the second harmonic voltages shown together with the first harmonic signal in the *longitudinal* measurement scheme. The fitted H_L and H_T are calculated from the fitting (red curve). (b) Current dependence of the longitudinal spin orbit effective field following the range of current density values for the *longitudinal* measurement scheme, (c) Experimental fitting of the second harmonic voltages shown together with the first harmonic signal in the *transverse* measurement scheme. The fitted H_L and H_T are calculated from the fitting (red curve). (d) Current dependence of the transverse spin orbit effective field following the range of current density values for the *transverse* measurement scheme

Assuming H_L and H_T are constants, the derived equation was used to fit the experimental results within $-1.1 \text{ kOe} \leq H_{x(y)} \leq 1.1 \text{ kOe}$. The device has known parameters, $R_{AHE} = 1.35 \text{ } \Omega$ and $R_{PHE} = 0.97 \text{ } \Omega$. Equations [4.6] and [4.7] were used to fit the curve as shown in Figure 4.7 (a) and Figure 4.7(c) for longitudinal and transverse measurement schemes respectively by utilising the 2nd harmonic signal. The effective field in the longitudinal scheme was determined to be $H_L = -12 \text{ Oe}$ and $H_T = 8 \text{ Oe}$ per 10^6 A/cm^2 with $H_k = 3.84 \text{ kOe}$. In the transverse scheme, $H_L = -12 \text{ Oe}$ and $H_T = -17 \text{ Oe}$ per 10^6 A/cm^2 . The inclusion of the PHE was necessary to attain a sufficiently accurate value of H_L and H_T . The values determined were larger than earlier reports. Several reports had shown that the H_L arises due to the spin Hall effect^{39,53,144} while H_T arises due to the Rashba effect^{37,49,117,120,121}. Assuming that the spin Hall effect contributed significantly to H_L , the parameters θ_{SH} and H_L can be related by, $H_L = \hbar \theta_{SH} |j_e| / 2 |e| M_s t_f$, where $M_s = 1.4 \times 10^6 \text{ A/m}$, t_f is the ferromagnetic thickness. The spin Hall angle was determined to be $\theta_{SH}^{Ta} \approx -0.14 \pm 0.05$ and is similar to earlier reports at 300K³⁶. The current-induced effective fields are strongly correlated to the current density, as indicated in Figure 4.7(c) and (d). The longitudinal effective field, H_L , in both the longitudinal and transverse measurement scheme shows clear monotonic behaviour with increasing current density. This is mainly caused by the spin Hall effect^{55,145}. Since the determination of H_T was through the PHE term in the derived equation, we can conclude that significant PHE was present in CoFeB (R_{PHE}) that gave rise to a large coexistence of the H_T . The magnitude of H_T is generally smaller than H_L , as the bulk SHE dominates in the longitudinal measurement scheme.

4.5 Summary

In summary, dendritic domain expansion was observed in the in-plane harmonic Hall measurement via *in-situ* Kerr imaging. Interfacial DMI in the CoFeB/Ta layer was responsible for the domain dynamics during the measurement, which was affirmed through micromagnetic simulations. At low frequency, the coexisting H_L and H_T at varying current densities were $H_L = -12$ Oe and $H_T = 8$ Oe per 10^6 A/cm² for the longitudinal scheme and $H_L = -12$ Oe and $H_T = -17$ Oe at 10^6 A/cm² for the transverse scheme. While the spin Hall angle, $\theta_{SH}^{Ta} \approx -0.14 \pm 0.05$. Due to the substantial PHE contribution, the perpendicularly magnetised MgO/CoFeB/Ta film exhibited the coexistence of H_L and H_T . The results reveal that the magnetisation reversal is achieved via the formation and expansion of dendritic domain, and is responsible for the effective fields determined.

Chapter 5: Enhanced Spin-orbit Torques in Rare Earth Pt/(CoNi)₂/Tb systems

Abstract

The enhanced spin-orbit torque (SOT) effect due to the rare-earth metal Tb in a Pt/[Co/Ni]₂/Co/Tb multilayer with perpendicular magnetic anisotropy (PMA) was reported. Due to the large spin-orbit coupling in Tb, the effective damping like efficiency, ξ_{DL}^{eff} , is determined to be 0.55 for 9-nm Tb as compared to $\xi_{DL}^{eff} = 0.18$ for the reference Pt/[Co/Ni]₂/Co/Ta stack. A relatively sizeable effective spin Hall efficiency of Tb, $\theta_{SH,Tb}^{eff}$, is determined to be -0.480 for Tb with a 9nm through a H_z -bias harmonic measurement. Enhanced magnetisation switching efficiency is observed with increasing Tb thickness affirming the sizable anti-damping torque responsible for deterministic switching. Furthermore, due to a more substantial interfacial coupling in Co/Tb interface than in Co/Ta interface, strong angular dependence of the damping-like and field-like terms is present but diminishes with increasing Tb thickness. These results affirmed that Tb plays a significant role in tuning the SOT efficiency in these structures as the increased Tb concentration leads to an enhancement of the effective spin Hall angle and switching efficiency.

Q. Y. Wong *et al.* , "Enhanced Spin-orbit Torques in Rare Earth Pt/[CoNi]₂/Co/Tb systems", Physical Review Applied 11, 024057 (2019).

5.1 Introduction

SOT can be expressed as the damping-like (DL)⁹⁸ and field-like (FL)⁴⁶ terms, where the DL torque originates from the SHE and is responsible for the magnetisation reversal because of its anti-damping form. The relative efficiency of the DL-torque is a commonly used figure of merit for spin Hall material that relates proportionally to the effective spin Hall angle, $\xi_{DL}^{eff} \propto \theta_{SH}^{eff}$. The ongoing studies of *5d* transition heavy metal (HM)/ferromagnetic (FM) interface has revealed strong intrinsic^{146,147} spin Hall effects in Pt ($\theta_{SH,Pt}^{eff} = +0.09$)^{36,60,127}, β -Ta ($\theta_{SH,Ta}^{eff} = -0.12$)^{36,61}, and β -W ($\theta_{SH,W}^{eff} = -0.30$)¹⁴⁸, as reported in Chapter 4. As such, we continued to pursue the search for alternate spin Hall materials from a class of comparatively unexplored elements, the *f*-electron rare-earth (RE) metals. The motivation for our work was based on the recent theoretical prediction that RE metals have substantial spin Hall effect contribution arising from considerable orbital angular momentum, *l*, and spin, *s* due to its partially filled *f*-orbitals^{146,149}. More recently, several works have examined SOT in RE ferrimagnets^{43,70,158,150–157}, which has gathered considerable attention in the field of spintronics. In Tb-based ferrimagnets, Tb was used to provide antiferromagnetic coupling to compensate the magnetisation in the Co-Tb alloys^{151,156}. Ueda *et al.*⁷⁰ have reported high spin Hall efficiency in Pt/Co/Gd due to the presence of Gd, while Reynolds *et al.*¹⁴⁹ have characterised spin Hall efficiency of RE metals such as Gd, Dy, Ho, Lu as the contributing interfacial layer on devices with in-plane magnetic anisotropy. Within the period of the Lanthanide series, Tb lies between Gd and Dy and have *4f*-level electronic configurations which contributed to the enhancement of the spin Hall effect.

Furthermore, Tb is considered chemically stable with partially filled *f*-shells with spin Hall enhancing potential but remains unexplored in terms of its spin Hall efficiency. Due to its similarity with other RE metals explored earlier, it is intuitive that Tb is likely to enhance the SOTs. Bang *et al.*¹⁵⁰ have reported large interfacial SOT due to the Co/Tb interfaces in a Co/Tb multilayer structure from enhanced DW speed due to an inner interfacial effect caused by Tb induced skew scattering. In our study, the heterostructure of Pt/FM/Tb allows Tb to serve as a heavy metal layer replacement for

the enhancement of the SOT while enabling the quantification of the effective spin Hall or dampinglike efficiency in Tb. As such, Pt/FM/Tb structures should be explored further to determine its spin Hall efficiency.

In this chapter of this thesis, enhanced SOTs were generated by the Co/Tb interface in a Pt/[Co/Ni]₂/Co/Tb system with perpendicular magnetic anisotropy (PMA), and strong azimuthal angle dependence via systematic variation of Tb thickness from 5 to 9 nm are reported. The results were compared with the control (reference) sample with a Ta cap layer. The SOTs were characterised as a function of Tb thickness with a modified H_z biased harmonic measurement setup. The magnitude of enhanced SOTs was contrasted by the magnitudes of their damping-like effective field, H_{DL} , field-like effective field, H_{FL} , effective spin Hall angle, θ_{SH}^{eff} as well as the $J_{threshold}$ in the current-induced magnetisation switching. Subsequently, the azimuthal angle dependence of the Pt/[Co/Ni]₂/Co/Tb (5 - 9 nm) stacks, was also determined.

5.2 Results & Discussions

5.2.1 Thin Film Material Properties

The thin-film multilayers consisting of Ta(2nm)/Pt(6nm)-/[Co(0.3nm)/Ni(0.6nm)]₂/Co(0.3nm)/RE(5–9nm)/Ta(2nm) and reference sample Ta(2nm)/Pt(6nm)/[Co(0.3nm) /Ni(0.6nm)]₂/Co(0.3nm)/Ta(5nm) were deposited onto thermally oxidised Si substrates using ultra-high vacuum d.c. magnetron sputtering, as shown schematically in Figure 5.1(a). Ta was used both as a bottom seed layer and capping layer to prevent oxidation of the Tb layer. Pt serves as an underlayer with (111) crystal orientation to promote PMA in the Co layer^{129,130,159,160}. The deposition was carried out at room temperature under 3mTorr Xe with a base pressure of 3×10^{-8} Torr. The Tb films capped with Ta exhibited electrical resistivity, $\rho_{Tb} = 108 \mu\Omega \text{ cm}$, indicating minimal oxidation of the Tb layer¹⁶¹. Figure 5.2 (a) shows the schematic of the H_z biased harmonic Hall measurement technique to estimate the effective fields. In this work, the Co/Tb interface is contrasted to the Co/Ta interface in terms of the SOT efficiency from the estimated effective fields and the current induced magnetisation switching.

The magnetic properties of the film structure, such as saturation magnetisation, M_s and the perpendicular anisotropic energy density, K_u , are determined from the vibrating sample magnetometer (VSM) measurements as shown in Figure 5.2(b). K_u can be expressed as $K_u = H_k M_s / 2$ and $H_k = H_{sat} + 4\pi M_s$, where H_k is the anisotropy field and H_{sat} is the hard axis saturation field. All the films exhibited PMA induced by the bottom Pt/Co interface. As summarised in Figure 5.2(b), M_s and K_u were both reduced with increasing Tb thickness suggesting that Tb is a critical factor in achieving strong PMA. When the thickness of Tb, t_{Tb} was varied from 5nm to 9 nm, it led to a 54% reduction in the magnitudes of K_u and M_s . This suggests that the drop in the calculated K_u was significantly due to the reduction in M_s value. The reduction in M_s value could potentially be due to compensation from the ferrimagnetic properties of Tb. From the experimental data, M_s for the Co/Ni multilayer stack decreases with increasing Tb thickness from 5nm to 9 nm. The saturation magnetisation decreased from 408 emu/cm³ to 157 emu/cm³ for the film stacks with 5nm Tb and 9nm Tb, respectively. The reference stack (Co/Ta) has a saturation magnetisation of 596 emu/cm³. The decrease in the M_s and H_k can be accounted for by the intermixing at the Co/Tb interface. Tb atomic layers adjacent to the Co have non-zero magnetic moments. The non-zero magnetic moments are antiparallel to the Co/Ni layers, resulting in an anti-ferromagnetic coupling between Co and Tb. Figure 5.1 shows the vibrating sample magnetometer (VSM) measurements of the stacks.

Figure 5.1 VSM Measurements of the stack structures.

At the interface, the atomic moments of Co and Tb couples in an antiparallel arrangement such that the total net magnetisation M_s of Co/Ni/Co_{1-x}Tb_x can be expressed as:

$$M_s = (1 - x)M_{CoNiCo} - xM_{Tb}, \quad (5.1)$$

where M_{CoNiCo} and M_{Tb} are the magnetisations of the ferromagnetically coupled Co/Ni multilayer and the Tb layer, respectively, and x stands for the average atomic percentage of Tb in the entire film stack.

Taking the M_s value from the reference film stack of 596 emu/cm³, without the influence of Tb and M_{Tb} as $8.9\mu_B$,¹⁶² M_{Tb} can be determined from the atomic moment of Tb as follows, $M_{Tb} = N_A \left(\rho_{Tb} / A_{R,Tb} \right) 8.9\mu_B = 2587 \text{ emu/cm}^3$, where N_A is the Avogadro constant, ρ_{Tb} is the density of Tb, $A_{R,Tb}$ is the relative molecular mass of Tb and μ_B is the Bohr magneton respectively. The average ratio of Tb involved in the mixing increases with increasing thickness until it plateaus at 9 nm Tb. The most substantial proportion occurs at 14% when the thickness of Tb, t_{Tb} is at 9 nm. With the trend of M_s and H_k observed, this methodology is adequate to account for the pattern of a compositional gradient at the Co/Tb interface. Similarly, due to the antiferromagnetic properties of Tb in the film stacks, the observed coercivity, H_c decreases with increasing t_{Tb} .

Figure 5.2 (a) Schematic illustration of reference sample Ta/Pt/[Co/Ni]₂/Co/Ta with Tb-based sample Ta/Pt/[Co/Ni]₂/Co/RE/Ta alongside the H_z biased measurement configuration for evaluation of the SOT in cartesian coordinates. (b) Saturation magnetisation (M_s) and perpendicular magnetic anisotropic energy density (K_u) versus capping layer thickness with percentage concentration of Tb in film stack inset (c) Relation of channel Hall bar conductance and coercivity, H_c with metallic capping layer thickness, $t_{Tb/Ta}$. The red line denotes a fit of linearity. (d) Anomalous Hall resistance measurement under a H_z sweep in films with various t_{Tb} ranging from 5 nm to 9 nm and $t_{Ta} = 5$ nm under a non-perturbing measurement current density of 1×10^{10} A/m². Coercivity of the film stacks decreases with the increase of t_{Tb} .

5.2.2 Enhanced SOT Fields in Rare Earth, Tb

Next, harmonic Hall voltage measurements were conducted by detecting Hall voltage variation as a function of the tilting of the magnetisation under an in-plane field to quantify SOT effective fields. In these measurements, symmetrical Hall bars with dimensions $5\mu\text{m}$ -width and $50\mu\text{m}$ -long were patterned using EBL and argon ion milling techniques. Electrical contact pads consisting of Ta (5nm)/Cu (100nm)/Ta(5nm) were sputtered at the ends of the Hall bars to inject low-frequency a.c. current using a Keithley 6221 a.c. current source and using a 7265 Dual-Phase DSP Lock-in Amplifier to detect the Hall voltage, V_H , while having a constant $\pm 300\text{Oe}$ H_z field as shown in the measurement geometry in Figure 5.2(a). The presence of the H_z field enables the magnetisation to remain at small polar angles for effective quantification of the SOT fields. The derivation of the modified harmonic measurements is found in the Supplemental section at the end of the chapter. Figure 5.2 (c) indicates the relation between the coercivity, H_c and capping layer thickness of Tb and Ta, while Figure 5.2 (d) shows the anomalous Hall loops when the films were swept under a H_z field for various t_{Tb} ranging from 5nm to 9 nm. The quantification of Tb resistivity ρ_{Tb} was done by measuring the resistance of the Hall bar devices with various t_{Tb} and their conductance is plotted as a function of t_{Tb} as shown in the top graph of Figure 5.2 (c). The inverse of the gradient corresponds to $\rho_{Tb} = 108 \pm 2 \mu\Omega\text{cm}$ while the vertical intercept corresponds to the conductance of the remaining part of the film stack. The current densities that flow through the HM and rare earth metal (RE) were accounted for by assuming the parallel resistance model of current distribution within the Hall cross-device, through the ratio of resistivities.

Longitudinal Measurement Scheme

Transverse Measurement Scheme

Figure 5.3 (a) First harmonic voltage, V_{ω} and (b) second harmonic voltage, $V_{2\omega}$ as a function of H_x in the longitudinal measurement scheme. (c) and (d) as a function of H_y in the transverse measurement scheme. Measurement geometry of both measurement schemes is illustrated on the left of each corresponding plot.

SOT harmonic measurements were carried out by passing a.c. current I_{ac} along the x-direction with frequency, $\omega = 333$ Hz, in the presence of an additional H_z biased field. The voltage measurement was recorded in two measurement schemes: the longitudinal and transverse schemes. The first and second harmonic, V_{ω} and $V_{2\omega}$, respectively, were concurrently measured from the Hall voltage using a lock-in amplifier and recorded as a function of the applied field H_x (H_y) as shown in Figure 5.3. The modulation amplitudes of the polar, $\Delta\theta$ and azimuthal axes, $\Delta\phi$, for a H_z biased measurement were determined to be (See detailed derivation in Supplemental section at the end of this chapter):

$$\Delta\theta = \frac{(\Delta H_x \cos \phi + \Delta H_y \sin \phi)}{(H_{k,eff} + H_z)}, \quad (5.2)$$

$$\Delta\phi = \frac{\Delta H_y H_x - \Delta H_x H_y}{H_x^2 + H_y^2}, \quad (5.3)$$

where $H_{k,eff}$ is the effective anisotropic field of the material stack, H_z is the magnitude of the biased field, $\Delta H_x = H_{DL}$ and $\Delta H_y = H_{FL}$, and the H_x and H_y correspond to the in-plane field sweep in the harmonic measurement.

The coefficients of the first and second harmonics are expressed as:

$$V_\omega = \frac{1}{2} I_{ac} [\Delta R_{AHE} \cos \theta + \Delta R_{PHE} \sin^2 \theta \sin 2\phi], \quad (5.4)$$

$$V_{2\omega} = -\frac{1}{2} I_{ac} \left(\frac{1}{2} [-\Delta R_{AHE} \sin \theta + \Delta R_{PHE} \sin 2\theta \sin 2\phi] \Delta \theta + \Delta R_{PHE} \sin^2 \theta \cos 2\phi \Delta \phi \right), \quad (5.5)$$

where ΔR_{AHE} is the change in anomalous Hall resistance, ΔR_{PHE} is the change in planar Hall resistance and I_{ac} represents the current in the wire, by substituting (5.2) and (5.3) into (5.5), the H_{DL} and H_{FL} can be estimated by utilising the following relation by eliminating the $(H_{k,eff} + H_z)$ and I_{ac} terms:

$$H_{DL(FL)} = -2 \left(\frac{dV_{2\omega}}{dH_{x(y)}} \right) / \left(\frac{d^2 V_\omega}{dH_{x(y)}^2} \right), \quad (5.6)$$

Figures 5.3(a) and 5.3(b) show V_ω and $V_{2\omega}$ as a function of H_x for $t_{Tb} = 5nm$ with initial magnetisation state set to be $+M_z$ and $-M_z$, while Figures 5.3(c) and 5.3(d) show V_ω and $V_{2\omega}$ as a function of H_y . $V_{2\omega}$ varies linearly for both H_x and H_y with similar slopes for H_x and reversed for H_y when the initial magnetisation state was reversed, which was the expected behaviour from the H_{DL} and H_{FL} contribution^{47,56,60,70,141}. The effects of Joule heating were negligible since the effective fields determined vary linearly with the input current density. The sign of H_{DL} is M_z dependent while the H_{FL} is independent on M_z , as previously reported^{53,141}.

To take into account the planar Hall effect component, the PHE correction factor, $\zeta = R_{PHE}/R_{AHE}$ can be used to correct the H_{DL} and H_{FL} given by the following relations⁵⁷:

$$H_{DL, corrected} = \frac{H_{DL} + 2\zeta H_{FL}}{1 - 4\zeta^2}, \quad (5.7)$$

$$H_{FL, corrected} = \frac{H_{FL} + 2\zeta H_{DL}}{1 - 4\zeta^2}, \quad (5.8)$$

The PHE corrected longitudinal ($H_{DL, corrected}$) and transverse ($H_{FL, corrected}$) fields, together with the H_{DL} and H_{FL} are plotted as a function of current density in Figure 5.4(a). The PHE correction factor, ζ , is determined via the relation related to the R_{AHE} :

$$\zeta = \frac{1}{2} \frac{(\partial^2 V_{\omega} / \partial H)_{\phi=-45^\circ} - (\partial^2 V_{\omega} / \partial H)_{\phi=45^\circ}}{(\partial^2 V_{\omega} / \partial H)_{\phi=-45^\circ} + (\partial^2 V_{\omega} / \partial H)_{\phi=45^\circ}} \quad (5.9)$$

The values of the PHE correction for each heavy metal or rare earth metal thickness $5 \text{ nm} \leq t_{Tb} \leq 9 \text{ nm}$ were calculated based on (5.9). The corrected values of the DL-SOT and FL-SOT fields are as shown in Figure 5.4 (a). $H_{DL, corrected}$ are contrasted with H_{DL} , and $H_{FL, corrected}$ are compared to the H_{FL} for $t_{Tb} = 5 \text{ nm}$. Here forth, for simplicity, the corrected fields are named, H_{DL} and H_{FL} . The corresponding PHE correction values are shown in Table 5.1.

Thicknesses (nm)	R_{AHE} / Ω	$\zeta = R_{PHE} / R_{AHE}$
Tb 5	0.113	0.220
6	0.136	0.324
7	0.120	0.243
8	0.112	0.327
9	0.080	0.053
Ta 5	0.148	0.318

Table 5.1. *Thicknesses of Tb and Ta and their corresponding PHE correction factor.*

The current induced effective fields H_{DL} and H_{FL} increase linearly with the input J_{ac} as shown in Figure 5.4(b) and the effective fields per unit current density relation

with capping layer thickness are shown in Figure 5.4(c). $|H_{DL}/J_{ac}|$ and $|H_{FL}/J_{ac}|$ for the Tb series of films have significantly larger induced effective fields as compared to the Ta capping. $|H_{DL}/J_{ac}|$ increases considerably with increasing Tb thickness from ~10 Oe per 10^{10}A/m^2 for $t_{Tb} = 5\text{ nm}$ to ~51 Oe per 10^{10}A/m^2 for $t_{Tb} = 9\text{ nm}$. The calculations were corrected based on the current distributions in the film stack, assuming that, the Pt, Co, Ni and Tb (Ta) layers act as resistances in parallel, such that the current flowing in each layer was distributed according to the resistivities. As the rest of the structure remains congruent throughout, the increase in current in Tb is the sole cause of the rise and is invariant with Pt thickness. Therefore, since $|H_{DL}/J_{ac}|$ increases with t_{Tb} as shown in Figure 5.4(c), it is inferable that the Tb metal at the top interface produces a DL-SOT that acts on the ferromagnetically coupled Co/Ni multilayer, since the t_{Pt} remains the same for all film stacks. The SOT efficiency can be expressed as, $\xi_{DL(FL)}^{eff} = 2eH_{DL(FL)}M_s t_{FM}/\hbar J_{ac}$, where $\xi_{DL(FL)}^{eff}$ is the SOT efficiency, $H_{DL(FL)}$ is the effective field, J_{ac} is the input a.c. current density, M_s is the saturation magnetisation and t_{FM} is the effective thickness of the ferromagnetic layer. The enhancement of the $|H_{DL}/J_{ac}|$ is likely due to Tb having $\xi_{SH,Tb}^{eff}$ that is the opposite sign to Pt, such that the top and bottom interfaces have an additive effect which gives rise to an effective damping-like efficiency, ξ_{DL}^{eff} . Also, in Figure 5.4 (d), $|H_{FL}/J_{ac}|$ as a function of t_{Tb} shows that with increasing t_{Tb} , the FL term increases systematically with a factor of 2.3, indicating the enhancement of FL-SOT caused by the presence of Tb. $|H_{FL}/J_{ac}|$ increases from ~2 Oe per 10^{10}A/m^2 for $t_{Tb} = 5\text{ nm}$ to ~13 Oe per 10^{10}A/m^2 for $t_{Tb} = 9\text{ nm}$. The FL-efficiency calculated was for $t_{Tb} = 5\text{ nm}$ to be $|\xi_{FL}^{eff}| = 0.12$ and for $t_{Tb} = 9\text{ nm}$ to be $|\xi_{FL}^{eff}| = 0.05$. This shows that even though the magnitude of the FL-term increases with Tb thickness, the overall efficiency of the FL-term on the FM layer diminishes, due to the M_s . The DL-efficiency, $|\xi_{DL}^{eff}|$ was approximately twice the FL-efficiency, $|\xi_{FL}^{eff}|$, for $t_{Tb} = 5\text{ nm}$ and this ratio of $|\xi_{DL}^{eff}|/|\xi_{FL}^{eff}|$ increases to approximately one order of magnitude, i.e. ten times.

Therefore, these results indicate that SHE is likely the more dominant mechanism in the studied stack variation^{163,164}.

Figure 5.4 (a) H_{DL} and $H_{DL,corrected}$, H_{FL} and $H_{FL,corrected}$ for a film stack with $t_{Tb} = 5 \text{ nm}$ as a function of the applied current density determined from the longitudinal and transverse harmonic measurement scheme, respectively. (b) H_{DL} and H_{FL} as a function of applied current density for various capping layer thickness for Tb (5 nm to 9 nm) and Ta (5 nm). (c) The absolute value of the effective fields per unit 10^{10} A/m^2 for $|H_{DL}/J_{ac}|$ and $|H_{FL}/J_{ac}|$ (d) Relation of SOT efficiency, $\xi_{SH}^{Pt-HM(RE)}$ with a thickness of Ta and Tb.

In this SHE dominant system, we assume this mechanism is responsible for the DL-term such that, the ξ_{DL}^{eff} relates directly to the effective spin Hall angle of the Pt-Tb system, θ_{SH}^{eff} , $\theta_{SH}^{eff} \approx \xi_{DL}^{eff}$. As such, the magnitude of the effective spin Hall angle θ_{SH}^{eff} was determined to be ranging from $|\theta_{SH,Ta}^{eff}| \sim 0.18 \pm 0.01$ for $t_{Ta} = 5 \text{ nm}$ capping layer, $|\theta_{SH,Tb}^{eff}| \sim 0.25 \pm 0.01$ for $t_{Tb} = 5 \text{ nm}$ to $|\theta_{SH,Tb}^{eff}| \sim 0.55 \pm 0.01$ for $t_{Tb} = 9 \text{ nm}$, as shown in Figure 5.4(d). In this film stack, the growth of the Pt/Co interface not only serves as an underlying surface that promotes PMA, but it also affects the spin-mixing conductance as there may be intermixing and disorder at the Pt/Co interface¹⁶⁵. The $\theta_{SH,Tb}^{eff}$ increases

with the increasing influence of Tb. At larger t_{Tb} , the shunting of current in Pt significantly reduces, as such, this increases the contributing SHE from the Co/Tb interface. Therefore, taking the spin Hall angle of Pt, $\theta_{SH,Pt}^{eff} \approx +0.07$ from known literature^{36,60,127}, the reference sample gives rise to $\theta_{SH,Ta}^{eff} \approx -0.11$, which is within a reasonable range of the spin Hall angle reported for Ta^{36,61}. Similarly, the spin Hall angle of Tb, $\theta_{SH,Tb}^{eff}$ can subsequently be derived from the ξ_{DL}^{eff} such that its value ranges from $-0.18 \leq \theta_{SH,Tb}^{eff} \leq -0.48$ for $5 \leq t_{Tb} \leq 9$ nm.

5.2.3 Current-Induced Magnetisation Switching in Pt/[Co/Ni]₂/Co/Tb (5-9nm) and reference sample Pt/[Co/Ni]₂/Co/Ta (5nm)

Current-induced magnetisation switching measurements were performed on the Hall cross structure. As expected, when a charge current passes through, a non-zero SOT is present due to the lack of inversion symmetry in Pt/[Co/Ni]₂/Co/Tb systems. The function of Hall resistance, R_H against an in-plane d.c. current, I_{dc} was determined through a constant in-plane magnetic field, H_x , applied along the x-axis to overcome the rotational symmetry and assist in the magnetisation reversal. 10ms current pulses were swept with varying magnitudes from – 25 mA to +25 mA, and the R-I loops observed reveal the change between two resistance states. The magnitude of change in resistance agrees with the observation in the $R-H_z$ loops, indicating deterministic magnetisation reversal between the $\pm z$ directions due to the current pulse. Figure 5.5 shows the determination of the threshold current density, $J_{threshold}$, which can be further illustrated in a single R-I loop switching. Here, $J_{threshold}$ is defined as the threshold current density at which magnetisation switching originates that corresponds to an initial abrupt jump in the resistance of the RI loop. As the DC pulse current was flown through the sample, it led to the exposure of nucleation sites and domain expansion which contributed to magnetisation switching.

Figure 5.5 *Current-induced magnetisation switching curve.*

Current-induced switching measurements were conducted for all samples in the study, and the consolidated R_H vs I_{dc} loops are as shown in Figure 5.6.

Figure 5.6 R_H vs I_{dc} Loop for Hall cross structures with varying top layer thicknesses.

As shown in Figure 5.6, the switching direction changes with the direction of the in-plane magnetic field; a clockwise loop was obtained for $-H_x$, and an anti-clockwise loop was acquired for $+H_x$. This indicates the Slonczewski-like symmetry induces switching that is determined by the direction of H_x . Due to the sizable H_{DL} , SOT mainly originates from the SHE in both the Pt and Ta or Tb channels. When $H_x = 0$, magnetisation switching diminishes, as there was insufficient tilt of the magnetisation, and cannot assist in SOT switching for both $t_{Ta} = 5$ nm and $t_{Tb} = 5$ nm, as shown in Figure 5.7 (a). For the Tb sample, the magnetic moments of the FM layer can be switched with low H_x , such that $H_x < 100$ Oe. The deterministic change of magnetisation polarity in the device reveals the substantial anti-damping torque enhanced by Tb. The corresponding phase diagram of the various Tb thicknesses shown in Figure 5.7(b) illustrates the behaviour of threshold switching current density, $J_{threshold}$ in relation to the thickness of Tb and H_x . In this SOT regime, the change of resistance in the RI loop is comparable to the AHE measurement, demonstrating the deterministic switching of Tb based Co/Ni systems are possible and effective. At $H_x = 100$ Oe, the reference sample was unable to switch magnetisation polarity, while the $t_{Tb} = 5$ nm sample was able to effectively switch at least 60% of the entire magnetisation of the Hall cross. The results affirm that the Co/Tb interface was indeed responsible for the enhanced SOT generation due to the presence of a sizable anti-damping torque. This is consistent with the earlier characterised H_{DL} for the reference and the Tb samples, where the damping-like field for the $t_{Tb} = 5$ nm is twice that in the magnitude than $t_{Ta} = 5$ nm. For various Tb thicknesses, the $J_{threshold}$ phase diagram shown in Figure 5.7(b) has an optimum magnitude of $J_{threshold}$ in the blue and purple regions.

Figure 5.7 (a) Current-induced magnetisation switching curves for the reference sample, $t_{Ta} = 5$ nm and Tb-based sample, $t_{Tb} = 5$ nm structure measured with different in-plane external magnetic fields H_x . (b) Switching phase diagram for the film stacks variation from $5 \text{ nm} \leq t_{Tb} \leq 9 \text{ nm}$ film stacks, where $J_{threshold}$ is the minimum current density required for magnetisation switching to occur.

These current densities in the area correspond to equal orders of magnitude for the spin-transfer torque (STT) switching mechanism¹⁴². For low in-plane bias field, the optimum Tb thicknesses for effective switching varies between 7 nm to 9 nm, with applied in-plane biased fields, $H_x < 1000$ Oe. The Tb based devices are capable of deterministic switching with relatively small in-plane bias field along with low $J_{threshold}$ as compared to the reference sample.

5.2.4 Angular dependence of SOT fields in Pt/[Co/Ni]₂/Co/Tb (5–9 nm) and reference sample Pt/[Co/Ni]₂/Co/Ta (5 nm)

Angular dependence of the SOT fields was conducted to determine its relation with the azimuthal angle, $\phi_H = \phi$ between current direction and the magnetisation in Tb-based PMA samples similar to the barium titanate, BaTiO₃ (BTO) samples¹⁶⁶. The V_ω and $V_{2\omega}$ were measured at different azimuthal angle values, with magnetisation set to be in the $-z$ orientation for all measurements. The current density was set at 1×10^{11} A/m² in the wire to have a better signal-to-noise ratio without compromising on joule heating effects. At each azimuthal angle field sweep, the values, $A_\omega = \partial^2 V_\omega / \partial H^2$ and $B_{2\omega} = \partial V_{2\omega} / \partial H$ were determined, and the function of A_ω against ϕ_H and $B_{2\omega}$ against ϕ_H are as shown in Figure 5.8 (a) and (b) respectively. As shown in Figure 5.8 A_ω exhibits an evident $-\sin 2\phi$ behaviour as predicted and $B_{2\omega}$ depicts a $-\cos \phi$ trend. This trend relates itself to the initial state of magnetisation of the sample, such that the pattern will have a positive behaviour of $\sin 2\phi$ and $\cos \phi$. By eliminating the dependence on I_{ac} , H_z and $H_{k,eff}$, the ratio of $\beta_\phi = B_{2\omega} / A_\omega$ is computed and plotted in Figure 5.8 (c). A fitting function can be used to determine the mean SOT effective fields,

$$\beta_\phi = \left(-\frac{1}{2} \sin \phi + \frac{\zeta \cos \phi \cos 2\phi}{1 - 2\zeta \sin 2\phi} \right) \Delta H_{FL} + \left(-\frac{1}{2} \cos \phi - \frac{\zeta \sin \phi \cos 2\phi}{1 - 2\zeta \sin 2\phi} \right) \Delta H_{DL}, \quad (5.10)$$

$$\beta_\phi = a \Delta H_{FL} + b \Delta H_{DL}, \quad (5.11)$$

Figure 5.8 A sample of $t_{Tb} = 8$ nm magnetised in the $-z$ orientation (a) Second derivative of the parabolic first harmonic Hall voltage as a function of azimuthal angle, ϕ , with a $-\sin 2\phi$ fit. (b) The first derivative of the second harmonic Hall voltage as a function of azimuthal angle, ϕ , with a $-\cos \phi$. (c) β_ϕ as a function of the azimuthal angle, ϕ , and a fit from (5.10).

Using the fitting of (5.10) on the function of β_ϕ against ϕ on Figure 5(c), the mean values of the DL-term and FL-term were determined to be 35 ± 2 Oe per 10^{10}A/m^2 and 6 ± 2 Oe per 10^{10} A/m^2 respectively for $t_{\text{Tb}} = 8 \text{ nm}$, with $\zeta = 0.327$ which is consistent with the values determined through the harmonic measurements based on (5.9). To evaluate the SOT fields on an azimuthal angle for the variation of Tb thicknesses, the SOT fields are calculated based on these analytical expressions given by¹⁶⁷:

$$\Delta H_{DL} = \frac{b\beta_\phi - a\beta_{90-\phi}}{b^2 - a^2} \quad (5.12)$$

$$\Delta H_{FL} = \frac{a\beta_\phi - b\beta_{90-\phi}}{a^2 - b^2} \quad (5.13)$$

where a and b are coefficients related to (5.10) and are dependent on the orientation of the applied magnetic field, ϕ and the PHE correction factor, ζ . It was noted that there exists a mirror symmetry at $\phi = 45^\circ$ such that $a(\zeta, \phi) = b(\zeta, 90 - \phi)$ and $b(\zeta, \phi) = a(\zeta, 90 - \phi)$ ¹⁶⁶.

Figure 5.9 Computed SOT effective fields for the Tb samples ranging from $5 \text{ nm} \leq t_{Tb} \leq 9 \text{ nm}$ as a function of ϕ ; (a) damping-like effective field according to (5.12). (b) Consolidated field-like effective field according to (5.13). The dashed lines represent regions where no effective fields were computed at the symmetrical axes. (c) Damping-like effective field and (d) field-like effective field for $t_{Tb} = 5 \text{ nm}$.

The symmetrical axes are indicated in red dashed lines, as shown in Figure 5.9. At these axes, there was no effective field computed, as $a^2 = b^2$, cancels out the function at multiples of $\phi = 45^\circ$. As shown in Figure 5.9, there are clear indications of the azimuthal angular dependence of both the DL-term and the FL-term, in which their mean values were determined to be 10 Oe per 10^{10}A/m^2 and 2 Oe per 10^{10}A/m^2 , which is similar to the values obtained at 0° and 90° respectively. Two minimums were observed at azimuthal angles of 45° and 225° , which indicates the angular dependence of the DL-term in Pt/[Co/Ni]₂/Co/Tb/Ta.

Similarly, the FL-term have minimums at the same angle, also indicating the angular dependence of the FL-term. The angular dependence of the DL-term is non-trivial and can be attributed to the combined effects of the Fermi surface distortion and Fermi sea contribution at the intermediate Rashba regime at the Co/Tb interface^{167–169}. The angular dependence of the FL-term is attributed to the Fermi surface distortion, which becomes significant when the Rashba spin-orbit coupling energy is comparable to the exchange energy¹⁶⁸. In the Pt/[Co/Ni]₂/Co/Ta reference sample, there was no indication of angular dependence as expected due to the weak Rashba spin-orbit coupling caused by oxidation of the top Ta layer^{167,168}. Since angular dependence is absent in the reference sample and present in the samples with Co/Tb interfaces. It can be mentioned that the presence of strong Rashba spin coupling at the Co/Tb interface contributes to both DL and FL effective field.

Comparing the t_{Tb} , the angular dependence of both the DL-term and FL-term decreases with the increase in Tb thickness. At $t_{\text{Tb}} = 5\text{ nm}$, there is an indication of the angular dependence of both the DL and FL terms, but this dependence on magnetisation direction diminishes with increasing thickness. This occurs due to the diminishing ratio of Rashba spin-orbit coupling to the exchange coupling of the magnetic material¹⁶⁷. The diminishing ratio of the Rashba spin-orbit coupling to exchange coupling could originate from one of these or a combination of these mechanisms, which disrupts the quality of the Co/Tb interface. Firstly, the shift towards poor crystallinity¹⁶⁸ in thicker Tb samples, secondly, the increase in oxidation¹⁶⁷ of thicker Tb at the sidewalls of the Hall cross and

thirdly, greater intermixing between Co and Tb at the Co/Tb interface, can lead to the diminishing of the Rashba coupling. With an indirect method of determining the crystallinity, atomic force microscopy (AFM) scanning was performed on Hall-cross devices for various Tb thickness, t_{Tb} from 5 nm to 9 nm to determine the effect of Tb thickness on roughness. The surface topology was characterised by the average roughness parameter, $\mathfrak{R}_{av} = (1/L) \int_0^L |Z(x)| dx \approx (|Z_1| + |Z_2| + \dots + |Z_N|)/N$, which describes the average surface roughness of the surface, where L is the evaluation length and $Z(x)$ is the height of the profile and N is the number of regions taken into account. Through the comparison of the AFM images, for samples between $t_{Tb} = 5\text{nm}$ and $t_{Tb} = 9\text{ nm}$, the surface roughness is shown as a function of Tb thickness in Figure 5.10 (a) and (b) respectively.

Figure 5.10 (a) AFM image for $t_{Tb} = 5\text{ nm}$ and $t_{Tb} = 9\text{nm}$. (b) Average surface roughness, $\mathfrak{R}_{av}$, as a function of Tb thickness.

This dependence of the Rashba coupling on the Tb thickness was previously determined from the FL-efficiency in the H_z -biased harmonic measurement. Even as ξ_{FL}^{eff} drops with increasing t_{Tb} , the ξ_{FL}^{eff} for Co/Tb stacks are still more significant in magnitude than ξ_{FL}^{eff} in the reference Co/Ta, which affirms that Tb is responsible for more significant Rashba coupling leading to more significant ξ_{FL}^{eff} .

The spin diffusion length of Tb, λ_{Tb} can be extracted by utilising the spin Hall angle or damping-like efficiency of Tb, $\theta_{SH,Tb}^{eff}$. Taking the normalised value of $\theta_{SH,Tb}$ to the case where $t_{Tb} = 9$ nm and calculating $\theta_{SH,Tb}$ by assuming that $\theta_{SH,Pt} \approx 0.09$, the λ_{Tb} can be extracted from the equation given by¹⁷⁰,

$$\frac{\theta_{SH,Tb}}{\theta_{SH,Tb(9\text{ nm})}} = 1 - \text{sech}\left(\frac{t_{Tb}}{\lambda_{Tb}}\right), \quad (5.14)$$

where θ_{SH} is the spin Hall angle, t_{Tb} is the thickness of the Tb layer, λ_{Tb} is the spin-diffusion length of Tb.

Figure 5.11 Experimental fitting of the normalised spin Hall angle, $\theta_{Tb,normalised}$ as a function of the Tb thickness, t_{Tb} .

The fitting result extracted from the $\theta_{Tb,normalised}$ shown in Figure 5.11 reveals that the spin diffusion length of Tb is approximately 10 nm, which is close to earlier reports¹⁷¹. Thus, the results show that the thickness of Tb studied in this work is within λ_{Tb} , thus confirming the bulk SHE as the dominant mechanism in the DL-SOT.

5.3 Summary

This chapter conclusively reports the enhancement of the DL-torque and FL-torque on a perpendicularly magnetised Pt/[Co/Ni]₂/Co/Tb sample ($|\xi_{DL}^{eff}| \approx 0.25 \pm 0.01$ for $t_{Tb} = 5$ nm) as compared to a reference Pt/[Co/Ni]₂/Co/Ta sample ($|\xi_{DL}^{eff}| \approx 0.18 \pm 0.01$ for $t_{Tb} = 5$ nm) which are attributed to the sizeable spin-orbit coupling in rare earth, Tb. Due to a SHE dominant mechanism, the $|\xi_{DL}^{eff}|$ increases to 0.55 ± 0.01 but $|\xi_{FL}^{eff}|$ diminishes with increasing t_{Tb} , characterised using a H_z -bias harmonic Hall measurement. The current-induced magnetisation switching shows that the enhanced DL-term, enabled successful deterministic switching with the greatest efficiency when $t_{Tb} > 7$ nm. Due to a stronger Rashba coupling in Co/Tb as compared to the reference Co/Ta, angular dependence of the DL and FL terms was observed for the Tb samples, while the reference stack had no angular dependence. In the Tb thickness variation, the Rashba coupling diminishes with the increasing thickness due to oxidation in the Tb sidewalls, poor crystallinity of Tb and the increase in Co/Tb intermixing at the Co/Tb interface which disrupts the Co/Tb interface.

In summary, the enhancement of the DL and FL terms by rare earth, Tb, can open up new potential avenues of research on rare earth materials as spin Hall material alternatives for SOT-based devices. As such, in an overview of alternative RE materials with similar electronic configurations, similar enhancements can be expected. However, it depends on the proximity of the 4f electrons to the Fermi level. In order to determine a suitable RE material which can further enhance the SHE, RE materials and alloys that are either closer to the peak in the 4f density of states or have more delocalised electrons in the 4f shell, should be considered. To accomplish this, engineering and optimisation of the spin Hall conductivity by insertion of high conductivity doping atoms, i.e. Cu or Ag to enhance electrical conductivity could be explored. Thus, in doing so, the charge to spin current conversion efficiency in RE materials can be exploited for future technological and SOT-memory applications.

5.4 Supplemental Section: Analytical Derivations

Section 1 to 3 of the Supplemental section describes the analytical derivations for the magnetisation polar and azimuthal angular modulation, Hall voltage modulation via an external current-induced magnetic field and the azimuthal angular dependence of the magnetisation in a H_z -biased harmonic Hall measurement.

1. H_z Biased Harmonics: Polar and Azimuthal Angle Modulation

In the Pt/[Co/Ni]₂/Co/Tb or Ta system with perpendicular magnetic anisotropy (PMA), the magnetic energy of the system can be expressed in spherical polar coordinates as:

$$E = -K_{u,eff} \cos^2 \theta - K_l \sin^2 \phi \sin^2 \theta - \mathbf{M} \cdot \mathbf{H}, \quad (\text{S5.1})$$

where $K_{u,eff}$ is the effective out-of-plane anisotropy constant and K_l is the effective in-plane anisotropy constant. θ and ϕ are the polar and azimuthal angles of the magnetisation vector. The vectors $\mathbf{M}$ and $\mathbf{H}$ can be expressed as follows:

$$\mathbf{M} = M_s \begin{pmatrix} \sin \theta \cos \phi \\ \sin \theta \sin \phi \\ \cos \theta \end{pmatrix}, \quad \mathbf{H} = \begin{pmatrix} H_x \\ H_y \\ H_z \end{pmatrix} \equiv \begin{pmatrix} \sin \theta_H \cos \phi_H \\ \sin \theta_H \sin \phi_H \\ \cos \theta_H \end{pmatrix}, \quad (\text{S5.2})$$

M_s is the saturation magnetisation, H_x, H_y, H_z are the components of the external applied magnetic field in the directions of the x, y, z axes. θ_H and ϕ_H are the polar and azimuthal angles of the applied external field in spherical polar coordinates. When an external applied magnetic field, $\mathbf{H}$ is applied to the system, the magnetic energy evolves, according to (S5.1). In the typical nanosecond relaxation regime, the magnetisation relaxes to a new equilibrium position with a new energy minimum.

As such, the new energy minima position can be derived by the partial derivatives of the energy with respect to the polar, $\partial E / \partial \theta$ and azimuthal, $\partial E / \partial \phi$ angles respectively. By inserting (S5.2) to (S5.1) and assuming that $K_{eff} \gg K_l$ for a system with strong PMA, K_l is neglected,

$$E = -K_{u,eff} \cos^2 \theta - [M_s H_x \sin \theta \cos \phi + M_s H_y \sin \theta \sin \phi + M_s H_z \cos \theta], \quad (S5.3)$$

$$\frac{\partial E}{\partial \theta} = \frac{2K_{u,eff}}{M_s} \sin \theta \cos \theta - [H_x \cos \theta \cos \phi + H_y \cos \theta \sin \phi - H_z \sin \theta], \quad (S5.4)$$

$$\frac{\partial E}{\partial \phi} = -[-M_s H_x \sin \theta \sin \phi + M_s H_y \sin \theta \cos \phi], \quad (S5.5)$$

Substituting $2K_{u,eff}/M_s = H_{k,eff}$, $\partial E/\partial \theta = \partial E/\partial \phi = 0$, θ is small even when a constant H_z field is applied, such that $\cos \theta \approx 1$, $\partial E/\partial \theta = 0$ can be expressed as:

$$\sin \theta = \frac{H_x \cos \phi + H_y \sin \phi}{H_{k,eff} + H_z}, \quad (S5.6)$$

$$\theta = \arcsin \left(\frac{H_x \cos \phi + H_y \sin \phi}{H_{k,eff} + H_z} \right), \quad (S5.7)$$

$$\Delta \theta = \frac{\partial \theta}{\partial H} \Delta H = \left(\frac{\partial \theta}{\partial H_x} \Delta H_x + \frac{\partial \theta}{\partial H_y} \Delta H_y + \frac{\partial \theta}{\partial H_z} \Delta H_z \right), \quad (S5.8)$$

$$\Delta \theta = \frac{(H_{k,eff} + H_z)(\Delta H_x \cos \phi + \Delta H_y \sin \phi) + \Delta H_z (H_x \cos \phi + H_y \sin \phi)}{(H_{k,eff} + H_z)^2}, \quad (S5.9)$$

Since $\Delta H_z \ll H_{k,eff} + H_z$,

$$\Delta \theta \approx \frac{(\Delta H_x \cos \phi + \Delta H_y \sin \phi)}{(H_{k,eff} + H_z)}, \quad (S5.10)$$

Similarly, the azimuthal angle modulation was determined by its energy derivative, $\partial E/\partial \theta = 0$,

$$\tan \phi = \frac{H_y}{H_x}, \quad (S5.11)$$

$$\phi = \arctan \left(\frac{H_y}{H_x} \right), \quad (S5.12)$$

$$\Delta \phi = \frac{\partial \phi}{\partial H} \Delta H = \frac{\Delta H_y H_x - \Delta H_x H_y}{H_x^2 + H_y^2}, \quad (S5.13)$$

where $\partial \theta/\partial H$ and $\partial \phi/\partial H$ represent the change in angles θ and ϕ caused by the current induced effective field ΔH

2. H_z Biased Harmonics: Hall Voltage Measurement Derivation

For a magnetic Hall cross, the Hall voltage, V_H , the Hall effect contributions are mainly from the anomalous Hall effect, V_{AHE} (AHE) and planar Hall effect, V_{PHE} (PHE) such that:

$$V_H = V_{AHE} + V_{PHE} = \frac{1}{2} I \Delta R_{AHE} \cos \theta + \frac{1}{2} I \Delta R_{PHE} \sin^2 \theta \sin 2\phi, \quad (S5.14)$$

ΔR_{AHE} represents the change in AHE resistance and ΔR_{PHE} represents the change in resistance from the PHE. I represents the current in the wire. An alternating current (AC) source will cause a modulation of the current, polar, and azimuthal angles given by:

$$\begin{aligned} I &\xrightarrow{\text{a.c modulation}} I_{ac} \sin \omega t \\ \theta + \Delta\theta &\xrightarrow{\text{a.c modulation}} \theta + \Delta\theta \sin \omega t, \\ \phi + \Delta\phi &\xrightarrow{\text{a.c modulation}} \phi + \Delta\phi \sin \omega t \end{aligned} \quad (S5.15)$$

The applied frequency $\omega = 2\pi f$ is small such that it is significantly lower than the Larmor frequency (in the GHz regime). These modulations are accounted for by substituting into (S5.14) and separated into the AHE and PHE:

$$V_{AHE} = \frac{1}{2} I_{ac} \sin \omega t \Delta R_{AHE} \cos(\theta + \Delta\theta \sin \omega t), \quad (S5.16)$$

$$V_{PHE} = \frac{1}{2} I_{ac} \sin \omega t \Delta R_{PHE} \sin^2(\theta + \Delta\theta \sin \omega t) \sin(2(\phi + \Delta\phi \sin \omega t)), \quad (S5.17)$$

To simplify the expression (S5.16) and (S5.17), first-order Taylor expansion was done to attain:

$$V_{AHE} \approx \frac{1}{2} I_{ac} \sin \omega t \Delta R_{AHE} (\cos \theta - \Delta\theta \sin \theta \sin \omega t), \quad (S5.18)$$

$$V_{PHE} \approx \frac{1}{2} I_{ac} \sin \omega t \Delta R_{PHE} (\sin^2 \theta + 2\Delta\theta \sin \theta \cos \theta \sin \omega t) (\sin 2\phi + 2\Delta\phi \cos 2\phi \sin \omega t), \quad (S5.19)$$

Utilising the trigonometric identity, $\sin^2 x = 1/2(1 - \cos 2x)$, and re-expressing the Hall voltage in an alternate form, $V_H = V_0 + V_\omega \sin \omega t + V_{2\omega} \cos 2\omega t$ such that the $V_0, V_\omega, V_{2\omega}$ coefficients are:

$$V_0 = \frac{1}{2} I_{ac} \left(\frac{1}{2} \left[-\Delta R_{AHE} \sin \theta + \Delta R_{PHE} \sin 2\theta \sin 2\phi \right] \Delta \theta + \Delta R_{PHE} \sin^2 \theta \cos 2\phi \Delta \phi \right), \quad (S5.20)$$

$$V_\omega = \frac{1}{2} I_{ac} \left[\Delta R_{AHE} \cos \theta + \Delta R_{PHE} \sin^2 \theta \sin 2\phi \right], \quad (S5.21)$$

$$V_{2\omega} = \frac{1}{2} I_{ac} \left(\frac{1}{2} \left[-\Delta R_{AHE} \sin \theta + \Delta R_{PHE} \sin 2\theta \sin 2\phi \right] \Delta \theta + \Delta R_{PHE} \sin^2 \theta \cos 2\phi \Delta \phi \right), \quad (S5.22)$$

The current induced effective fields $\Delta \mathbf{H}$ are mainly noticeable in the second harmonics Hall voltage through the angular modulations in the polar and azimuthal angles.

3. H_z Biased Harmonics: Azimuthal Angular Dependence

The magnetisation modulation angles, (S5.10) and (S5.13) can be substituted into the (S5.22) to attain the angular dependence of the magnetic system. Utilising the identity, $\cos \theta = \sqrt{1 - \sin^2 \theta} = 1 - 1/2 \sin^2 \theta$, the harmonic Hall voltages, V_ω and $V_{2\omega}$ expression can be written as:

$$V_\omega = \frac{1}{2} I_{ac} \left[\Delta R_{AHE} + \left(\Delta R_{PHE} \sin 2\phi - \frac{\Delta R_{AHE}}{2} \right) \left(\frac{H}{H_{k,eff} + H_z} \right)^2 \right], \quad (S5.23)$$

$$V_{2\omega} = I_{ac} \frac{H}{(H_{k,eff} + H_z)^2} \left\{ \left[-\frac{1}{4} \Delta R_{AHE} + \Delta R_{PHE} \sin 2\phi \right] (\Delta H_x \cos \phi + \Delta H_y \sin \phi) \right. \\ \left. - \frac{1}{2} \Delta R_{PHE} \cos 2\phi (\Delta H_y \cos \phi_H - \Delta H_x \sin \phi_H) \right\}, \quad (S5.24)$$

The dependence on H can be removed by taking the derivatives, $A_\omega = \partial^2 V_\omega / \partial H^2$ and $B_{2\omega} = \partial V_{2\omega} / \partial H$, and are expressed as,

$$A_\omega = -\frac{I_{ac}}{2(H_{k,eff} + H_z)^2} (\Delta R_{AHE} - 2\Delta R_{PHE} \sin 2\phi), \quad (S5.25)$$

$$B_{2\omega} = \frac{I_{ac}}{(H_{k,eff} + H_z)^2} \left\{ \left[-\frac{1}{4} \Delta R_{AHE} + \Delta R_{PHE} \sin 2\phi \right] (\Delta H_x \cos \phi + \Delta H_y \sin \phi) \right. \\ \left. - \frac{1}{2} \Delta R_{PHE} \cos 2\phi (\Delta H_y H_x - \Delta H_x H_y) \right\} \quad (S5.26)$$

The azimuthal angular dependence of the SOT effective fields can be derived by eliminating I_{ac} , $H_{k,eff}$, H_z such that, $\beta_\phi = B_{2\omega}/A_\omega$, $\Delta H_x \equiv \Delta H_{DL}$; $\Delta H_y \equiv \Delta H_{FL}$ is given by as

$$\beta_\phi = -\frac{1}{2}(\Delta H_{DL} \cos \phi_H + \Delta H_{FL} \sin \phi_H) + \frac{R_{PHE} \cos 2\phi_H (\Delta H_{FL} \cos \phi_H - \Delta H_{DL} \sin \phi_H)}{(R_{AHE} - 2R_{PHE} \sin 2\phi_H)}, \quad (S5.27)$$

$$\beta_\phi = \left(-\frac{1}{2} \sin \phi + \frac{\zeta \cos \phi \cos 2\phi}{1 - 2\zeta \sin 2\phi} \right) \Delta H_{FL} + \left(-\frac{1}{2} \cos \phi - \frac{\zeta \sin \phi \cos 2\phi}{1 - 2\zeta \sin 2\phi} \right) \Delta H_{DL}, \quad (S5.28)$$

$$\beta_\phi = a\Delta H_{FL} + b\Delta H_{DL}, \quad (S5.29)$$

The fitting function for the azimuthal angle, ϕ derived in (S31) for the H_z biased harmonic measurement becomes similar to C. Engel *et al.*, as the dependence on the out-of-plane H_z field is eliminated through $\beta_\phi = B_{2\omega}/A_\omega$. The derivations also show that the harmonic measurement is invariant with the applied H_z field, as it was mainly applied to ensure that θ is small and domain nucleation could be prevented at sweeping fields

Adopting the derived equation from C. Engel *et al.*, the azimuthal angle dependence of the field can be given as,

$$\Delta H_{DL} = \frac{b\beta_\phi - a\beta_{90-\phi}}{b^2 - a^2}, \quad (S5.30)$$

$$\Delta H_{FL} = \frac{a\beta_\phi - b\beta_{90-\phi}}{a^2 - b^2}, \quad (S5.31)$$

The error of propagation for the functions a and b to estimate the angle offset is derived as:

$$\Delta(a) = \frac{da}{d\phi} \Delta\phi = \left(-\frac{1}{2} \cos \phi - \frac{2\zeta \cos \phi \sin 2\phi}{1 - 2\zeta \sin 2\phi} + \frac{4\zeta^2 \cos \phi \cos^2 2\phi}{(1 - 2\zeta \sin 2\phi)^2} - \frac{\zeta \sin \phi \cos 2\phi}{1 - 2\zeta \sin 2\phi} \right), \quad (S5.32)$$

$$\Delta(b) = \frac{db}{d\phi} \Delta\phi = \left(\frac{1}{2} \cos \phi + \frac{2\zeta \sin \phi \sin 2\phi}{1 - 2\zeta \sin 2\phi} - \frac{4\zeta^2 \sin \phi \cos^2 2\phi}{(1 - 2\zeta \sin 2\phi)^2} - \frac{\zeta \cos \phi \cos 2\phi}{1 - 2\zeta \sin 2\phi} \right), \quad (S5.33)$$

Due to an overestimate of the coefficients a and b because of their sole dependence on ζ and ϕ , the uncertainties for the effective fields can be computed as, such that $\Delta(\phi) = 0.5^\circ$, due to the limits of the rotation stage:

$$\Delta(\Delta H_{DL}) = \frac{\partial \Delta H_{DL}}{\partial a} \Delta(a) + \frac{\partial \Delta H_{DL}}{\partial b} \Delta(b), \quad (S5.34)$$

$$\Delta(\Delta H_{FL}) = \frac{\partial \Delta H_{FL}}{\partial a} \Delta(a) + \frac{\partial \Delta H_{FL}}{\partial b} \Delta(b), \quad (S5.35)$$

where the derivatives can be expressed as:

$$\frac{\partial \Delta H_{DL}}{\partial a} = \frac{\beta_\phi (a^2 - b^2) - 2a(a\beta_\phi - b\beta_{90-\phi})}{(a^2 - b^2)^2}$$

$$\frac{\partial \Delta H_{DL}}{\partial b} = \frac{-\beta_{90-\phi} (a^2 - b^2) + 2b(a\beta_\phi - b\beta_{90-\phi})}{(a^2 - b^2)^2}$$

$$\frac{\partial \Delta H_{FL}}{\partial b} = \frac{2a(b\beta_\phi - a\beta_{90-\phi})}{(b^2 - a^2)^2}$$

$$\frac{\partial \Delta H_{FL}}{\partial a} = \frac{-2b(b\beta_\phi - a\beta_{90-\phi})}{(b^2 - a^2)^2}$$

Chapter 6: Direct current-biased harmonic probing of spin accumulation magnetoresistance in rare earth systems

Abstract

In this chapter, the observation of spin accumulation magnetoresistance in perpendicularly magnetised Pt/[Co/Ni]₂/Co/Tb samples in a dc biased harmonic Hall measurement is reported. An anomalous behaviour was observed in the second harmonic Hall resistance when a biasing current was applied to the structures. Conventionally, spin-orbit torque effects are exclusively found through the second harmonic Hall resistances. The experimental results and analytical derivations reveal that the anomalous second harmonic Hall resistance originates from spin accumulation induced by the spin Hall effect. The spin accumulation was adiabatically quantified through the anomalous second harmonic Hall resistance in the structures. The spin accumulation depends on the thickness of the Tb or Ta layers, such that it ranges from 3.0% in $t_{Tb} = 5$ nm to 6.6% in $t_{Tb} = 8$ nm, and 1.3 % in the reference stack of $t_{Ta} = 5$ nm of the local magnetisation when the total applied current density is 10^{11} A/m².

6.1 Introduction

In-plane current injection into non-magnetic (NM)/ferromagnetic (FM) structures are known to generate torques due to the presence of spin-orbit coupling (SOC), also known as the spin-orbit torque (SOT). SOT serves as an efficient alternative for reversal of the magnetisation in the FM layer^{36,37,39,117,119,130} and driving of DWs^{8,74,121,122,172,173}. The origin of the SOT is attributed to both the spin Hall effect^{36,47,61,148,174} (SHE) in the NM layer and the interfacial Rashba effect^{48,49,58}. Recently, the *f*-electron rare-earth (R) metals have drawn increasing attention because of their substantial SHE contribution due to their partially filled *f*-orbitals^{146,149}. The R-ferrimagnets^{43,70,150–152,154–157,175} have been examined to show promising spintronic application with enhanced SOTs and faster DWM¹⁵⁰. In Chapter 5, enhancement of SOT in a Pt/FM/Tb heterostructure was reported, where the R metal, Tb served as a heavy metal (HM) layer replacement, with significant spin Hall angle, $\theta_{SH,Tb} = -0.48$ ⁶². Although the relative efficiency of the dampinglike (DL) torque, ξ_{DL} is commonly used as a figure of merit for the spin Hall material, the direct quantification of spin accumulation caused by the portion diffusing into the FM to cause the H_{FL} and H_{DL} remain elusive. Several experiments utilising magneto-optical Kerr imaging have been used to characterise spin-separation of electrons in semiconductors^{176,177}, and in heavy metals^{178–181}. Stamm *et al.*¹⁸¹ reported an indirect detection of spin accumulation in Pt and W utilising a combination of current modulation and linear magneto-optical Kerr effect (MOKE) measurement while comparing with *ab-initio* calculations. Other optical methods of detection include Brillouin light scattering¹⁸², and second harmonic generation¹⁸³. The utilisation of optical methods depends on the reflectivity of the material which may change due to Joule heating, and on the sensitivity of the MOKE detection. An all-electrical direct detection of the spin accumulation magnetoresistance utilising a dc biased harmonic Hall measurement is proposed in this chapter.

In this chapter, the observation of an anomalous resistance behaviour in the second harmonic Hall resistance measurement with biasing direct current in Pt/[CoNi]₂/Co/Tb structures with perpendicular magnetic anisotropy (PMA) is reported. Conventionally, the second harmonic Hall resistance, $R_{2\omega}$, reveals only the SOT dampinglike (DL) and fieldlike (FL) terms. Here, we show that the anomalous behaviour in $R_{2\omega}$ originates from the spin accumulation due to the spin Hall effect. Subsequently, the $R_{2\omega}$ is used to quantify the spin accumulation adiabatically after considering the thermoelectric contributions caused by Joule heating.

6.2 Results and Discussions

The measurements were carried out in Ta (2)/Pt (6)/[Co (0.3)/Ni (0.6)]₂/Co (0.3)/Tb (5 – 8) structures and reference stack Ta (2)/Pt (6)/[Co (0.3)/Ni (0.6)]₂/Co (0.3)/Ta (5) with perpendicular magnetic anisotropy. All nominal thicknesses are in nm where the films were deposited with process steps similar to our previous report⁶². Both direct current (DC) and alternating current (AC) were applied in the nanowire throughout the harmonic Hall measurement with a constant H_z bias field of ± 1100 Oe to obtain the first (V_{ω}^{ac+dc}) and second ($V_{2\omega}^{ac+dc}$) harmonic Hall voltages simultaneously with a lock-in amplifier. Figure 6.1 (a) shows a schematic illustration of the measurement setup with the input of a biasing current, such that $I_{input} = \pm I_{dc} + I_{ac} \sin \omega t$, and a Scanning Electron Micrograph (SEM) image of the device with arrows indicating the field directions for both the longitudinal (H_x) and transverse (H_y) measurement schemes. Figure 6.1(b) illustrates the spin-selective separation of electrons in the Pt layer due to SHE, such that the spin-polarized electrons diffuse into the FM layer to transfer spin torque on the magnetisation of the [Co/Ni]₂/Co layer. Due to the magnetoresistive effects, spins accumulated near the Pt-FM and FM-Tb interfaces are analogous to the STT idea that S. Zhang *et al.*²⁹ proposed, in which spin accumulation transfers spin torque to the FM. As this transfer occurs in the sub-nanosecond regime, the spins can be reasonably considered to be in the lowest energy state and oriented in the direction of the localised magnetisation of the [Co/Ni]₂/Co layer, M/m where M is the magnetisation and m is

the magnetisation unit vector. Similarly, when extended to a dc bias regime, an identical approximation can be considered as the diffused spins align along the $\mathbf{m}$ direction. Hence, the resultant direction of the diffused spin is aligned along the magnetisation direction of the FM layer after momentum transfer. Thus, the magnitude of the diffusing spin per unit volume, which is related to the magnitude of the SHE induced spin accumulation, can be written as $s\mathbf{m}$ in the harmonic Hall measurement, such that the total magnetisation becomes, $M\mathbf{m} + s\mathbf{m}$. Figure 6.1 (c) shows the anomalous Hall (AHE) loops of the samples with $5 \leq t_{Tb} \leq 8$ nm and reference stack $t_{Ta} = 5$ nm where all devices demonstrate sharp switching when an external H_z field is swept with sufficiently small dc measurement current density of 1×10^{10} A/m², that will not perturb the system during the AHE measurement. These AHE results demonstrate that the films have perpendicular magnetic anisotropy (PMA).

Figure 6.1 (a) Schematic illustration of the measurement geometry and the SEM image of the Hall cross structure with the measurement parameters in determining the spin accumulation, (b) schematic of spins accumulating near the Pt-FM and Tb-FM interface where the spins scatter due to the spin Hall effect align itself, and in the millisecond regime, the spins exert a torque on the FM layer and behaves like the localised magnetisation in the FM. (c) Anomalous Hall (AHE) loops of the various samples with $t_{Ta} = 5 \text{ nm}$ and $5 \leq t_{Tb} \leq 8 \text{ nm}$ indicating the presence of PMA

6.2.1 Analytical Expressions for Spin Accumulation Quantification in PMA systems

The Hall voltage comprises of the anomalous Hall (AHE) voltage, V_{AHE} and the planar Hall voltage, V_{PHE} , i.e. $V_H = V_{AHE} + V_{PHE}$ which can be expressed in the form:

$$V_{AHE} = I.k_a M_0 \cos \theta, \quad (6.1)$$

$$V_{PHE} = I.k_p M_0^2 \sin^2 \theta \sin 2\phi, \quad (6.2)$$

where M_0 is the intrinsic magnetisation value excluding spin accumulation, θ is the polar angle of the magnetisation, ϕ is the azimuthal angle of the magnetisation and I is the applied current, k_a and k_p are proportionality constants associated with the AHE and PHE respectively.

The spin accumulation which induces SOT can be expressed as $-\gamma_0 \mathbf{M} \times \mathbf{J}_s$ acting on the ferromagnetic layer, where γ_0 is the gyromagnetic ratio, $\mathbf{M}$ is the magnetisation of the ferromagnetic layer, and J is a diffusion coefficient related to the spin diffusion length of the spin accumulation, s in the FM layer from Pt and Tb or Ta. When expressed in the form of effective fields, SOT composes of the fieldlike term, $\mathbf{H}_{FL} = H_{FL} \mathbf{p}$ and the dampinglike term, $\mathbf{H}_{DL} = H_{DL} (\mathbf{m} \times \mathbf{p})$ and which can be alternatively expressed as, $\mathbf{J}_s = H_{FL} \mathbf{p} + H_{DL} (\mathbf{m} \times \mathbf{p})$. Since spin accumulation occurs in nanoseconds, and the applied current density has a period significantly greater than that, the spin accumulation can be analogously viewed as a localised magnetic moment in the lowest energy state. As such, s changes the magnetisation of the FM layer from $M_0 \mathbf{m}$ to $M_0 \mathbf{m} + s \mathbf{m}$, which leads to the re-expression of the AHE and PHE voltages given by,

$$V_{AHE} = k_a (M_0 I + \zeta I^2) \cos \theta, \quad (6.3)$$

$$V_{PHE} = k_p (I M_0^2 + 2 I^2 \zeta + I^3 \zeta^2) \sin^2 \theta \sin 2\phi, \quad (6.4)$$

where in the steady-state condition, the localised magnetic moment, s is proportional to the applied current density, $s = \zeta I$, as the rate of spin decay is equivalent to the spin generation, where ζ is a proportionality constant related to the spin accumulation. $k_a = R_{AHE} / M_0$ and $k_p = R_{PHE} / M_0^2$ are proportionality constants, and M_0 is equivalent to the saturation magnetisation, M_s .

When a DC current is added together with an AC current, the spins are preferentially accumulated at one interface more than the other, which enables the effective determination of the spin accumulation, such that $s = \zeta (I_{ac} \sin \omega t + I_{dc})$, ω is the angular frequency of the applied AC current and can be substituted into (6.3) and (6.4). This spin accumulation contributes to the second harmonic Hall resistance and can be expressed as a generic second harmonic Hall voltage given by,

$$V_{2\omega, H}^{ac+dc} \Big|_{\parallel (\perp)} = -\frac{R_{AHE}}{2} \begin{pmatrix} \zeta I_{ac}^2 \cos \theta_0 \\ -\Delta \theta_{ac} (I_{ac} + 2\zeta I_{ac} I_{dc}) \sin \theta_0 \\ -\zeta \Delta \theta_{dc} I_{ac}^2 \sin \theta_0 \end{pmatrix} \mp R_{PHE} \sin^2 \theta_0 \left(I_{ac} \Delta \phi_{ac} + \frac{\zeta}{M_0} \left(4I_{dc} I_{ac} \Delta \phi_{ac} + 2I_{ac}^2 \Delta \phi_{dc} \right) \right), \quad (6.5)$$

where $\zeta = \zeta/M_0$ is the spin accumulation factor, $\Delta \theta_{ac}$ and $\Delta \theta_{dc}$ are the polar angle oscillations induced by the ac and dc current respectively, $\Delta \phi_{ac}$ and $\Delta \phi_{dc}$ are the azimuthal angle oscillations induced by the ac and dc current respectively, $H_{k,eff}$ is the effective anisotropic field of the system, $\parallel$ refers to the case where $\phi = 0^\circ$, $\perp$ refers to the case where $\phi = 90^\circ$, and H_z is the constant bias field applied in the z-direction. In the case where only an ac current is applied in the system, such that $I_{dc} = 0$, $\Delta \theta_{dc} = \Delta \phi_{dc} = 0$, and the contribution due to the spin accumulation is assumed to be small due to the alternating nature of the ac source ($\zeta \rightarrow 0$). The equation (6.5) can be expressed as the general second harmonic expression at the longitudinal and transverse scheme of measurement given as,

$$V_{2\omega, H}^{ac} \Big|_{\parallel (\perp)} \approx \frac{1}{2} I_{ac} R_{AHE} \Delta \theta_{ac} \sin \theta_0 \mp I_{ac} R_{PHE} \Delta \phi_{ac} \sin^2 \theta_0, \quad (6.6)$$

where $\Delta \theta_{ac} \Big|_{\parallel (\perp)} = \Delta H_{x(y), ac} / (H_{k,eff} + H_z)$, $\Delta \phi_{ac} \Big|_{\phi=0^\circ (90^\circ)} = \Delta H_{y(x), ac} / H_{x(y)}$.

The second harmonic Hall voltage maintains its linear correlation with the applied external field when only ac current is applied to the system. This experimental

comparison between the ac-case and the ac inclusive of dc bias case is shown later in Figure 6.2. The SOT effective fields induced by both the ac and dc currents can be expressed in terms of the DL efficiency, ξ_{DL} and FL efficiency, ξ_{FL} given by:

$$\Delta H_{x(y),ac/dc} = \frac{\hbar J_{ac/dc} \xi_{DL(FL)}}{2eM_s t}, \quad (6.7)$$

where e is the electron charge, t is the thickness of the ferromagnetic layer, M_s is the saturation magnetisation of the film stack.

After substituting (6.7) into (6.5), the generic expression for the second harmonic Hall resistance, $R_{2\omega,ac}^{dc}$ can be expressed in terms of the applied magnetic field as,

$$R_{2\omega,ac}^{dc} \Big|_{\parallel(\perp)} = \left(-\frac{R_{AHE}}{2} \zeta I_{ac} + \frac{R_{AHE}}{4} \zeta I_{ac} \left(\frac{H_{x(y)}}{H_{k,eff} + H_z} \right)^2 + Z^{x(y)} H_{x(y)} \right), \quad (6.8)$$

$$Z^{x(y)} = \frac{\hbar}{2eM_s t (H_{k,eff} + H_z)^2} \left[\frac{R_{AHE} \xi_{DL(FL)}}{2} (J_{ac} (1 + 2\zeta I_{dc}) + \zeta J_{dc} I_{ac}) \mp \gamma R_{AHE} \xi_{FL(DL)} \left(J_{ac} + \frac{\zeta}{M_s} (4I_{dc} J_{ac} + 2I_{ac} J_{dc}) \right) \right], \quad (6.9)$$

where $\xi_{DL(FL)}$ is the effective DL or FL efficiency coefficient, γ is the PHE correction factor, e is the electron charge, M_s is the saturation magnetisation, t is the thickness of the FM layer, the ac and dc represent the applied current densities in orders of 10^{10} A/m² respectively. As $Z^{x(y)}$ exists as a function of I_{dc} , $Z^{x(y)}$ will be utilised to calculate the spin accumulation coefficient, $\zeta = \zeta/M_s$ which is subsequently used to calculate the percentage spin accumulation given by, % Spin Accumulation = s/M_s , where $s = \zeta I_{ac+dc}$, at a total current density, $J_{ac+dc} = 1 \times 10^{11}$ A/m². Detailed derivation can be found in the Supplemental section at the end of this chapter.

6.2.2 Experimental Determination of Spin Accumulation in PMA systems

Experimentally, a mixture of both I_{ac} and I_{dc} are injected into the Hall cross-device, in the presence of a fixed H_z bias field of ± 1100 Oe to simultaneously attain the first and second harmonic Hall voltages, $V_{\omega,ac}^{dc}$ and $V_{2\omega,ac}^{dc}$ respectively in both schemes of measurement. The ac current density, ac was fixed at 4×10^{10} A/m², while varying the dc current density, dc from 1×10^{10} to 6×10^{10} A/m², such that the maximum total current density does not exceed 1×10^{11} A/m². This is to minimise the influence of Joule heating and to prevent current from irreversibly changing the magnetisation state of the device. Nonetheless, thermoelectric contributions were subsequently characterised in the latter portions of this chapter to correct the raw $R_{2\omega,ac}^{dc}$. Figure 6.3 (a) and (b) shows the first and second harmonic Hall voltage when dc biased current densities are passed through the Hall cross-device in the longitudinal and transverse scheme respectively without thermoelectric corrections. The second harmonic voltages reveal an anomalous parabolic behaviour which deviates from the conventional linear form. The parabolic behaviour varies with the increase in dc bias and is absent in a pure ac harmonic case as presented in the harmonic Hall measurement of chapter 5 and in Figure 6.2. The black data points shown in Figure 6.2 shows the linear correlation with the applied in-plane external field in both x and y directions of the longitudinal and transverse measurement schemes, respectively. Thus, the behaviour could be due to spin accumulation arising from the application of a dc bias in the system.

The measured data of the second harmonic in the longitudinal and transverse measurement scheme are shown in Figure 6.3 before thermoelectric correction are done and where the polar angle, $\theta < 10^\circ$ when the applied in-plane field sweep is between ± 2.5 kOe.

Figure 6.2 Second harmonic Hall resistances before ANE corrections for up and down magnetisation state in the (a) longitudinal, and (b) transverse measurement schemes.

Figure 6.3 First and second harmonic Hall voltages of the up and down magnetisation states when a mixed ac and dc current is passed into the Hall cross-device with $t_{Tb}=5\text{nm}$ in the (a) longitudinal measurement scheme and (b) transverse measurement scheme.

In metallic systems, the most significant thermoelectric contribution is due to the anomalous Nernst effect (ANE), which produces an electric field in the form, $E_{ANE} = -\alpha \nabla T \times \mathbf{m}$, where α is the ANE coefficient, ∇T is the temperature gradient and $\mathbf{m}$ is the magnetisation. Aside from the PHE and AHE contribution to $R_{2\omega}$, thermoelectric contribution due to the anomalous Nernst effect (ANE) were accounted for when both ac and dc currents are applied. When the current flows in the Hall cross, a temperature gradient exists due to non-homogeneous heating in correspondence to possible fabrication defects at the edges of the sample. Since heating is modulated by both the ac and dc current, the thermoelectric effects contribute to both the first and second harmonic Hall voltage given as, $R_{\omega}^{\nabla T} \propto \nabla T_{\omega,ac}^{\pm dc} = 2R_s J_{dc} J_{ac}$ and $R_{2\omega}^{\nabla T} \propto \nabla T_{2\omega,ac}^{\pm dc} = -R_s J_{ac}^2 / 2$ where R_s is the sample sheet resistance, J_{ac} is the ac current density, and J_{dc} is the dc current density. Since the ANE exists in the xy -, yz - and xz -planes, the total ANE contribution should be the sum of the ANE resistances originating from all three planes of geometry.

The magnetisation dependent ANE, R_{ANE} that has voltage perpendicular to the temperature gradient and the magnetisation, m_z , contributes to the $R_{2\omega}$ by mimicking the AHE but with much smaller amplitudes as shown in Figure 6.4(c). These measurements were performed by sweeping the applied magnetic field in the z -direction to determine the x - y plane contribution. In this measurement geometry, the SOT and Oersted fields do not contribute to the $R_{2\omega}$ since the variation of the Hall resistance is symmetric with respect to $\theta = 0^\circ$. The ac was fixed at $4 \times 10^{10} \text{ A/m}^2$, while varying the dc from 1×10^{10} to $6 \times 10^{10} \text{ A/m}^2$, the ratio of $R_{ANE} / 2R_{AHE}$ was experimentally determined for various dc current densities. Likewise, the field can be swept in the H_x and H_y direction for determination of the ANE resistance in the y - z and x - z planes, as shown in Figure 6.4 (a) and (b), respectively. When the magnetisation is close to saturation in the x or y direction, the difference in the V_{ω} at saturation will correspond

to the $2R_{PHE}$ while the $V_{2\omega}$ at saturation will correspond to thermoelectric contribution to $R_{2\omega}$ as shown in Figure 6.4.

Figure 6.4 First and second harmonic Hall in the (a) H_x field sweep (y - z plane), (b) H_y field sweep (x - z plane), and (c) H_z field sweep (x - y plane) regimes in the determination of the ANE correction factors in each plane of the ANE field.

The thermoelectric contribution can be subtracted from the raw data to attain the thermoelectrically correct, $(R_{2\omega,ac}^{dc})_{corr}$ can be expressed as^{54,184},

$$(R_{2\omega,ac}^{dc})_{corr} = (R_{2\omega,ac}^{dc})_{raw} - \psi R_{ac}^{dc}, \quad (6.10)$$

$$\psi = \left[R_{ANE}|_{xy-plane} + \frac{1}{\gamma} (R_{ANE}|_{xz-plane} + R_{ANE}|_{yz-plane}) \right] / 2R_{AHE}$$

where γ is the PHE correction factor.

Figure 6.5 Second harmonic Hall resistances after thermoelectric corrections in the longitudinal and transverse scheme of measurements, for (a), (b) up magnetised and (c), (d) down magnetised structures in $t_{Tb} = 5\text{nm}$ sample.

Table 6.1 shows the total ANE correction factors for all three planes, i.e. xy -, yz -, and xz -planes. Thus, upon correction, the second harmonic signal should only have contributions which relate purely to the spin accumulation and SOT fields. The corrected second harmonic Hall resistances are calculated and are shown in Figure 6.5.

Thicknesses (nm)	<u>yz- and xy-plane</u>		<u>xz- and xy-plane</u>	
	ψ_4^{-6}	ψ_4^{+6}	ψ_4^{-6}	ψ_4^{+6}
Tb 5	0.017	0.004	0.005	0.005
6	0.048	0.002	0.010	0.010
7	0.074	0.013	0.017	0.015
8	0.098	0.010	0.022	0.023
Ta 5	0.010	0.006	0.006	0.005

Table 6.1. ANE correction factors, ψ_{ac}^{dc} for various capping layer thicknesses of Tb and 5 nm Ta, when $dc = \pm 6 \times 10^{10} \text{ J/m}^2$ and $ac = 4 \times 10^{10} \text{ J/m}^2$ for the yz - and xy -plane, and the xz - and yz -plane respectively.

The ANE correction factors, ψ for all three planes, were considered and subsequently utilised to correct the raw $R_{2\omega,ac}^{dc}$ through (6.10) and as shown in Figure 6.5. Thus, upon correction, the second harmonic signal should only have contributions which relate purely to the spin accumulation and SOT fields. The corrected second harmonic Hall resistances are calculated and are shown in Figure 6.5. The thermoelectrically corrected $R_{2\omega,ac}^{dc}|_{\parallel(\perp)}$ is fitted according to (6.8) and a partial derivative of $R_{2\omega,ac}^{dc}|_{\parallel(\perp)}$ with respect to I_{dc} will yield,

$$\frac{\partial R_{2\omega,ac}^{dc}|_{\parallel(\perp)}}{\partial I_{dc}} = \frac{\partial Z|_{\parallel(\perp)}}{\partial I_{dc}} = \frac{3\zeta J_{ac} \hbar R_{AHE}}{2eM_s t (H_{k,eff} + H_z)^2} \left[\frac{\xi_{DL(FL)}}{2} \mp \frac{2\gamma \xi_{FL(DL)}}{M_s} \right], \quad (6.11)$$

Utilising the coefficient Z determined experimentally from the various I_{dc} , a function of Z against J_{dc} is plotted as shown in Figure 6.6 for both the longitudinal and transverse measurement schemes.

Figure 6.6 Plot of Z against J_{dc} in the (a) longitudinal scheme, and (b) transverse scheme of measurement for reference stack $t_{Ta} = 5$ nm and $5 \leq t_{Tb} \leq 8$ nm

The experimental relation $Z(J_{dc})$ can be utilised to calculate the spin accumulation factor, ζ , based on (6.11) with the available constant values shown in the analytical expression. This enables the quantification of s as a function of the thickness of the capping layer, t_{Tb} or t_{Ta} for both the longitudinal and transverse measurement

scheme, as shown in Figure 6.6. As $s = \zeta I_{ac+dc}$, the ratio of s/M_s is calculated to investigate the influence of the spin accumulation on the local magnetisation. As shown in Figure 6.7 (a), when the applied current density is 1×10^{11} A/m², the spin accumulation ratio is $\sim 3.0\%$ for $t_{Tb} = 5$ nm, $\sim 6.6\%$ for $t_{Tb} = 8$ nm, $\sim 1.3\%$ for the reference stack of $t_{Ta} = 5$ nm, when measured in the longitudinal scheme of measurement. As for the transverse scheme of measurement, the spin accumulation ratio is $\sim 3\%$ for $t_{Tb} = 5$ nm, $\sim 6.7\%$ for $t_{Tb} = 8$ nm and $\sim 1.5\%$ for the reference stack of $t_{Ta} = 5$ nm.

Figure 6.7 The magnitude of the spin accumulation and the reported (a) damping like efficiency in the longitudinal measurement scheme with table inset showing the percentage spin accumulation at different capping thicknesses per 10^{11} A/m², and (b) fieldlike efficiency in the transverse measurement scheme, with table inset showing the percentage spin accumulation at different capping thicknesses per 10^{11} A/m².

Since the spin accumulation is a phenomenon commonly associated with the spin Hall effect, s is expected to follow a similar trend to the spin Hall angle of the various samples with varying thickness of Ta or Tb irrespective of their magnitudes as observed in Figure 6.7 (a). s represents the spin accumulation generated, while the $\xi_{DL,eff}$ describes the effective DL efficiency corresponding to the effective spin current conversion through the Pt-Tb or Pt-Ta layer, similar to the effective spin Hall angle, θ_{SH}^{eff} . As such, s and $\xi_{DL,eff}$ are expected to exhibit similar trends with the thickness of the capping layer. The value of s seems to decrease at $t_{Tb} = 8$ nm due to a comparatively lower M_s . Thus, the percentage of spin accumulation was used to compare with $\xi_{DL,eff}$, which shows a similar trend to the θ_{SH}^{eff} . Furthermore, in Figure 6.7 (b), although the FL-efficiency of the SOT decreases with increasing t_{Tb} , the amount of spin accumulation, s generally increases. This indicates that the spin accumulation is not likely to be caused by the interfacial accumulation but instead occurring in bulk due to the SHE. Thus, the observation of the anomalous second harmonic Hall signals is a result of the spin accumulation at the FM layer which originates from the Pt, Ta or Tb layers. These results are in agreement with the effective spin Hall angle quantified in our previous report⁶² and in chapter 5 of this thesis, where the increase in t_{Tb} or t_{Ta} leads to an increase in the spin Hall efficiency. Therefore, as SHE is dominant in our stack structures, it is expected to have significant spin accumulation acting on the local magnetisation, as quantified in this section.

6.3 Summary

In conclusion, spin accumulation induced by electric current in stacks of Pt/[Co/Ni]₂/Co/Tb (5 – 8 nm) or Ta (5 nm) has been quantified using a dc biased harmonic Hall technique. The quantification results show that the spin accumulation is approximately 6.6% of the local magnetisation when the total applied current density is 10^{11} A/m². Our results demonstrate that, apart from the convention SOT fields, spin accumulation can contribute to the second harmonic Hall resistance. The ratio of spin accumulation over the applied electric current is consistent with the DL efficiencies. Hence, the ratio can be used to evaluate the spin current conversion efficiency of heavy metal or R metal in converting electrical current to spin current. This method provides an all-electrical alternative to determine spin accumulation through a commonly used harmonic Hall resistance approach.

6.4 Supplemental Section: Analytical Derivations

Anomalous Hall Contribution

The anomalous Hall (AHE) voltage can be expressed as, $V_{AHE} = R_{AHE} I$ and represented in the form:

$$V_{AHE} = k_a M_0 \cos \theta I, \quad (S6.1)$$

where M_0 is the intrinsic magnetisation value excluding spin accumulation, k_a is a proportionality constant, θ is the polar angle of the magnetisation and I is the applied current density.

In the presence of spin accumulation, S , which completes its transfer in the nanosecond time regime, this spin accumulation can be analogously viewed as localised magnetic moments in the relaxed state, since the period of the applied current density is significantly greater than the nanosecond regime. As such, this spin accumulation, S changes the magnetisation of the ferromagnetic layer from $M_0 m$ to $M_0 m + sm$, which leads to the re-expression of the AHE voltage given by,

$$V_{AHE} = k_a (M_0 + s) \cos \theta I = k_a (M_0 I + \zeta I^2) \cos \theta = V_{AHE,a} \cos \theta, \quad (S6.2)$$

where in the steady-state condition, the localised magnetic moment, S is proportional to the applied current density, $S = \zeta I$, as the rate of spin decay is equivalent to the spin generation, where ζ is the spin accumulation proportionality coefficient which can be related to the effective spin Hall angle of the film stack.

When a fixed DC current is added together with an AC current, the spins are preferentially accumulated at one interface more than the other, which enables the effective determination of the spin accumulation, such that $I = I_{ac} \sin \omega t + I_{dc}$, where ω is the angular frequency of the applied AC current.

$$\begin{aligned} \cos \theta &= \cos(\theta_0 + \Delta\theta) \\ &\approx \cos \theta_0 - \Delta\theta \sin \theta_0 = \cos \theta_0 - (\Delta\theta_{ac} \sin \omega t + \Delta\theta_{dc}) \sin \theta_0 \\ \cos \theta &\approx \cos \theta_0 - (\Delta\theta_{ac} \sin \omega t + \Delta\theta_{dc}) \sin \theta_0 \end{aligned} \quad (S6.3)$$

where $\Delta\theta_{ac}$ and $\Delta\theta_{dc}$ are polar angle modulations due to the ac current and dc current respectively.

$$\begin{aligned} V_{AHE,a} &= k_a (M_0 I + \zeta I^2) \\ &= k_a \left(M_0 [I_{dc} + I_{ac} \sin \omega t] + \zeta [I_{dc}^2 + 2I_{ac} I_{dc} \sin \omega t + I_{ac}^2 \sin^2 \omega t] \right) \\ &= k_a \left[\begin{aligned} &M_0 I_{dc} + \zeta I_{dc}^2 \\ &+ (M_0 I_{ac} + 2\zeta I_{ac} I_{dc}) \sin \omega t \\ &+ \zeta I_{ac}^2 \sin^2 \omega t \end{aligned} \right] \\ V_{AHE,a} &= k_a \left[M_0 I_{dc} + \zeta I_{dc}^2 + (M_0 I_{ac} + 2\zeta I_{ac} I_{dc}) \sin \omega t + \zeta I_{ac}^2 \sin^2 \omega t \right] \end{aligned} \quad (S6.4)$$

$$\begin{aligned}
V_{AHE}^{ac+dc} &= V_{AHE,a} \cos \theta \\
&\approx k_a \left[\cos \theta_0 \left[\begin{aligned} &\left(M_0 I_{ac} + 2\zeta I_{ac} I_{dc} \right) \sin \omega t \\ &+ \zeta I_{ac}^2 \sin^2 \omega t \end{aligned} \right] - \left[\begin{aligned} &\left(M_0 I_{dc} + \zeta I_{dc}^2 \right) (\Delta \theta_{ac} \sin \omega t) \\ &+ \left(M_0 I_{ac} + 2\zeta I_{ac} I_{dc} \right) (\Delta \theta_{ac} \sin^2 \omega t + \Delta \theta_{dc} \sin \omega t) \\ &+ \zeta I_{ac}^2 \sin^2 \omega t (\Delta \theta_{dc}) \end{aligned} \right] \sin \theta_0 \right] \\
&= k_a \left[\begin{aligned} &\left(\begin{aligned} &\left(M_0 I_{ac} + 2\zeta I_{ac} I_{dc} \right) \cos \theta_0 \\ &- \Delta \theta_{ac} \left(M_0 I_{dc} + \zeta I_{dc}^2 \right) \sin \theta_0 \\ &- \Delta \theta_{dc} \left(M_0 I_{ac} + 2\zeta I_{ac} I_{dc} \right) \sin \theta_0 \end{aligned} \right) \sin \omega t \\ &+ \left(\begin{aligned} &\zeta I_{ac}^2 \cos \theta_0 \\ &- \Delta \theta_{ac} \left(M_0 I_{ac} + 2\zeta I_{ac} I_{dc} \right) \sin \theta_0 \\ &- \Delta \theta_{dc} \zeta I_{ac}^2 \sin \theta_0 \end{aligned} \right) \sin^2 \omega t \end{aligned} \right] \\
&= R_{AHE} \left[\begin{aligned} &\left(\begin{aligned} &\left(I_{ac} + 2\frac{\zeta}{M_0} I_{ac} I_{dc} \right) \cos \theta_0 - \Delta \theta_{ac} \left(I_{dc} + \frac{\zeta}{M_0} I_{dc}^2 \right) \sin \theta_0 \\ &- \Delta \theta_{dc} \left(I_{ac} + 2\frac{\zeta}{M_0} I_{ac} I_{dc} \right) \sin \theta_0 \end{aligned} \right) \sin \omega t \\ &- \frac{1}{2} \left(\begin{aligned} &\frac{\zeta}{M_0} I_{ac}^2 \cos \theta_0 - \Delta \theta_{ac} \left(I_{ac} + 2\frac{\zeta}{M_0} I_{ac} I_{dc} \right) \sin \theta_0 \\ &- \Delta \theta_{dc} \frac{\zeta}{M_0} I_{ac}^2 \sin \theta_0 \end{aligned} \right) \cos 2\omega t \end{aligned} \right]
\end{aligned}$$

where $k_a = R_{AHE}/M_0$; $\varsigma = \frac{\zeta}{M_0}$.

$$V_{\omega,AHE}^{ac+dc} = R_{AHE} \left(\begin{aligned} &\left(I_{ac} + 2\varsigma I_{ac} I_{dc} \right) \cos \theta_0 - \Delta \theta_{ac} \left(I_{dc} + \varsigma I_{dc}^2 \right) \sin \theta_0 \\ &- \Delta \theta_{dc} \left(I_{ac} + 2\varsigma I_{ac} I_{dc} \right) \sin \theta_0 \end{aligned} \right), \quad (S6.5)$$

$$V_{2\omega,AHE}^{ac+dc} = -\frac{R_{AHE}}{2} \left(\begin{aligned} &\varsigma I_{ac}^2 \cos \theta_0 - \Delta \theta_{ac} \left(I_{ac} + 2\varsigma I_{ac} I_{dc} \right) \sin \theta_0 \\ &- \varsigma \Delta \theta_{dc} I_{ac}^2 \sin \theta_0 \end{aligned} \right), \quad (S6.6)$$

When there is no applied DC current, $I_{DC} = 0$, $\Delta \theta_{dc} = 0$, $\zeta \rightarrow 0$, since the current direction alternates.

$$V_{\omega,AHE}^{ac} = R_{AHE} I_{ac} \cos \theta_0 \approx I_{ac} \left(R_{AHE} - \frac{R_{AHE}}{2} \sin^2 \theta_0 \right), \quad (S6.7)$$

$$V_{2\omega,AHE}^{ac} = \frac{1}{2} R_{AHE} I_{ac} \Delta \theta_{ac} \sin \theta_0, \quad (S6.8)$$

The 1st and 2nd harmonic expressions arising from the AHE can be reduced to the case for the pure AC harmonic measurement analysis.

Planar Hall (PHE) Contribution

The planar Hall (AHE) voltage can be expressed as, $V_{PHE} = R_{PHE} I$ and represented in the form:

$$V_{PHE} = k_p M_0^2 \sin^2 \theta \sin 2\phi \quad (S6.9)$$

where M_0 is the intrinsic magnetisation value excluding spin accumulation, k_p is a proportionality constant, θ is the polar angle of the magnetisation, ϕ is the azimuthal angle of the magnetisation and I is the applied current density.

Similar to the AHE case, the V_{PHE} can be re-expressed in the form,

$$\begin{aligned} V_{PHE} &= k_p I (M_0 + s)^2 \sin^2 \theta \sin 2\phi \\ &\approx k_p I (M_0 + I\zeta)^2 \sin^2 (\theta_0 + \Delta\theta) \sin (2\phi_0 + \Delta\phi) \\ &\approx k_p (IM_0^2 + 2I^2\zeta + I^3\zeta^2) \sin^2 (\theta_0 + \Delta\theta) \sin (2\phi_0 + \Delta\phi) \\ &= V_{PHE,a} \sin^2 (\theta_0 + \Delta\theta) \sin (2\phi_0 + \Delta\phi) \end{aligned} \quad (S6.10)$$

Taking the harmonic amplitudes up to the second-order, and using the substitutions given by:

$$I = I_{ac} \sin \omega t + I_{dc} \quad (S6.11)$$

$$I^2 = (I_{ac} \sin \omega t + I_{dc})^2 = I_{ac}^2 \sin^2 \omega t + 2I_{dc} I_{ac} \sin \omega t + I_{dc}^2 \quad (S6.12)$$

$$I^3 = (I_{ac} \sin \omega t + I_{dc})^3 = I_{ac}^3 \sin^3 \omega t + 3I_{dc} I_{ac}^2 \sin^2 \omega t + 3I_{dc}^2 I_{ac} \sin \omega t + I_{dc}^3 \quad (S6.13)$$

$$\sin^2 (\theta_0 + \Delta\theta) = \sin^2 \theta_0 + 2\Delta\theta \sin \theta_0 \cos \theta_0 \quad (S6.14)$$

$$\sin 2(\phi_0 + \Delta\phi) = \sin 2\phi_0 + 2\Delta\phi \cos 2\phi_0 \quad (S6.15)$$

(S6.14) and (S6.15) can further be expressed as,

$$\sin^2 (\theta_0 + \Delta\theta) = \sin^2 \theta_0 + 2 \sin \theta_0 \cos \theta_0 \Delta\theta_{dc} + 2 \sin \theta_0 \cos \theta_0 \Delta\theta_{ac} \sin \omega t$$

$$\sin 2(\phi_0 + \Delta\phi) = \sin 2\phi_0 + 2 \cos 2\phi_0 \Delta\phi_{dc} + 2 \cos 2\phi_0 \Delta\phi_{ac} \sin \omega t$$

$$\sin^2 (\theta_0 + \Delta\theta) \cdot \sin 2(\phi_0 + \Delta\phi) \approx \left[\begin{aligned} &\left(\sin^2 \theta_0 (\sin 2\phi_0 + 2 \cos 2\phi_0 \Delta\phi_{dc}) \right) \\ &+ 2 \sin \theta_0 \cos \theta_0 \Delta\theta_{dc} (\sin 2\phi_0) \end{aligned} \right] \\ + \left[\begin{aligned} &2 \sin^2 \theta_0 \cos 2\phi_0 \Delta\phi_{ac} \\ &+ 2 \sin \theta_0 \cos \theta_0 \Delta\theta_{ac} (\sin 2\phi_0) \end{aligned} \right] \sin \omega t$$

where $\Delta\phi_{ac}$ and $\Delta\phi_{dc}$ are azimuthal angle oscillations induced by the applied ac and dc current, respectively.

$$\begin{aligned} V_{PHE,a} &= k_p \left(I M_0^2 + 2 I^2 \zeta + I^3 \zeta^2 \right) \\ &= k_p \left(A_{PHE} + B_{PHE} + C_{PHE} \right) \end{aligned} \quad (S6.16)$$

$$k_p A_{PHE} = \frac{R_{PHE}}{M_0^2} I M_0^2 = R_{PHE} \left(I_{dc} + I_{ac} \sin \omega t \right) \quad (S6.17)$$

$$k_p B_{PHE} = 2 R_{PHE} \frac{\zeta}{M_0^2} I^2 = \frac{2 R_{PHE} \zeta}{M_0^2} \left(I_{dc}^2 + 2 I_{dc} I_{ac} \sin \omega t + I_{ac}^2 \sin^2 \omega t \right) \quad (S6.18)$$

$$k_p C_{PHE} = \frac{R_{PHE}}{M_0^2} \zeta^2 I^3 \approx R_{PHE} \zeta^2 \left(I_{dc}^3 + 3 I_{dc}^2 I_{ac} \sin \omega t + 3 I_{dc} I_{ac}^2 \sin^2 \omega t \right) \quad (S6.19)$$

where $\zeta = \zeta / M_0$, $k_p = R_{PHE} / M_0^2$. As ζ^2 is small,

$$V_{PHE,a} \approx k_p \left(I M_0^2 + 2 I^2 \zeta \right) = k_p \left(A_{PHE} + B_{PHE} \right), \quad (S6.20)$$

Substituting the coefficients, we can expand and express $V_{\omega, PHE}^{ac+dc}$ as,

$$V_{\omega, PHE}^{ac+dc} \approx R_{PHE} \left[\begin{aligned} &\left(I_{ac} + \frac{4\zeta}{M_0} I_{dc} I_{ac} \right) \sin^2 \theta_0 \sin 2\phi_0 \\ &+ \sin 2\theta_0 \sin 2\phi_0 \left(\frac{I_{ac} \Delta \theta_{dc} + I_{dc} \Delta \theta_{ac}}{M_0} + \frac{2\zeta}{M_0} I_{dc}^2 \Delta \theta_{ac} + \frac{4\zeta}{M_0} I_{dc} I_{ac} \Delta \theta_{dc} \right) \\ &+ \sin^2 \theta_0 \cos 2\phi_0 \left(\frac{2 I_{ac} \Delta \phi_{dc} + 2 I_{dc} \Delta \phi_{ac}}{M_0} + \frac{8\zeta}{M_0} I_{dc} I_{ac} \Delta \phi_{dc} + \frac{4\zeta}{M_0} I_{dc}^2 \Delta \phi_{ac} \right) \end{aligned} \right], \quad (S6.21)$$

$$V_{2\omega, PHE}^{ac+dc} \approx -\frac{1}{2} R_{PHE} \left[\begin{aligned} &I_{ac} \left(2 \sin^2 \theta_0 \cos 2\phi_0 \Delta \phi_{ac} + \sin 2\theta_0 \sin 2\phi_0 \Delta \theta_{ac} \right) \\ &+ \frac{2\zeta}{M_0} \left(\sin^2 \theta_0 \cos 2\phi_0 \left(\frac{4 I_{dc} I_{ac} \Delta \phi_{ac}}{M_0} + 2 I_{ac}^2 \Delta \phi_{dc} \right) + \sin 2\theta_0 \sin 2\phi_0 \left(\frac{2 I_{dc} I_{ac} \Delta \theta_{ac}}{M_0} + I_{ac}^2 \Delta \theta_{dc} \right) \right) \end{aligned} \right] \quad (S6.22)$$

When there is no applied DC current, $I_{DC} = 0$, $\Delta \theta_{dc} = 0$, $\zeta \rightarrow 0$, since the current direction alternates, and does not have a net effect on the magnetisation.

$$V_{\omega, PHE}^{ac} \approx I_{ac} R_{PHE} \sin^2 \theta_0 \sin 2\phi_0 \quad (S6.23)$$

$$V_{2\omega, PHE}^{ac} \approx - \left(I_{ac} R_{PHE} \sin^2 \theta_0 \cos 2\phi_0 \Delta \phi_{ac} + \frac{1}{2} I_{ac} R_{PHE} \sin \theta_0 \cos \theta_0 \sin 2\phi_0 \Delta \theta_{ac} \right) \quad (S6.24)$$

In the longitudinal scheme of measurement, the first, $V_{\omega,ac}^{dc} \Big|_{\parallel}$ and second, $V_{2\omega,ac}^{dc} \Big|_{\parallel}$ harmonic Hall voltages can be expressed as,

$$V_{\omega,ac}^{dc} \Big|_{\parallel} = R_{AHE} \left(\begin{aligned} & \left(I_{ac} + 2\zeta I_{ac} I_{dc} \right) \cos \theta_0 - \Delta \theta_{ac} \left(I_{dc} + \zeta I_{dc}^2 \right) \sin \theta_0 \\ & - \Delta \theta_{dc} \left(I_{ac} + 2\zeta I_{ac} I_{dc} \right) \sin \theta_0 \end{aligned} \right) + 2R_{PHE} \sin^2 \theta_0 \left(I_{ac} \Delta \phi_{dc} + I_{dc} \Delta \phi_{ac} + \frac{4\zeta}{M_0} I_{dc} I_{ac} \Delta \phi_{dc} + \frac{2\zeta}{M_0} I_{dc}^2 \Delta \phi_{ac} \right), \quad (S6.25)$$

$$V_{2\omega,ac}^{dc} \Big|_{\parallel} = -\frac{R_{AHE}}{2} \left(\begin{aligned} & \zeta I_{ac}^2 \cos \theta_0 - \Delta \theta_{ac} \left(I_{ac} + 2\zeta I_{ac} I_{dc} \right) \sin \theta_0 \\ & - \zeta \Delta \theta_{dc} I_{ac}^2 \sin \theta_0 \end{aligned} \right) - R_{PHE} \sin^2 \theta_0 \left(I_{ac} \Delta \phi_{ac} + \frac{\zeta}{M_0} \left(4I_{dc} I_{ac} \Delta \phi_{ac} + 2I_{ac}^2 \Delta \phi_{dc} \right) \right), \quad (S6.26)$$

In the transverse scheme of measurement, the first, $V_{\omega,ac}^{dc} \Big|_{\perp}$ and second, $V_{2\omega,ac}^{dc} \Big|_{\perp}$ harmonic Hall voltages can be expressed as,

$$V_{\omega,ac}^{dc} \Big|_{\perp} = R_{AHE} \left(\begin{aligned} & \left(I_{ac} + 2\zeta I_{ac} I_{dc} \right) \cos \theta_0 - \Delta \theta_{ac} \left(I_{dc} + \zeta I_{dc}^2 \right) \sin \theta_0 \\ & - \Delta \theta_{dc} \left(I_{ac} + 2\zeta I_{ac} I_{dc} \right) \sin \theta_0 \end{aligned} \right) - 2R_{PHE} \sin^2 \theta_0 \left(I_{ac} \Delta \phi_{dc} + I_{dc} \Delta \phi_{ac} + \frac{4\zeta}{M_0} I_{dc} I_{ac} \Delta \phi_{dc} + \frac{2\zeta}{M_0} I_{dc}^2 \Delta \phi_{ac} \right), \quad (S6.27)$$

$$V_{2\omega,ac}^{dc} \Big|_{\perp} = -\frac{R_{AHE}}{2} \left(\begin{aligned} & \zeta I_{ac}^2 \cos \theta_0 - \Delta \theta_{ac} \left(I_{ac} + 2\zeta I_{ac} I_{dc} \right) \sin \theta_0 \\ & - \zeta \Delta \theta_{dc} I_{ac}^2 \sin \theta_0 \end{aligned} \right) + R_{PHE} \sin^2 \theta_0 \left(I_{ac} \Delta \phi_{ac} + \frac{\zeta}{M_0} \left(4I_{dc} I_{ac} \Delta \phi_{ac} + 2I_{ac}^2 \Delta \phi_{dc} \right) \right), \quad (S6.28)$$

(S6.25) to (S6.28) can be generically expressed as,

$$V_{\omega,ac}^{dc} \Big|_{\parallel(\perp)} = R_{AHE} \left(\begin{aligned} & \left(I_{ac} + 2\zeta I_{ac} I_{dc} \right) \cos \theta_0 - \Delta \theta_{ac} \left(I_{dc} + \zeta I_{dc}^2 \right) \sin \theta_0 \\ & - \Delta \theta_{dc} \left(I_{ac} + 2\zeta I_{ac} I_{dc} \right) \sin \theta_0 \end{aligned} \right) \pm 2R_{PHE} \sin^2 \theta_0 \left(I_{ac} \Delta \phi_{dc} + I_{dc} \Delta \phi_{ac} + \frac{4\zeta}{M_0} I_{dc} I_{ac} \Delta \phi_{dc} + \frac{2\zeta}{M_0} I_{dc}^2 \Delta \phi_{ac} \right), \quad (S6.29)$$

$$V_{2\omega,ac}^{dc} \Big|_{\parallel(\perp)} = -\frac{R_{AHE}}{2} \left(\begin{aligned} & \zeta I_{ac}^2 \cos \theta_0 - \Delta \theta_{ac} \left(I_{ac} + 2\zeta I_{ac} I_{dc} \right) \sin \theta_0 \\ & - \zeta \Delta \theta_{dc} I_{ac}^2 \sin \theta_0 \end{aligned} \right) \mp R_{PHE} \sin^2 \theta_0 \left(I_{ac} \Delta \phi_{ac} + \frac{\zeta}{M_0} \left(4I_{dc} I_{ac} \Delta \phi_{ac} + 2I_{ac}^2 \Delta \phi_{dc} \right) \right), \quad (S6.30)$$

For a system with both ac and dc in the harmonic measurements, the analytical equations can be expressed as,

$$\Delta\theta_{dc} = \frac{\Delta H_{x,dc} \cos\phi_0 + \Delta H_{y,dc} \sin\phi_0}{(H_{k,eff} + H_z)} ; \quad \Delta\theta_{ac} = \frac{\Delta H_{x,ac} \cos\phi_0 + \Delta H_{y,ac} \sin\phi_0}{(H_{k,eff} + H_z)}, \quad (S6.31)$$

$$\Delta\phi_{dc} = \frac{\Delta H_{y,dc} H_x - \Delta H_{x,dc} H_y}{H_x^2 + H_y^2} ; \quad \Delta\phi_{ac} = \frac{\Delta H_{y,ac} H_x - \Delta H_{x,ac} H_y}{H_x^2 + H_y^2} \quad (S6.32)$$

$$\sin\theta_0 = \frac{H}{H_{k,eff} + H_z} \quad (S6.33)$$

where $\Delta H_{x,dc}$ and $\Delta H_{y,dc}$ is the DL-SOT fields, $\Delta H_{x,ac}$ and $\Delta H_{y,ac}$ are the FL-SOT fields induced by the ac and dc current densities respectively, H_x and H_y are the applied magnetic fields in the x - and y -directions respectively, and H is either H_x or H_y .

Utilising the damping-like and field-like (FL) efficiency, ξ_{DL} and ξ_{FL} respectively, determined previously, we are able to determine the current dependency of both the ac and dc SOT fields through the relation,

$$\Delta H_{x(y),ac/dc} = \frac{\hbar J_{ac/dc} \xi_{DL(FL)}}{2eM_s t}, \quad (S6.34)$$

(S6.29) to (S6.32) are substituted into (S6.28), to attain a generical second harmonic Hall expression given by,

$$R_{2\omega,ac}^{dc} \Big|_{\parallel(\perp)} = \left(-\frac{R_{AHE}}{2} \varsigma I_{ac} + \frac{R_{AHE}}{4} \varsigma I_{ac} \left(\frac{H_{x(y)}}{H_{k,eff} + H_z} \right)^2 + Z^{x(y)} H_{x(y)} \right), \quad (S6.35)$$

$$Z^{x(y)} = \frac{\hbar}{2eM_s t (H_{k,eff} + H_z)^2} \left[\frac{R_{AHE} \xi_{DL(FL)}}{2} \left(\frac{J_{ac} (1 + 2\varsigma I_{dc})}{+ \varsigma J_{dc} I_{ac}} \right) \right. \\ \left. \mp \gamma R_{AHE} \xi_{FL(DL)} \left(J_{ac} + \frac{\varsigma}{M_s} \left(\frac{4I_{dc} J_{ac}}{+ 2I_{ac} J_{dc}} \right) \right) \right], \quad (S6.36)$$

where $\xi_{DL(FL)}$ is the effective DL or FL efficiency coefficient, γ is the PHE correction factor, e is the electron charge, $M_s = M_0$ is the saturation magnetisation, t is the thickness of the FM layer, α and δ are the ac and dc applied current densities in orders of 10^{10} A/m² respectively.

$R_{2\omega,ac}^{dc}$ behaves like a parabolic function with H , such that,

$$f(H) \equiv R_{2\omega,ac}^{dc} \equiv A + Z^{x(y)} H_{x(y)} + B H_{x(y)}^2, \quad (S6.37)$$

where

$$A = -\frac{1}{2} R_{AHE} \varsigma I_{ac} , \quad (S6.38)$$

$$Z = \frac{\hbar}{2eM_s t (H_{k,eff} + H_z)^2} \left[\frac{\xi_{DL(FL)}}{2} \left(J_{ac} (1 + 2\varsigma I_{dc}) \right) \mp \gamma \xi_{FL(DL)} \left(J_{ac} + \frac{\varsigma}{M_s} \left(4I_{dc} J_{ac} \right) \right) \right] \quad (S6.39)$$

$$B = \frac{R_{AHE} \varsigma I_{ac}}{4 (H_{k,eff} + H_z)^2} , \quad (S6.40)$$

By finding the partial derivative with respect to I_{dc} ,

$$\left(\frac{\partial A}{\partial I_{dc}} \right) = 0 \quad ; \quad \left(\frac{\partial B}{\partial I_{dc}} \right) = 0 \quad ; \quad \left(\frac{\partial Z^{x(y)}}{\partial I_{dc}} \right) \neq 0 , \quad \text{as such only the } Z^{x(y)} \text{ coefficient}$$

corresponds to a change in I_{dc} which relates to $\varsigma = \zeta / M_s$.

$$\frac{\partial Z^{x(y)}}{\partial I_{dc}} = \frac{3\varsigma J_{ac} \hbar R_{AHE}}{2eM_s t (H_{k,eff} + H_z)^2} \left[\frac{\xi_{DL(FL)}}{2} \mp \frac{2\gamma \xi_{FL(DL)}}{M_s} \right] , \quad (S6.41)$$

Rearranging the terms,

$$\varsigma = \left(\frac{\partial Z^{x(y)}}{\partial I_{dc}} \right) \cdot \frac{2eM_s t (H_{k,eff} + H_z)^2}{3J_{ac} \hbar R_{AHE} \left[\xi_{DL(FL)} / 2 \mp 2\gamma \xi_{FL(DL)} / M_s \right]} , \quad (S6.42)$$

The spin accumulation coefficient, ζ can expressed as,

$$\zeta = \varsigma M_s = \left(\frac{\partial Z^{x(y)}}{\partial I_{dc}} \right) \cdot \frac{2eM_s^2 t (H_{k,eff} + H_z)^2}{3J_{ac} \hbar R_{AHE} \left[\xi_{DL(FL)} / 2 \mp 2\gamma \xi_{FL(DL)} / M_s \right]} , \quad (S6.43)$$

$\left(\frac{\partial Z^{x(y)}}{\partial I_{dc}} \right)$ can be attained experimentally by plotting the function of Z against I_{dc} and

determined to calculate ζ and subsequently, the spin accumulation, s . The percentage spin accumulation with respect to the magnetisation is given by:

$$\%SpinAccumulation = \frac{s}{M_s} , \quad (S6.44)$$

Thermoelectric Contribution

In metallic systems, the most significant thermoelectric contribution is due to the anomalous Nernst effect (ANE), which produces an electric field given as,

$$E_{ANE} = -a \nabla T \times m, \quad (S6.45)$$

where a is the ANE coefficient.

$$E_{ANE} = \begin{pmatrix} \nabla T_x \\ \nabla T_y \\ \nabla T_z \end{pmatrix} \times \begin{pmatrix} m_x \sin \theta \cos \phi \\ m_y \sin \theta \sin \phi \\ m_z \cos \theta \end{pmatrix} = \begin{pmatrix} \nabla T_y m_z \cos \theta - \nabla T_z m_y \sin \theta \sin \phi \\ \nabla T_z m_x \sin \theta \cos \phi - \nabla T_x m_z \cos \theta \\ \nabla T_x m_y \sin \theta \sin \phi - \nabla T_y m_x \sin \theta \cos \phi \end{pmatrix}, \quad (S6.46)$$

In order to determine the ANE contributions from the various planes, i.e. y - z , x - z and x - y planes, large fields are swept such that the samples are saturated in the various, x , y , z -directions where m remains only in the m_x , m_y and m_z , respectively and, the expression can be written as,

$$H_x \text{ field sweep: } E_{ANE} = \begin{pmatrix} 0 \\ \nabla T_z m_x \\ -\nabla T_y m_x \end{pmatrix} \sin \theta \cos \phi \quad (y\text{-}z \text{ plane}) \quad (S6.47)$$

$$H_y \text{ field sweep: } E_{ANE} = \begin{pmatrix} -\nabla T_z m_y \\ 0 \\ \nabla T_x m_y \end{pmatrix} \sin \theta \sin \phi \quad (x\text{-}z \text{ plane}) \quad (S6.48)$$

$$H_z \text{ field sweep: } E_{ANE} = \begin{pmatrix} \nabla T_y m_z \\ -\nabla T_x m_z \\ 0 \end{pmatrix} \cos \theta \quad (x\text{-}y \text{ plane}) \quad (S6.49)$$

where $\nabla T_{x,y,z}$ is the temperature gradient occurring in the various planes of geometry.

Considering Joule heating as the sole source of the thermal gradient, it can be described via the expression,

$$V_{ANE} \propto \nabla T \propto I^2 R_s, \quad (S6.50)$$

where ∇T is the temperature gradient, R_s is the sample resistance. For an ac current, the thermal gradient, ∇T_{ac} can be expressed as:

$$\nabla T_{ac} \propto (I_{ac}^2 \sin^2 \omega t) R_s = \frac{1}{2} I_{ac}^2 [1 - \cos 2\omega t] R_s, \quad (S6.51)$$

For the case when dc current is added, we can express the ∇T_{ac+dc} as:

$$\nabla T_{ac+dc} \propto (I_{ac} \sin \omega t + I_{dc})^2 R_s, \quad (S6.52)$$

$$\nabla T_{ac+dc} = \left(I_{dc}^2 + \frac{1}{2} J_{ac}^2 + 2 I_{dc} I_{ac} \sin \omega t - \frac{1}{2} I_{ac}^2 \cos 2\omega t \right) R_s, \quad (S6.53)$$

$$\nabla T_{\omega}^{ac+dc} = 2 R_s I_{dc} I_{ac} \sin \omega t, \quad (S6.54)$$

$$\nabla T_{2\omega}^{ac+dc} = -\frac{1}{2} R_s J_{ac}^2 \cos 2\omega t, \quad (S6.55)$$

The thermal contribution to the second harmonic Hall voltage exists similarly to the case of the pure ac case. As $R_H^{2\omega}$ is a dynamic effect resulting from the magnetisation oscillations, but the thermal contribution arises due to a static effect entering the $R_H^{2\omega}$. To ignore the SOT contributions, the H_{ext} should be sufficiently large to force magnetisation to rigidly align along the direction of the field. When saturated, the R_{ANE} in the x-y plane can be determined, i.e. the field is swept in the H_z direction, and the $V_{2\omega}^{ANE}$ can be determined as the SOT fields, and Oersted fields do not contribute to the $V_{2\omega}$. Likewise, the field can be swept in the H_x and H_y direction for the y-z and x-z planes, respectively. When the magnetisation is close to saturation in the x or y direction, the difference in the V_{ω} at saturation will correspond to the R_{PHE} while the $V_{2\omega}$ at saturation will correspond to thermoelectric contribution. As such, $R_{2\omega}^{ANE}$ can be determined for various DC and AC current densities, and thus be used to determine a corrected second harmonic signal given by^{54,184},

$$R_{2\omega,corr} = R_{2\omega,raw} - R_{\omega} \frac{R_{ANE}|_{xy-plane} + \frac{1}{\gamma} (R_{ANE}|_{xz-plane} + R_{ANE}|_{yz-plane})}{2R_{AHE}}, \quad (S6.56)$$

where γ is the PHE correction factor. Thus, upon correction, the second harmonic signal should only have contributions which relate purely to the spin accumulation and SOT fields.

Raw Experimental 2nd Harmonic Hall Resistance

Figure S6.1. Raw 2nd harmonic Hall resistance as a function of H_x field for varying thickness of Ta and Tb when a dc bias current is swept from $\pm 4 \times 10^{10}$ to $\pm 6 \times 10^{10}$ A/m².

Figure S6.2. Raw 2nd harmonic Hall resistance as a function of H_y field for varying thickness of Ta and Tb when a dc bias current is swept from $\pm 4 \times 10^{10}$ to $\pm 6 \times 10^{10}$ A/m².

In both Figure S6.1 and S6.2, in the longitudinal and transverse scheme, respectively, the second harmonic Hall resistance reveals an anomalous parabolic relation before the thermoelectric correction was done, due to the addition of a dc bias into the measurement current.

Determination of ANE in the x-y, x-z, and y-z plane

Experimental determination of the ANE contributions to the second harmonic Hall voltage can be determined through a field saturation in the x-field (longitudinal scheme) and y-field (transverse scheme) for the y-z and x-z plane thermoelectric contribution, respectively. For the sake of comparison, the large field sweep is done at ac-only current density, $J_{ac} = 1 \times 10^{11} \text{ A/m}^2$ which reveals a hysteresis-like behaviour depicted in the first harmonic Hall resistances as shown in Figure S6.3 (a) and (b), for the longitudinal and transverse scheme of measurement, respectively. The corresponding second harmonic Hall resistances are shown in Figure S6.3 (c) and (d) across the various samples of $t_{tb} = 5 \text{ nm}$ and $5 \leq t_{tb} \leq 8 \text{ nm}$.

Figure S6.3. Large field sweep regime harmonic measurement without dc bias. (a) and (b) showing the first harmonic, (c) and (d) showing the second harmonic Hall resistances in the longitudinal and transverse measurement, respectively.

Figure S6.4 and Figure 6.5 shows the determination of the $R_{ANE,xz}$ and $R_{ANE,yz}$, in a large in-plane field sweep in the large x-field and y-field regime, respectively. This is accomplished through the application of fixed $J_{ac} = 4 \times 10^{10} \text{ A/m}^2$ for J_{dc} ranging from $1 \times 10^{10} \text{ A/m}^2$ to $6 \times 10^{10} \text{ A/m}^2$ with an increment of 10^{10} A/m^2

Figure S6.4. Large field regime harmonic measurement with dc bias in the longitudinal scheme of measurement(H_x sweep).

Figure S6.5. Large field regime harmonic measurement with dc bias in the transverse scheme of measurement (H_y sweep).

Chapter 7: Conclusion & Future Outlook

7.1 Thesis Summary

In this thesis, the fundamental investigation of SOT effective fields in both single ferromagnetic and multilayer structures was conducted given the potential of SOT in future industrial applications. As such, this thesis investigates SOT in Hall bar devices through an *in-situ* Kerr and harmonic Hall method, H_z harmonic Hall method and a DC biased H_z harmonic Hall method.

In chapter 4 of this thesis, the experimental determination of the SOT effective fields in a MgO/CoFeB/Ta structure utilising a combination of the harmonic measurement and *in-situ* Kerr imaging was reported. The SOT effective fields were determined through an analytical energy approach by transforming the anomalous Hall effect and planar Hall effect (PHE) voltage into a field dependency while imaging the magnetisation behaviour by differential Kerr microscopy. The analytical fitting to the measurement data indicates the significant coexistence of both transverse field, H_T , and longitudinal field, H_L , in the longitudinal ($H_L = -12$ Oe, $H_T = 8$ Oe per 10^6 A cm $^{-2}$) and transverse ($H_L = -12$ Oe, $H_T = -17$ Oe per 10^6 A cm $^{-2}$) measurement schemes, respectively, due to the PHE. Additionally, the dendritic-like domains which indicate the influence of the interfacial Dzyaloshinskii–Moriya interaction (DMI) at the CoFeB/Ta interface, were observed by *in-situ* Kerr imaging. Micromagnetic simulations were performed to confirm the dendritic domain formation and edge tilting of the magnetisation, as result of DMI.

In chapter 5 of this thesis, the enhanced spin-orbit torque (SOT) effect due to the rare-earth metal Tb in a Pt/[Co/Ni] $_2$ /Co/Tb multilayer with perpendicular magnetic anisotropy (PMA) was reported. Due to the large spin-orbit coupling in Tb, the effective damping like efficiency, ξ_{DL}^{eff} , was determined to be 0.55 for 9-nm Tb as compared to $\xi_{DL}^{eff} = 0.18$ for the reference Pt/[Co/Ni] $_2$ /Co/Ta stack. A relatively sizeable effective spin Hall efficiency of Tb, $\theta_{SH,Tb}^{eff}$, was determined to be -0.480 for Tb with a 9nm through a H_z -bias harmonic measurement. Enhanced magnetisation switching efficiency was

observed with increasing Tb thickness, thus affirming the sizable anti-damping torque responsible for deterministic switching. Furthermore, as the Co/Tb interface has more substantial interfacial coupling than in Co/Ta interface, strong angular dependence of the damping-like and field-like terms was present. This angular dependence diminishes with increasing Tb thickness. These results affirm that Tb plays a significant role in tuning the SOT efficiency through the variation of Tb concentration, which led to an enhancement of the effective spin Hall angle and switching efficiency.

In chapter 6 of this thesis, an application of a dc bias alternating current in the structure induces an anomalous second harmonic Hall resistance which is not entirely due to the widely investigated SOT. Analytical derivations demonstrate that apart from the SOT, spin accumulation can contribute to the second harmonic Hall resistance. This spin accumulation magnetoresistance indicates that the magnetisation of Co/Ni multilayers can be altered via spin injection from HM and R materials. As such, on top of the SOT-induced magnetisation switching, the spin accumulation is postulated to play a vital role in the SOT reversal process. Experimental quantification of the spin accumulation induced by the electrical current in stacks of Pt/[Co/Ni]₂/Co/Tb (5-8nm) or Pt/[Co/Ni]₂/Co/Ta (5nm) have shown that spin accumulation could be as much as 6.6% of the local magnetisation when the total current density applied is 10^{11} A/m². The ratio of spin accumulation over the applied electric current is consistent with the damping like efficiencies. Hence, the ratio can be used to evaluate the spin current conversion efficiency of heavy metal or R metal in converting electrical current to spin current. This provides an all-electrical alternative to determine spin accumulation through a commonly used harmonic Hall resistance approach.

7.2 The Future of Spin-Orbitronics

7.2.1 Influence of DMI on SOT Switching

Having investigated the enhancement of SOT in Pt-Rare Earth systems as presented in Chapter 5 of the thesis, one can expect either an enhancement or cancellation of SOT by changing the material of the capping layer. This is done by sandwiching a FM between two different metals of opposite spin Hall angle. As a result of the change in material, the magnitude and sign of DMI can be tuned as well^{73,141,185}. Thus the induced Neel wall with different chiralities can be driven either more efficiently or less efficiently by the SHE depending on the type of materials used. In earlier proposals of nucleation driven magnetisation reversal of inversion asymmetric structures¹⁸⁶, the DMI field must be overcome by a large external magnetic field in order to enable spin Hall driven expansion of the nucleated domains. In J. W. Yu *et al.*¹⁸⁷, they report that large negative DMI requires a larger in-plane field for full magnetisation switching, and DMI structural engineering can be done separately from SOT. As such, further investigation on the DMI in relation to the SOT can be further studied to determine an optimally engineered DMI value to improve the switching efficiency of the SOT device.

We propose to utilise a microwire expansion method to have better control over the magnetic domains, based on the principles of the asymmetric DW expansion of circular DWs previously reported^{188–190}, where the circular domain will result in an elliptical domain when an in-plane magnetic field is applied. The benefit of utilising the microwire method as compared to the bubble expansion method is that the injection line allows a control nucleation site at the centre of the microwire. This removes the need to locate isolated nucleation sites which may not exist on thin films. This method also prevents the interaction between the neighbouring nucleation sites, as such large OOP fields and IP fields can be applied to observe the domain expansion. Figure 7.1 (a) shows the proposed geometry of microwires with an injection line in which DMI can be studied in the film structure, (b) shows the asymmetric expansion of the DWs when $+H_x$ and $-H_x$ are applied, and the graph of left DW and right DW against the in-plane field for samples (c) MgO/CoFeB/Ta, and (d) Pt/[Co/Ni]₂/Co/Tb (5nm). The measurement

parameters were attained through determining the most optimal injection current and applied OOP field for domain nucleation to occur at the centre of the wire. After nucleation, the DWs were propagated through the use of an OOP field, and IP field applied one after another. This propagation can be measured in terms of pixels using a Kerr microscope and subsequently used to calculate the DW velocity. The H_{DMI} field is determined at the minimum DW velocity in which the applied in-plane field equalises with that of the H_{DMI} . Thus, preliminarily the sample MgO/CoFeB (1.4nm)/W (5nm) is determined to have DMI strength of 0.5 mJ/m², while the sample Pt/[Co/Ni]₂/Co/Tb(5nm) has DMI strength of 0.27 mJ/m².

Figure 7.1 (a) shows the proposed geometry of microwires with an injection line in which DMI can be studied in the film structure, (b) shows the asymmetric expansion of the DWs when $+H_x$ and $-H_x$ are applied, and the graph of left DW and right DW against the in-plane field for samples (c) MgO/CoFeB/Ta, and (d) Pt/[Co/Ni]₂/Co/Tb (5nm)

7.2.2 Voltage Control Magnetic Anisotropy (VCMA)-Coupled SOT Switching

Following the work of this thesis, upon the characterisation of the SOT fields using a H_z bias harmonic Hall technique, the control of the K_u of the system will provide the potential for greater understanding of the underlying physics in *in-situ* VCMA-harmonic measurements. This can be done through the application of an electric field to control and modulate the H_k of the material. A proposed schematic of the device is demonstrated in Figure 7.2.

Figure 7.2 Schematic diagram illustrating the possible geometry for a VCMA-harmonic Hall measurement and the cross-sectional view of the stack structure.

Harmonic measurements on the device can be carried out in its pristine state without any application of a gate voltage. Subsequently, an application of a sufficiently high gate voltage of $\sim \pm 0.8 \text{ MV/cm}$ for a period of 2000s. In essence, the application of a gate voltage alters the $H_{k,eff}$ of the sample where coefficients of the harmonic Hall voltages can be expressed as,

$$V_{\omega} \approx \pm \frac{1}{2} R_H I_0 \left(1 - \frac{H}{H_{k,eff}} \right)^2, \quad (7.1)$$

$$V_{2\omega} \approx - \left(\pm \frac{1}{4} R_H I_0 \right) \Delta H_{DL,FL} \frac{H}{H_{k,eff}^2}, \quad (7.2)$$

where V_{ω} and $V_{2\omega}$ are the first and second harmonic amplitude, respectively. R_H is the Hall resistance, I_0 is the current, $H_{k,eff}$ is the effective anisotropic field, H is the applied external magnetic field and $\Delta H_{DL,FL}$ are the SOT fields.

7.2.3 Azimuthal Angular Dependence of Spin Accumulation

Having quantified the spin accumulation in chapter 6 of this thesis in both the longitudinal ($\phi = 0^\circ$) and transverse ($\phi = 90^\circ$) measurement schemes, the second harmonic voltage could be further generalised to the azimuthal angle dependence, in which one can investigate the dependence of the magnitude of spin accumulation in relation to the azimuthal angle of the magnetisation in a perpendicularly magnetised stack.

$$V_{2\omega, AHE}^{ac+dc} = -\frac{1}{4} \left(\zeta I_{ac}^2 \left(1 - \frac{H^2}{(H_{k,eff} + H_z)^2} \right) - \frac{H}{(H_{k,eff} + H_z)^2} \left((\Delta H_{x,ac} \cos \phi + \Delta H_{y,ac} \sin \phi) (R_{AHE} I_{ac} + 2\zeta I_{ac} I_{dc}) \right) + (\Delta H_{x,dc} \cos \phi + \Delta H_{y,dc} \sin \phi) \zeta I_{ac}^2 \right)$$

$$V_{2\omega, PHE}^{ac+dc} = \left(\sin^2 \theta_0 \frac{\cos 2\phi}{H_x^2 + H_y^2} \left[R_{PHE} I_{ac} \begin{pmatrix} \Delta H_{y,ac} H_x \\ -\Delta H_{x,ac} H_y \end{pmatrix} + 2\zeta \begin{pmatrix} 4I_{dc} I_{ac} (\Delta H_{y,ac} H_x - \Delta H_{x,ac} H_y) \\ +2I_{ac}^2 (\Delta H_{y,dc} H_x - \Delta H_{x,dc} H_y) \end{pmatrix} \right] \right. \\ \left. + \sin 2\theta_0 \frac{\sin 2\phi}{H_{k,eff} + H_z} \left[\frac{1}{2} R_{PHE} I_{ac} \begin{pmatrix} \Delta H_{x,ac} \cos \phi \\ +\Delta H_{y,ac} \sin \phi \end{pmatrix} + 2\zeta \begin{pmatrix} 2I_{dc} I_{ac} (\Delta H_{x,ac} \cos \phi + \Delta H_{y,ac} \sin \phi) \\ +I_{ac}^2 (\Delta H_{x,dc} \cos \phi + \Delta H_{y,dc} \sin \phi) \end{pmatrix} \right] \right. \\ \left. + 2\zeta I_{ac}^2 \sin^2 \theta_0 \sin 2\phi \right)$$

Preliminary work on the azimuthal angular dependence from macro-spin simulation has shown shifts in the parabolic relation in both the first and second harmonic Hall resistance, which can be attributed to the SOT effective fields as well as the spin accumulation present, as shown in Figure 7.3.

Figure 7.3 First harmonic Hall relation (left) and the second harmonic Hall relation with respect to the varying azimuthal angle, ϕ , $J_{dc} = 6 \times 10^{10} \text{ A/m}^2$ and $J_{ac} = 4 \times 10^{10} \text{ A/m}^2$.

7.2.4 Alternative Field-Free Switching SOT

In this thesis, although the samples of study have shown enhanced SOT, field-free switching was not realised. The samples studied required an in-plane field to break symmetry for current-induced magnetisation reversal to occur. SOT devices must switch without a field for it to be useful for industrial applications. As such, there is a need to investigate alternative mechanisms for field-free SOT switching. We propose using a structure with an IMA-coupled with PMA structure as shown in Figure 7.4, where the IMA layer not only provides a stray field to break symmetry but provide an additional spin z component as reported by S. C Baek *et al.*¹⁹¹, which favours opposite magnetisation directions on the top PMA-FM layer for opposite current directions.

Figure 7.4 *Proposed structure of the FM trilayer, with the bottom FM having IMA and the top recording layer, with PMA-FM.*

In Baek *et al.*, they reported field-free switching using Ti instead of the conventional HMs such as Ta, Pt or W, in which most of the contribution comes from the IMA-FM layer. As such, by using an IMA at the bottom, it will facilitate symmetry breaking and provide an additional spin z component coupled with the HM layer contribution. This could potentially improve SOT switching efficiency.

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Appendix

Macro-Spin Simulation

Harmonic Hall Measurement

Harmonic Hall macro-spin simulations were conducted to simulate the harmonic Hall expresses as attained experimentally in the first and second harmonic Hall measurements. This is done by assuming a fixed SHE-induced and Rashba induced SOT fields given by the equations,

$$H_{SHE} = H_{SHE} (\mathbf{m} \times \hat{y}), \quad (A1)$$

$$H_R = H_R \hat{y}, \quad (A2)$$

where $\mathbf{m}$ is the unit vector of the magnetisation, $\hat{y}$ is the unit vector of the spin-polarised current induced in the HM layer. These SOT fields contribute to the effective field that the magnetisation experiences during the measurement given by,

$$H_{eff} = H_{SHE} + H_R + H_{k,eff} + H_z + H_{x(y)} + H_{sw}, \quad (A3)$$

where $H_{k,eff}$ is the effective anisotropic field of the sample, H_z is the field applied in the sample in the z-direction, $H_{x(y)}$ is the dynamic field that is swept, H_{sw} is the field-induced when a small angle is tilting the sample in the polar direction. In the large field sweep, where the film is tilted at an angle of 4° and swept between ± 1 T, the 1st and 2nd harmonic simulation results are as illustrated in Figure A1.1

Figure A1.1 Macro-spin simulation results for harmonic Hall voltage in the longitudinal and transverse scheme of measurements when no H_z bias field is applied in a magnetic field sweep between ± 1 T.

In the presence of a tilted sample, there will exist a small sweeping field in the z -direction as the H_x or H_y field is swept which results in a field-induced switching of the magnetisation when the applied external magnetic field is swept in the measurement setup. With the application of a bias H_z field, switching will no longer occur in the 1st harmonic Hall voltage which results in the 1st harmonic showing a parabolic relation while the 2nd harmonic showing a linear relation, as shown in Figure A1.2 and A1.3

Figure A1.2 Macro-spin simulation results for harmonic Hall voltage in the longitudinal scheme of measurements when H_z bias field, i.e. $-550 \leq H_z \leq 550$ Oe is applied in a magnetic field sweep of 0.6T.

Figure A1.3 Macro-spin simulation results for harmonic Hall voltage in the transverse scheme of measurements when H_z bias field, i.e. $-550 \leq H_z \leq 550$ Oe is applied in a magnetic field sweep of 0.6T.

When the applied magnetic field is applied at different azimuthal angles, the SOT fields dependence on the angle of the magnetisation may be determined through the ratio of the differentials of the 2nd harmonic coefficient to the 1st harmonic as shown in Figure A1.4 as shown. The β_ϕ is fitted with the expression given by (5.10) in Chapter 5 of the thesis,

$$\beta_\phi = \left(-\frac{1}{2} \sin \phi + \frac{\zeta \cos \phi \cos 2\phi}{1 - 2\zeta \sin 2\phi} \right) \Delta H_{FL} + \left(-\frac{1}{2} \cos \phi - \frac{\zeta \sin \phi \cos 2\phi}{1 - 2\zeta \sin 2\phi} \right) \Delta H_{DL}, \quad (\text{A4})$$

Figure A1.4 A sample that is up magnetised showing the schematic of the azimuthal angular dependence, the second derivative of the parabolic first-harmonic Hall voltage as a function of the azimuthal angle, ϕ . with a $-\sin 2\phi$ fit, the first derivative of the second-harmonic Hall voltage as a function of the azimuthal angle, ϕ . with a $\cos\phi$ fit, and the β_ϕ as a function of ϕ . (clockwise from top left corner)

Spin Accumulation

In the determination of spin accumulation using a mixed harmonic Hall technique as in Chapter 6 of this thesis. Macro-spin simulation is conducted similar to the above mentioned but takes into account an additional I_{dc} instead of just the alternating current, $I_{ac} \sin \omega t$. The harmonic Hall voltages are attained for both the longitudinal and transverse scheme of measurement as shown in Figure A1.5 and A1.6

Figure A1.5 Longitudinal measurement scheme harmonic Hall voltages of the up and down magnetisation states.

Figure A1.6 Transverse measurement scheme harmonic Hall voltages of the up and down magnetisation states.

Second harmonic Hall signals portray parabolic relations which suggest a contribution from the spin accumulation which can be related through a spin accumulation factor which is proportional to the applied current. As such, using a parabolic fit of the form, $f(x) \equiv C + Bx + Ax^2$, where A, B and C are coefficients of the experimental fit, which relates to the applied dc current density, since only the dc current density is being changed during the simulation. As such, by determining each coefficient's relation to the DC current density, further evaluation of the spin accumulation within the system like in Figure A1.6 can be determined.