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SINGAPORE

**INVESTIGATIONS OF MAGNETIC STRUCTURES IN ONE
DIMENSIONAL GEOMETRY FOR SPINTRONICS
APPLICATIONS**

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SCHOOL OF PHYSICAL AND MATHEMATICAL SCIENCES

2021

INVESTIGATIONS OF MAGNETIC STRUCTURES IN ONE DIMENSIONAL GEOMETRY FOR SPINTRONICS APPLICATIONS

SABPREET BHATTI

SCHOOL OF PHYSICAL AND MATHEMATICAL SCIENCES

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

2021

Statement of Originality

I hereby certify that the work embodied in this thesis is the result of original research done by me except where otherwise stated in this thesis. The thesis work has not been submitted for a degree or professional qualification to any other university or institution. I declare that this thesis is written by myself and is free of plagiarism and of sufficient grammatical clarity to be examined. I confirm that the investigations were conducted in accord with the ethics policies and integrity standards of Nanyang Technological University and that the research data are presented honestly and without prejudice.

5th July 2021

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Date

Sabpreet

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Sabpreet Bhatti

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Associate Professor S.N. Piramanayagam

Authorship Attribution Statement

This thesis contains material from four papers published in the following peer-reviewed journal(s) / from papers accepted at conferences in which I am listed as an author.

Chapter 4 is published as:

Sabpreet Bhatti, Chuang Ma, Xiaoxi Liu, and S. N. Piramanayagam. Stress-Induced Domain Wall Motion in FeCo-Based Magnetic Microwires for Realization of Energy Harvesting, *Advanced Electronics Materials* **5**, p.1800467 (2018).

DOI: 10.1002/aelm.201800467.

Sabpreet Bhatti, Chuang Ma, Xiaoxi Liu, and S. N. Piramanayagam. Realization of Energy Harvesting Based on Stress-Induced Modification of Magnetic Domain Structures in Microwires, *IEEE Transactions on Magnetics* **55**, p.2301107 (2019).

DOI: 10.1109/TMAG.2019.2902569.

The contributions of the co-authors are as follows:

- Assoc/Prof S.N. Piramanayagam (NTU, Singapore) and Prof Liu (Shinshu University, Japan) proposed the idea of this project to the Japan Society for Promotion of Science and received the grant.
- Assoc/Prof S.N. Piramanayagam and Prof Liu provided the initial project direction.
- All experiments, including sample preparation via optical lithography and material deposition, microscopy, and magnetic properties characterization were conducted by Dr Chuang Ma and I, in the Department of Electrical and Computer Engineering, Shinshu University, Nagano, Japan.
- Structure characterizations of the deposited material, through X-ray diffraction and scanning electron microscopy, were conducted by Prof Liu and Dr Ma.
- Device fabrication and mask preparation for pick-up coil deposition were carried out by Dr Ma and Prof Liu.
- Generation of energy through ambience vibration in the fabricated device was conducted by Dr Ma and I. We also analyzed the data together.
- Micromagnetic simulations on the application of stress were done by me and guided by Assoc/Prof S.N. Piramanayagam.

- I prepared the drafts of the manuscripts. The manuscripts were revised by Assoc/Prof S.N. Piramanayagam and Prof Liu.

Chapter 5 is published as:

Sabpreet Bhatt and S.N. Piramanayagam. High Amplitude Microwave Generation Using Domain Wall Motion in a Nanowire. *Physica Status (RRL)–Rapid Research Letters* **13**, p.1800479 (2019). DOI: 10.1002/pssr.201800479.

Sabpreet Bhatt and S.N. Piramanayagam. Effect of Dzyaloshinskii–Moriya Interaction Energy Confinement on Current-Driven Dynamics of Skyrmions. *Physica Status (RRL)–Rapid Research Letters* **13**, p.1900090 (2019). DOI: 10.1002/pssr.201900090.

The contributions of the co-authors are as follows:

- Assoc/Prof S.N. Piramanayagam came up with the initial idea of the works published in these papers. Together, we developed the ideas further.
- The design of experiments was planned by Assoc/Prof S.N. Piramanayagam and I together.
- I performed micromagnetic simulations to investigate the proposed techniques.
- I wrote the drafts of the manuscript, which were revised together with Assoc/Prof S.N. Piramanayagam.

Chapter 6 is based on unpublished work. The manuscript of this study is under preparation. The contributions of the expected co-authors are as follows:

- Assoc/Prof S.N. Piramanayagam and I came up with the idea of studying the variation of interfacial properties on the behaviour of skyrmions. Assoc/Prof S.N. Piramanayagam suggested the route of ion irradiation to vary the interface properties.
- Assistant Prof Anjan Soumyanarayanan (IMRE, A*STAR and National University of Singapore) supervised the tasks of material deposition and characterization techniques for skyrmions.
- Dr Sze Ter Lim (IMRE, A*STAR), Assistant Prof Anjan, and Assoc/Prof S.N. Piramanayagam oversaw the material characterization of pristine and irradiated samples.

- Prof Dafiné Ravelosona (University of Paris-Saclay/CNRS, France) administered the task of He^+ ions irradiation on the skyrmion stack. Mamour Sall (Spin Ion Technology, France) carried out the irradiation on the deposited skyrmions stack.
- I measured the MH loops of samples using vibrating sample magnetometer (VSM) and alternating gradient field magnetometer (AGFM). The measured data was analysed by me.
- The Dzyaloshinskii Moriya Interaction (DMI) measurements were carried out by Dr Vanessa Zhang Li (Wuhan University, Hubei, China) in the lab of Prof R. Mahendiran using Brillouin Light Scattering spectroscopy at Department of Physics, National University of Singapore. I assisted in the data analysis of DMI calculations.
- The imaging of skyrmions were persuaded by me using Magnetic Force Microscopy (MFM). M.I. Sim (IMRE, A*STAR) and I carried out analysis of skyrmion size and density.
- The idea to anneal samples for comparing magnetic properties and skyrmion properties with irradiated samples was proposed by me.
- Time of flight secondary ions mass spectroscopy was performed by Dr Z. Xing (IMRE, A*STAR) and the data was analyzed by me.
- Cross-section Transmission electron microscopy and energy dispersive X-ray spectroscopy was carried out at Covalent Metrology, California and I analysed the data.
- First order reverse curves were measured using AGFM and the data were analysed and plotted by me.
- H.K. Tan (IMRE, A*STAR) assisted with all magnetic characterization techniques, sample cutting, storing and preparations.

5th July 2021

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Date

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Sabpreet Bhatti

Abstract

Spintronics is a branch of magnetism, which exploits the individual spin of electrons. The devices based on spintronics are propitious for memory, computing, and sensing applications. Such devices utilize the spin degree of freedom of electrons. For spintronics devices, one-dimensional geometry has been explored extensively as it facilitates a guided motion of magnetic domain textures. This thesis emphasizes the physical and magnetic properties of some of these one-dimensional spintronics devices that can be used to harvest energy, generate microwaves, and racetrack memory applications.

The work reported in this thesis has been divided into three parts. The first part explores the energy harvesting devices for spintronics-based internet of things (IoT) sensors. In this work, an FeCo based alloy, which depicts magnetostrictive properties, has been used. The stress-induced magnetic behavior of the FeCo thin films and microwires has been studied. The application of external mechanical stress induces a change in the direction of the easy axis. The studied behavior assists to fathom the nuance of material properties, which were further used to demonstrate energy harvesting. We have fabricated a one-dimensional device with wrapped pick-up coils, which was introduced under external stress mimicking the ambient vibrations. The harvested power from the fabricated devices was in the range of μW , which can be used as a power source for spintronics IoT sensors.

The second part of this thesis explored new ideas for further exploration. The studies involved exploring the confined motion of domains walls and skyrmions for microwave generation and logic/computing applications, respectively. The demonstrations have been carried out by employing micromagnetic simulations. Microwave generation in GHz range was demonstrated using back and forth motion of domain walls in a nanotrack with synthetic pinning sites. The latter portion explores the skyrmion motion confined in a Dzyaloshinskii-Moriya interaction (DMI) energy well. We have demonstrated that the skyrmion Hall effect can be suppressed

after introducing energy barriers, which proved to be beneficial for high-density skyrmion devices.

The last part of the thesis explores how to achieve a high-density skyrmion state in thin films and demonstrates the DMI modification for confined motion of skyrmions experimentally. Intermixing of atoms at interface of Pt/CoFeB/MgO stack was utilized to vary the interfacial properties, viz. K_{eff} and DMI, via He^+ ion irradiation. The effect of irradiation dose was studied on the magnetic and skyrmion physical properties. The undertaken approach effectively enhanced the skyrmion density by five folds (reaching up to $50 \text{ skyrmion}/\mu\text{m}^2$). Moreover, the skyrmion size was reduced by $\sim 70 \text{ nm}$. The variation in the physical property of skyrmions can be construed due to a reduction in the domain wall energy. This technique can be used for generating high-density skyrmions at nucleation sites of racetrack memories. This technique also can be used to achieve the tracks with low DMI for confined motion of skyrmions.

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First and foremost, I would like to express my sincere and deep gratitude to my research supervisor, Associate Professor S.N. Piramanayagam, for believing in me and providing me the opportunities to fulfill my yearning to pursue research. His optimism, vision, sincerity, humbleness, motivation, patience, and perseverance have deeply inspired me. During my research under him, he has continuously polished my skills, rectified my mistakes, supported me, and stood by my side (whenever needed). I am extremely grateful for what he has offered me. I was fortunate that a diligent and sensible person like him happened to be my supervisor. It is beyond my words to express my heartfelt appreciation for him.

I would also like to extend my gratitude towards my collaborators from Shinshu University Nagano, Japan, especially Prof Xiaoxi Liu. It was a great privilege to work under Prof Liu and learn multiple sample preparations and microscopy skills from him. I am also thankful to Dr Chuang Ma for his assistance and valuable time during experiments and data analysis. I am grateful to Prof Liu, Prof. Morisako, Dr Ma, Mr Arai, and Mr Hara for their warm hospitality during my visits to Japan. Additionally, I am thankful to the Japan Society for Promotion of Sciences (JSPS) for granting the funds, which supported the study on energy-harvesting devices.

Additionally, I am profoundly grateful to my collaborators from Institute of Material Research and Engineering, Agency for Science, Technology and Research (A*STAR) Singapore. I would like to express my gratitude to Dr Sze Ter Lim for supervising me and providing the opportunity to work in A*STAR. I am grateful to him for all his valuable time and for being there for me whenever I was in need. It was he, who mitigated my work-related issues and differences and supported me throughout my journey in PhD. His joviality during my visits to his office has always uplifted my spirit. Further, I would like to extend my gratitude to Assistant Professor Anjan Soumyanarayanan, for overseeing my skyrmion-related work, providing his

insights and support. I am also grateful to Dr Xiaoye Chen (whom I often called ‘*Senpai*’) for introducing me to Python language and numerous discussions, which have improved my understanding of skyrmion fundamentals. I would also like to thank Dr Ho Pin for taking the time to go through my work updates and giving comments that directed my experimental investigations. Moreover, a sincere thanks go to Mr H.K. Tan, who has assisted my magnetic characterizations and sample preparation. Further, I would like to thank Ms May Inn Sim for her assistance in imaging and analysing the skyrmions. Also, I am grateful to Prof R. Mahendiran for allowing the measurement of DMI for skyrmion samples and Dr Vanessa Zhang Li for measuring the property. My sincere gratitude also goes to Dr Z. Xing for carrying out the ToF-SIMS. I am also thankful to Dr James Lourembam, Dr Abhijit Ghosh, Dr Dickson Thian, and Dr Jian Feng, for their support and discussions on various occasions. My special thanks are due to Dr Sarjoosing Goolaup for introducing me to mumax3. Additionally, I would like to give credit to SpOT-LITE research grant to facilitate the funds that supported the investigations on the skyrmion project.

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List of Publications

Journal Publications

1. **Sabpreet Bhatti** and SN Piramanayagam, “ Effect of Dzyaloshinskii - Moriya Interaction Energy Confinement on Current driven Dynamics of Skyrmion” *Physica Status Solidi (RRL) – Rapid Research Letters*, (2019), 13(11), p.1900090, DOI: 10.1002/pssr.201900090.
2. **Sabpreet Bhatti**, Chuang Ma, Xioaxi Liu and S.N. Piramanayagam, "Realization of Energy Harvesting Based on Stress-Induced Modification of Magnetic Domain Structures in Microwires." *IEEE Transactions on Magnetics*, (2019), 55(7), p.2301107, DOI: 10.1109/TMAG.2019.2902569.
3. **Sabpreet Bhatti** and S.N. Piramanayagam, “High Amplitude Microwave Generation using Domain Wall Motion in a Nanowire”, *Physica Status Solidi (RRL)–Rapid Research Letters*, (2019), 13(3), p.1800479, DOI: 10.1002/pssr.201800479.
4. **Sabpreet Bhatti**, Chuang Ma, Xioaxi Liu and S.N. Piramanayagam, “Stress induced Domain Wall motion in FeCo microwires for realization of Energy Harvesting”. *Advanced Electronic Materials*, (2019), 5(1), p.1800467, DOI: 10.1002/aelm.201800467.
5. **Sabpreet Bhatti**, Rachid Sbiba, Atsufumi Hirohata, Hideo Ohno and Shunsuke Fukami S.N. Piramanayagam “Spintronics based Random Access Memory: a Review.” *Materials Today*, (2017), 20(9): p. 530, DOI: 10.1016/j.mattod.2017.07.007.

Conference Presentations

6. “Enhancement of Skyrmion Density achieved via Interface Engineering” International Magnetic Conference (INTERMAG) 2021, April 26th to 30th 2021, Lyon France. (***Best Student Oral Presentation Finalist***)
7. “Transformation of skyrmion phase induced by ion irradiation” 9th MRS-Singapore Conference on Advanced Materials, November 23rd to 25th 2020, Singapore. (***Best Poster Award: Bronze***)
8. “Effect of Dzyaloshinskii-Moriya Interaction Energy Confinement on Current driven Dynamics of Skyrmion” IEEE Magnetics Summer School 2019, June 5th to June 8th 2019, Richmond, Virginia (USA).
9. “Stress induced domain wall motion in FeCo based magnetic microwires for realization of energy harvesting” IEEE International Magnetics Conference (INTERMAG) and the Conference on Magnetism and Magnetic Materials (MMM), January 14th to 19th 2019, Washington D.C. USA. (***Invited Talk by Prof. Piramanayagam***)
10. “Analysis of stress induced domain wall structure modification in FeCo based multi-layered microwires” 8th MRS-Singapore Conference on Advanced Materials, November 21st to 23rd 2018, Singapore. (***Best Poster Award***)
11. “Energy Harvesting Based on Stress Induced Domain Wall Motion in Soft Magnetic Microwires” The 42nd Annual conference on Magnetics in Japan, September 11th to 14th 2018, Surugadai, Tokyo, Japan (***Invited Talk by Prof. Piramanayagam***).
12. “Reducing the Magnetostatic Interactions for High Density Magnetic Random-Access Memory” MAINZ Summer School, August 11th to 17th 2018, Beijing, China.

13. "Harvesting Energy using stress induced domain wall motion in FeCo microwires"
International Colloquium on Magnetic Films and Surfaces (ICMFS), July 23rd to 27th
2018, Santa Cruz, California, USA (***Best Student Presentation Finalist***).
14. "Effect of FeCo microwires thickness on the Stress Regulated Domain Wall Motion"
International Magnetic Conference (INTERMAG) 2018, April 23rd to 27th 2018,
Singapore (***Best Poster Award***).
15. "Stress Induced Domain Wall Motion and its application in Energy Harvesting"
Institute of Physics Singapore (IPS) Meeting, March 7th to 9th 2018, Singapore (***Invited
Talk by Prof. Piramanayagam***).
16. "Stress Regulated Domain wall motion in CoFe" Conference on Magnetism and
Magnetic Materials (MMM), November 6th to 10th 2017, Pittsburgh, Pennsylvania,
USA.

List of Abbreviation

IBM	International Business Machines
HDD	Hard Disk Drives
GMR	Giant-Magnetoresistance
MRAM	Magnetic Random-Access Memory
RAM	Random-Access Memory
XRD	X-Rays Diffraction
IoT	Internet of Things
DMI	Dzyaloshinskii Moriya Interaction
MOKE	Magneto-Optic Kerr Effect
CMOS	Complementary Metal Oxide Semiconductor
MFM	Magnetic Force Microscopy
PZT	Lead Zirconate Titanate
SOC	Spin Orbit Coupling
PMA	Perpendicular Magnetic Anisotropy
FM	Ferromagnet
HM	Heavy Metal
MO _x	Metal Oxide
SOT	Spin Orbit Coupling
SHE	Spin Hall Effect
SHA	Spin Hall Angle
TMR	Tunnel Magnetoresistance
VSM	Vibrating-Sample Magnetometer
AGM	Alternative Gradient Field Magnetometry
RF	Radio-Frequency
FTS	Facing Targeting Sputtering
MH	Magnetic Hysteresis
BPI	Bitter Pattern Imaging
SEM	Scanning Electron Microscope
ToF-SIMS	Time of Flight Secondary ions mass spectroscopy

List of Symbols

K_{eff}	Effective Anisotropy Energy Density
A_{ex}	Exchange Coupling Energy Density
M_s	Saturation Magnetisation
M	Magnetisation
H	Magnetic Field
χ	Magnetic Susceptibility
B	Applied Magnetic Field
M_r	Remanent Magnetisation
E_d	Dipolar Energy
H_d	Dipolar Field
S_i	Spin vector
$\hat{J}$	Heisenberg Exchange Interaction Constant
E_{ex}	Exchange Energy
A	Exchange Stiffness Constant
H_{ext}	External Magnetic Field
ϵ_{Zeeman}	Zeeman Energy
$\epsilon_{anisotropy}$	Anisotropy Energy
α, β, γ	Cosine angles between direction of magnetisation and the crystal axis
K_c	Anisotropy constant
K_s	Surface Anisotropy Energy
K_v	Volume Anisotropy Energy
K_b	Bulk Magnetocrystalline anisotropy Energy
K_d	Shape Anisotropy
t	Thickness of the Ferromagnetic Film
t_c	Critical Thickness
B_{MI}	Magnetic Induction
s_e	Elastic Compliance
d'	Magnetoelastic Constant
μ_0	Magnetic Permeability
d^m	Piezomagnetic Constant

$m_{elastic}$	Magnetoelastic Constant
$\vec{v}$	Velocity of Electron
$\vec{E}$	Electric Field
$\vec{B}_{eff}$	Effective Magnetic Field
μ_B	Bohr's Magnetron
c	Speed of light
H_{SO}	Spin Orbit Field
H_{DMI}	Hamiltonian of Dzyaloshinskii Moriya Interaction
$\vec{d}_{ij}$	DMI vector
$\vec{r}_{ij}$	Distance between connecting two spins
$\vec{z}$	Vector representing direction normal to the film
E_{DMI}	Dzyaloshinskii Moriya Interaction Energy
D	Effective Dzyaloshinskii Moriya Interaction
m_i	Normalized Magnetisation
a	Lattice Constant
$E\hat{z}$	Interfacial Electric Field
H_{RSOC}	Magnetic Field due to Rashba Spin Orbit Coupling
α_R	Rashba Parameter
$\vec{k}$	Electron Momentum
$\vec{B}_R$	Effective Magnetic Field produced by Rashba effect
D_{Rashba}	DMI due to Rashba
K_R	Rashba Coefficient
m_e	Mass of electron
$\hbar$	Plank's constant
j_s	Spin Current
j_c	Applied Current
θ_{SH}	Spin Hall Angle
σ	Polarization of Spin Current
e	Elementary electron Charge
σ_{DWE}	Domain Wall Energy
S	Topological Number

κ	Thermal Stability Constant for Skyrmions
K_u	Uniform Anisotropy Energy
T	Temperature in Kelvin
V	Volume of the magnetic film
H_c	Coercivity Field
H_s	Saturation Field
F_z	Magnetic Force in z direction
F_{ext}	Sinusoidal Driving Force
ω	angular frequency
k_{spring}	Spring Constant
D_{grain}	Grain Size
K_{stress}	Stress Anisotropy
σ_{stress}	Applied Stress
$\lambda_{mstriction}$	Magnetostriction Constant
E_s	Young's Modulus

Chapter 1 Introduction

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1.1 Introduction to the thesis

Starting from the magnetic compass, magnetic materials-based devices have always been useful in our daily lives. In today's world, a myriad of devices based on magnetic materials can be found. Out of the myriad applications of magnetic materials, some are enormous, like the turbines of power plants, while some are tiny (nanometer scale), like magnetic bits of hard disk drives restricting to one or zero dimensions. Irrespective of the size of magnetic material-based devices, one cannot imagine their life without using any magnetic devices.

Magnetism has revolutionized the field of data storage. The application of magnetism as data storage can be traced back to the late 19th century when Oberlin Smith published a description on magnetic sound recording in 1888 [1]. Later in 1898, Valdemar Poulsen first demonstrated a magnetic medium for recording information [2]. In the 1940s, magnetic coated tapes with Fe₂O₃ particles were invented. In 1953, International Business Machines (IBM) Corporation shipped the first commercial magnetic tape drive. Later in 1956, IBM shipped the first commercial hard disk drive (HDD), which had data storage space of 5MB. Interestingly, the size of magnetic disks was 24 inches in diameter with a data storage density of merely 2 kbits/in² [3]. The improvement in data storage capacity continued in the 20th century due to advancements in material deposition techniques and lithography, which led to improved recording media and reading heads. These techniques made the thickness reach a few nanometres reliably, which assuage the exploitation of electron spins. The discovery of Giant-Magnetoresistance (GMR) effect by Albert Fert [4] and Peter Grunberg [5] in 1980 boosted the interests of researchers toward this field. The impact of this discovery boomed a new era which came with the advent of a new field in science named 'Spintronics'.

Spintronics is a field of science which deals with electronic spins and spin current. Dealing with spin facilitates another degree of freedom in addition to charge, making it a promising candidate for storage, computing, and other sensing applications.

Besides HDD, spintronics has found a niche application in the random-access memory (RAM), such as magnetic random-access memory (MRAM). Unlike the conventional semiconductor-based RAM, MRAM is a non-volatile memory, which means the data remains stored even when the power is switched off [6]. Further, there are other memory devices such as racetrack memory, which are based on their one-dimensional geometry. In these memory technologies, magnetic textures like domain walls and skyrmions move from one end of the geometry to another. The idea was first proposed for domain-wall racetrack memory by Stuart Parkin in 2007 [7]. The proposed idea worked like a shift register where a spin current moves magnetic domains (data bits) in the racetrack. Later, with the advent of thin-film skyrmions in 2013, the idea of racetrack memory was also extended to skyrmions-based memory [8]. Since then, much work has been carried out on the skyrmion-based one-dimensional devices for memory and computing devices [9]. Besides memory and computing applications, spintronics-based one-dimensional devices have been proposed for other applications like energy harvesters, spin-torque oscillators, microwave generators, and spin-wave generators. All these investigations on the devices aim to be the product of consumer electronics. In consumer electronics, it is better to have devices with small sizes. Having devices with small sizes increase the density of devices in a certain volume, and hence, enhances the output of devices. Keeping this as an aim, researchers are working on low-dimensional devices. In spintronics also, low-dimensional devices hold the potential to revolutionize information storage, computing, and communication. This thesis focuses on the physics of magnetic structures in one-dimensional geometry, which demonstrate promising applications for harvesting energy, microwave generation and skyrmions based devices.

1.2 Scope of the thesis

This thesis reports studies on the physical phenomena of magnetic structures in one-dimensional geometry (length is a few orders larger than the width) and exploits them for spintronics applications. The work reported in the thesis can apply to spintronics sensors, racetrack memory, microwave generators, and computing devices. The magnetic structures studied in the thesis varies in the range from sub-10 nm to μm . Hence, the studied magnetic structures in one-dimensional geometry should not be misconstrued with one-dimensional magnetic structures (the magnetic structure size is less than the magnetic exchange length).

The thesis has been divided into seven chapters. Chapter 2 provides the background framework of concepts. The beginning of the chapter explains the basic types of magnetic materials and their behaviours with respect to the applied magnetic field. Then, the focus of the chapter shifts to various magnetic energies. The fundamental information (provided in chapter 2) on different energies of magnetic materials helps to fathom the behaviour of magnetic textures and the current-induced dynamics of magnetisation studied in this thesis.

The next section of chapter 2 focuses on the magnetostrictive anisotropy of magnetic materials and their applications in harvesting energy for the internet of things (IoT). In the latter half of the chapter, the focus shifts to spin orbit coupling-related phenomena. Subsequently, different origins of Dzyaloshinskii Moriya interactions have been explained. Dzyaloshinskii Moriya interaction or DMI is an antisymmetric exchange interaction that has gained focus in the past few years due to its role in chiral textures. The following section explains the spin orbit torque and its origin, which can be traced to the spin Hall effect or Rashba Edelstein effect. Then the focus of the chapter shifts to domain walls and chiral domains walls. In the domain wall section, the behaviour of different domain walls in ultra-thin film, and their structures were defined schematically. In the second last section, a detailed description of skyrmions was given. The section explains different aspects of skyrmions, nucleation, and stability in thin films. The last

section of the chapter gives an insight into the modulation of magnetic properties achieved via irradiation. The last section explains the changes in the structural properties at the interface of a material, which lead to variation in the magnetic properties arising at the interface of materials. These variations subsequently change the density, size, chirality, and other physical properties of these magnetic textures.

Chapter 3 gives the details of the used methods and characterization techniques. The chapter commences with a short discussion on different techniques of thin-film deposition and an explanation of the two sputtering deposition methods used to deposit thin films in the investigations of this thesis. Then, the focus of the chapter shifts to the optical lithography technique used for the fabrication of microwires. The next section of the chapter explains the magnetometry techniques used for measuring the magnetic properties of samples. These techniques include vibrating sample magnetometry, Magneto-Optic Kerr effect (MOKE) magnetometry, and alternative gradient field magnetometry. Moreover, the method of calculating the effective anisotropy energy density was defined. Further, an explanation and underlying physics of used imaging techniques were done. These techniques include Bitter patterns and magnetic force microscopy. In addition, the micromagnetic simulations concepts were explained. Mumax3 was used to carry out the simulations, proven an essential tool for understanding magnetisation behaviour, current-induced dynamics of domain walls, skyrmions, and estimation of the exchange coupling energy density. In the final section of the chapter, different methods for applying stress on thin films and microwire has been discussed.

In chapter 4, I have shown the results of various experiments and simulations to explore the potential of a magnetostrictive material based energy harvesting device to power the internet of things (IoT). Internet of Things (IoT) devices, which work in interconnected nexus to share

data, have been booming recently. IoT is a compelling paradigm for smart living. The sensors of IoT are interconnected and collect the information of interest continuously. According to the application note released by Libelium [10], magnetic sensors occupy around 10% of the proportion in total for all smart sensors applications. Out of the magnetic sensors, the spintronics sensors, namely magnetoresistance sensors, are widely used in IoTs.

In the chapter 4, first, I have shown the results to describe the use of magnetostrictive FeCo microwire to generate energy to power the IoT devices. IoT devices hold the potential to scrutinize all the information related to public safety, human healthcare, industrial automation, and environmental changes. For IoT devices, these devices should harvest their energy. I have shown that stress-induced magnetic domain wall motion can generate power to harvest energy. The stress can be induced from ambient vibration. By applying stress, we (I and other contributors of the study) have demonstrated that the hard axis of the microwire can be changed to the easy axis. As a result, domain wall motion or collapse of domain walls occurs entirely by stress-induced bending of substrate. Such changes in the domain walls position introduce variation in the magnetic flux, which can harvest energy. We have demonstrated energy generation in the order of μJ in a time span of $50\ \mu\text{s}$ using a tri-layer magnetic stack [11].

Figure 1.1. Generation of energy induced by ambience stress. The FeCo thin films show a change in magnetisation direction under external stress. The magnetic domains direction was changed in fabricated FeCo microwires. On applying stress through a speaker on fabricated devices, we were able to harvest energy. In practical devices, stress may arise from ambient vibrations.

To further explore the potential of one-dimensional magnetic devices in spintronics, I have demonstrated in chapter 5 the use of energy confinement on the motion of domain walls and skyrmions. Chapter 5 demonstrates the computationally investigated two concepts and shortlist an application for further experimental investigations. The first half of the chapter demonstrates the generation of high amplitude microwave signals in a nanowire. Microwave generation has been extensively investigated due to its application in telecommunication and promising use in emerging technologies like IoT devices. The current approaches to generating microwaves using voltage-controlled oscillators have the problem of high-power consumption. Spin Torque Oscillators have been investigated as an alternative, but they suffer from a weak output signal [12]. Hence, there is a need for an alternative approach that overcomes these issues. We have carried out investigations on a new approach, which relies on the motion of domain walls to

generate microwaves with a strong signal and lower power consumption. For this purpose, we have simulated a magnetic nanowire with a sequence of oppositely magnetized domains in one-half of the nanowire. We moved the domain walls by applying spin-orbit torque to the nanowire. We propose to generate a microwave by detecting the changes in magnetisation value using a magnetic tunnel junction in an actual application. The oscillating frequency of magnetisation was found to be in GHz [13]. We have demonstrated that the frequency can be tuned by varying the applied current density. The outcome of this work promises the application of the reported method to generate microwaves in one-dimensional devices of the future.

In the second half of the chapter, I have computationally demonstrated a magnetic nanotrack with the controlled motion of skyrmions for racetrack memories, logic devices, or bio-inspired applications. Magnetic skyrmions are nanoscale topological spin textures, usually found in magnetic materials with asymmetric or frustrated exchange interactions. The possibility of skyrmions to circumvent tiny defects (inhomogeneities in material), and required low current for motion have made the skyrmion a propitious candidate for various applications like racetrack memories, logic computing devices, bioinspired applications, etc. However, skyrmions in a racetrack move with a transverse drift towards the edge of the track (skyrmion Hall effect) and eventually annihilates [14]. Several methods have been proposed to avoid this detrimental effect, including synthetic antiferromagnetic layer structures, ferromagnetic skyrmionium, potential barriers, etc. We have investigated a new scheme based on DMI energy wells by locally modifying the DMI. DMI of the track edges can be tuned locally by practical approaches such as ion-implantation or compositional modifications that can be easily adapted in a complementary metal-oxide-semiconductor (CMOS) -friendly manner. Using our proposed approach, we have demonstrated the restriction of the skyrmion's transverse motion to avoid the annihilation of the skyrmion [15]. In addition, we have carried out a comprehensive study to investigate the dynamics of skyrmion for various DMI energies of the wells. We have

demonstrated that the size of the skyrmions can be reduced, to some extent, by DMI energy wells' positions. Moreover, the velocity and path of skyrmion can be manoeuvred by varying the current and energy of DMI energy wells. In addition, we have investigated the effect of magnetostatic interaction between the skyrmions in closely spaced neighbouring tracks for high-density storage application. The outcome of our reported studies corroborates the use of skyrmion for future applications.

Figure 1.2. Controlled motion of magnetic domains/skyrmions through energy barrier for various applications. The introduction of an energy barrier was achieved by lowering DMI energy. The controlled motion domains (skyrmions) was used for generating microwaves (suppressing the skyrmion Hall effect).

In chapter 6, I investigated the methods to tweak skyrmion properties via varying the interfacial properties. Skyrmions can exist in a low-density state or a high-density state [16, 17]. A high-density skyrmion state is more favourable for high-density memory devices. However, the formation of the high-density state requires a subtle balance of magnetic properties. One of the critical requirements needed for the high-density state is a relatively high value of DMI as compared to the effective magnetic anisotropy (K_{eff}) and exchange coupling energy (A_{ex}) [18].

In a specific thin material system, DMI can be tuned by either changing the thickness of ferromagnet (FM) or by varying its stoichiometry[19-21]. However, these methods fell short in reducing K_{eff} and providing an unhindered skyrmion motion, respectively.

In this thesis, I report a reduction of K_{eff} by using He^+ ions induced interface intermixing without compromising DMI. I have used Pt/CoFeB/MgO stack to study the effect of varying ion irradiation dose and carried out Monte Carlo simulations to get the probability of the ions distributions and the intermixing in the sample [22], which has shown a linear increase in displacements of atoms with increasing dose. I have found a reduction in K_{eff} as atomic intermixing at interfaces between the FM and non-magnetic layers was increased. Further, an increase in the saturation magnetisation (M_s) was observed, due to increase of the spin moment at interface and migration of B atoms [23]. Interestingly, the interfacial DMI was increased by 50% after irradiation. Surface M_s and interfacial DMI were calculated to eliminate the effective thickness. Moreover, micromagnetic simulations were used to estimate the exchange coupling energy A_{ex} . Noticeably, both DMI and A_{ex} follow a similar trend as M_s .

To probe the change in magnetisation dynamics due to the modification of the magnetic properties, I have carried out magnetic force microscopy (MFM). A reduction in the zero-field textures by ~ 50 nm was noticed. The skyrmion density increased up to five folds as compared to the pristine sample. Furthermore, the skyrmion size was reduced by ~ 70 nm, down to 75 nm. Besides, to comprehend the underlying physics, a comparatively low energy treatment (annealing) of the deposited stack was done to explain the physical phenomena of skyrmion density enhancement.

Figure 1.3. Enhancement of skyrmion density. The irradiation of the sample introduced intermixing of atoms at the interface, which varied the interface properties. Subsequently, a high skyrmion density was achieved after irradiation.

In the last chapter, I have summarized the key results of the studies reported in this thesis. Then, I have given the outlook on the achieved results and their potential for applications. In the last part of the chapter, I have listed investigations related to the reported studies, which are promising and can be carried out in the future.

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2.1 Introduction

This chapter aims to introduce some basic information required to understand the relevant topics investigated in this thesis. The chapter introduces some basics of magnetism at first. Different energies of a magnetic system are discussed. Then the focus of the chapter shifts to the internet of things and energy harvesting techniques. The later part of the section describes the different phenomena originating from spin orbit interactions. In the end, the chapter provides a description of magnetic skyrmions and their structural behaviours.

2.2 Types of magnetic materials

Magnetism has always played an essential role in day-to-day life. The applications of magnetism-based devices are myriad. Physically, the origin of magnetism can be understood due to the spin of electron and electron orbital motion around a nucleus. Based on the occupation of electronic orbits, all materials can be categorized into one of the magnetic material types. A material with completely filled electronic orbits, where all the electrons are paired, has zero magnetic moment. Such materials are called diamagnetic. In contrast, materials with outermost electronic orbit with unpaired electrons can be classified into paramagnetic and ferromagnetic. The paramagnetic materials have zero net magnetic moment at room temperature due to random directions of the moment. Whereas, in the case of ferromagnetic materials, the exchange interaction between neighbouring moments results in a net magnetic moment. The magnetisation in these materials is calculated by dividing the total magnetic moment by the volume of the magnetic material. The magnetisation of the material is typically denoted by M . Further, magnetic orders are categorized as ferrimagnetic and antiferromagnetic based on the magnetic moment direction and amplitude. A ferrimagnetic material has oppositely oriented moments in two sublattices. As the magnitude of the moments is different in value, the magnetisation is not fully compensated. Mostly, a ferrimagnetic material behaves

similar to ferromagnetic materials. Whereas an antiferromagnetic material has oppositely oriented magnetic moments of identical magnitudes in the two sublattices, resulting in a zero net magnetic moment.

Figure 2.1. Schematic representation of different magnetic ordering in different magnetic materials.

Magnetic susceptibility is another benchmark to differentiate different types of magnetic materials. The magnetic susceptibility infers the sensitivity of the material to an applied magnetic field H . The magnetic susceptibility can be written $\chi = M/H$, where M is the magnetisation of material and H is the applied magnetic field on the material. The $M - H$ behaviour of different magnetic materials is shown in figure 2.2.

Figure 2.2. Magnetisation (M) of different magnetic materials as a function of the applied magnetic field (H); M_s : saturation magnetisation; M_r : remanent magnetisation. (The y-axis values are not-to-scale)

2.3 Types of different magnetic energies

The total energy of a magnetic system at any point in time depends on various interactions present in the magnetic material and the external magnetic field (if any). It is crucial to fathom these interactions, which play an essential role in the dynamics of a magnet. In this section, different energies have been explained based on both micromagnetic modelling and quantum mechanical approaches.

2.3.1 Dipolar energy

Dipolar interaction, which is resultant energy from magnetic moments interaction, is a long-range type of interaction. The arrangements of magnetic moments in a system result in magnetic charges at the surface. These arrangements lead to the generation of stray fields.

Therefore, magnetic moments lean to align in such a direction that the surface charges constitute minimum energy.

Figure 2.3. Magnetic field due to a hypothetical dipole.

The dipolar energy (E_d) of a system can be written in the following form:

$$E_d = -\frac{\mu_0}{2} \int_V M \cdot H_d dV$$

where V is the volume of the ferromagnet. $H_d = -nN \cdot M$, and N is a shape-dependent demagnetizing tensor. The shape of the system substantially controls the dipolar energy. In the case of thin films with uniform magnetisation, the shape-dependent tensor N is maximum for perpendicular direction and zero for parallel (in-plane) direction. The dipolar energy can be written as:

$$E_d = -\frac{\mu_0}{2} M_s^2 \sin^2 \theta$$

2.3.2 Exchange energy

The phenomenon of exchange coupling can be construed as the reason behind the lineup of individual atomic magnetic moments with the neighbouring magnetic moment of atoms. Exchange coupling energy is a consequence of quantum mechanical interaction. The exchange

interaction energy between two neighbouring spins S_1 and S_2 can be written with the help of Heisenberg exchange interaction constant ($\hat{j}_{12}$) as

$$\varepsilon_{12} = -\hat{j}_{12}\vec{S}_1 \cdot \vec{S}_2$$

Figure 2.4. The arrangement of spins under the different signs of exchange values (a) and (b). (a) A positive value of exchange coupling $\hat{j}_{12}$ gives rise to a ferromagnetic coupling and (b) negative value $\hat{j}_{12}$ gives rise to an antiferromagnetic coupling. (c) Effect of the external field H_{ext} on the magnetic spins of a system, resulting in an aligned direction parallel to the applied field.

The Heisenberg exchange constant favours a collinear arrangement of spins, implying spins align parallel or anti-parallel to each other. For $\hat{j} > 0$, the spins are oriented parallel to each other. Hence, the ferromagnetic order is favourable. However, for $\hat{j} < 0$, the spins are oriented anti-parallel to each other, which engenders antiferromagnetic ($\vec{S}_1 = \vec{S}_2$) or ferrimagnetic ordering ($\vec{S}_1 \neq \vec{S}_2$). In a chain of one-dimensional spin, the energy becomes

$$\varepsilon_{ij} = -\hat{j}S^2 \sum_{ij} \cos\phi_{i,j}$$

$$\text{where } \cos\phi_{i,j} \sim |m_i - m_j|, m = M/M_s$$

$$\varepsilon_{ij} = -\hat{J}S^2(1 - \frac{\phi_{ij}}{2}), \text{ for small } \phi_{ij}$$

The discrete nature of the lattice can be ignored, and total exchange energy can be written as

$$E_{ex} = A_{ex} \int_V [\nabla \vec{m}]^2 dV, \text{ where } A_{ex} \approx \hat{J}S^2 z / 2a$$

The A_{ex} is called the exchange stiffness constant (a - nearest neighbour distance, z - number of sites in a unit cell)

2.3.3 Zeeman energy

Zeeman energy is the energy arising from the interaction of magnetisation with an external magnetic field. The energy tends the magnetisation of the system to align along the direction of the applied external magnetic field. The Zeeman energy of a system with magnetisation M in the presence of an external magnetic field H_{ext} can be written as

$$\varepsilon_{Zeeman} = -\mu_0 \int_V M \cdot H_{ext} dV$$

2.3.4 Magnetocrystalline anisotropy energy

Magnetocrystalline energy is energy arising from the interaction of crystal field and magnetisation, causing the magnetisation to align in a specific crystallographic direction or two. The energy of the system is the lowest when the direction of magnetisation is parallel to a certain crystallographic direction (called the easy axis). However, the direction (plane) which are perpendicular to the easy direction is called the hard axis (plane). The energy required to align magnetisation in the direction of the hard axis is maximum. The anisotropy energy density can be written in terms of various angular functions associated with the symmetry of crystal structure. For materials with a cubic symmetry lattice structure, the anisotropy energy ε can be written as:

$$\varepsilon_{anisotropy} = K_{c0} + K_{c1}(\alpha^2\beta^2 + \beta^2\gamma^2 + \gamma^2\alpha^2) + K_{c2}(\alpha^2\beta^2\gamma^2)$$

where α, β, γ are the cosine of the angle between the direction of magnetisation and the crystal axis, and K_{c0}, K_{c1} , and K_{c2} are the anisotropy constants.

For a hexagonal crystal system such as Co, the c axis is an easy axis with sixfold rotation symmetry. The energy density can be written as

$$\varepsilon_{anisotropy} = K_{u1}\sin^2\theta + K_{u2}\sin^4\theta + \dots$$

Here, θ is the angle between the easy axis and the direction of magnetisation and K_{u1} , and K_{u2} are the anisotropy constants. In most systems, the higher-order terms can be neglected, owing to a small contribution of K_{u2} . Hence, the above expression can be written in simple terms with a uniaxial anisotropy constant (K_u) as:

$$\varepsilon_{anisotropy} = K_u\sin^2\theta$$

For tetragonal crystal lattice systems and rhombohedral crystal lattice systems, the anisotropy energy density can be expressed as:

$$\varepsilon_{anisotropy} = K_{c1}\sin^2\theta + K_{c2}\sin^4\theta + \dots$$

The higher terms can also be neglected in these lattice systems.

2.3.5 Surface anisotropy

Magnetic thin films with a high surface to volume (thickness of film less $\leq 1\text{nm}$) generate a substantial anisotropy in a perpendicular direction. This anisotropy is called surface anisotropy (K_s). The surface anisotropy arises in material systems with broken inversion symmetry. This phenomenon, which generally occurs at the interface, is contributed by the interfacial electronics or magnetoelastic effect.

The origin of surface anisotropy at the interface is the interaction between the electron orbitals of the ferromagnetic layer and the neighbouring layer. The studied material in this thesis consists of Pt/CoFeB/MgO stack. This stack has two interfaces. Interestingly, the interface anisotropy can come from both interfaces. The first source of anisotropy is due to the strong

spin orbit interaction between Co and Pt (heavy metal). The first-principle studies suggest significant interfacial anisotropy comes from Co/X interfaces combination (where X is Cu, Pd, Au, Pt, Ag) [1, 2]. The interfacial anisotropy in these interfaces can be construed due to the hybridization between the 3d orbitals of Co and 5d orbitals of the heavy metal. This strong hybridization results in energy splitting between $3d_{x^2-y^2}$ and $3d_z^2$ orbitals of the Co layer. Resultingly, the orbitals moments which are quenched become enhanced in the out of plane direction [3].

On the other hand, the second interface (CoFeB/MgO) can also lead to interface anisotropy. The study has shown that the interface between Co or Fe/M-O (with metals M =Ta, Mg, Al, Ru), causing a high degree of oxidation of metal M can strongly affect the magnetic anisotropy of the transition metal layer (Co in this case). The origin of this effect can be fathomed due to overlapping between the Oxygen- p_z orbitals and the hybridized $3d_z^2$ and $3d_{xz(yz)}$ orbitals of Co or Fe [4, 5]. The present spin orbit interaction, though weak, is still sufficient to raise the degeneracy of the out-of-plane 3d orbitals. The subsequent effect of oxidation is very susceptible to the disorder. Noticeably, the resulting interfacial anisotropy in the out of plane direction is stronger for Fe/MgO interfaces than the Co/Pt interface.

The effective anisotropy energy K_{eff} , measured from the experiments, can be written as

$$K_{eff} = (K_s + K_v)/t$$

where K_s is the surface anisotropy and $K_v = K_b + K_d$; K_b is the bulk magnetocrystalline anisotropy and K_d is the shape anisotropy (negative in sign), and t is the thickness of the ferromagnetic film. The contribution of K_s to the total magnetic anisotropy energy reduces with an increase in thickness of the ferromagnetic layer. Above a critical thickness (t_c), the effective anisotropy of the ferromagnetic layer will be negative, and the magnetisation will lie in the in-plane direction, which is parallel to the plane of the layer. On the other hand, if the thickness of a ferromagnetic layer is less than t_c then the K_{eff} is positive and an out of plane magnetic

anisotropy, which aligns the magnetisation perpendicular to the surface of the layer, is favoured.

For samples studied in chapter 6, the t_c is around ~ 0.6 nm.

Figure 2.5. Trend of effective magnetic anisotropy K_{eff} multiplied by the thickness (t) of the ferromagnetic material, plotted as a function of the ferromagnetic layer thickness. $K_{eff} > 0$ indicates anisotropy is out of plane magnetisation direction, whereas when $K_{eff} < 0$, the magnetisation is in-plane direction.

2.4 Magnetostrictive materials for harvesting energy

Magnetostriction is a phenomenon of some ferromagnetic materials in which magnetic domains rotate under applied external magnetic field. The magnetic domains can expand and contract according to the direction of the applied field.

The first description of the magnetostrictive effect can be traced back to 1842, as reported by J.P. Joule [6]. In his report, the length of an iron piece was changed when placed under a magnetic field.

The change in the dimension of the material can be understood as follow:

In crystalline solids, the atoms are arranged in regular patterns, and the distance between one atom or ion and its neighbours is fixed within certain limits. General attractive forces cause the

atoms to approach each other and be as close as possible. However, the limiting factor opposing this movement is the orbitals of the outer electrons of each atom or ion. The orbitals cannot cross over each other.

In an external magnetic field, some electron orbitals cannot align in the direction of the external magnetic field without a change taking place in the distances between the atoms or ions. These distances must increase to allow the orbitals to become aligned. These changes in length may cause the crystal structure to become elongated in the same direction as the magnetic field or to become elongated in one or more directions at right angles to this direction. In this last case, there may also be a contraction of the crystal in the same direction as the magnetic field [7].

Later, Villari discovered that the magnetisation of magnetostrictive material could be changed when the external stress is applied. The basic equations for magnetostriction can be written as below:

$$\varepsilon = s_e \sigma + d' H$$

$$B_{MI} = d' \sigma + \mu H$$

here ε is the induced strain due to applied stress σ , B_{MI} is the magnetic induction and H is the magnetic field, s_e is the elastic compliance, d' is the magnetoelastic constant and μ is the magnetic permeability. The d' can be written as $d' = d^m + m_{elastic} H$. d^m is the piezomagnetic constant and $m_{elastic}$ is the second-order magnetoelastic constant [8, 9].

Terfenol-D, which constitutes of $Tb_{0.3}Dy_{0.7}Fe_2$, has the highest magnetostriction constant at room temperature. Due to its magnetostriction property, Terfenol-D is currently being used for various applications[10]. Interestingly, the name Terfenol comes from the composition for alloy $TbFe_2$. Further, Galfenol which is an Fe-Ga alloy also has been reported for its magnetostriction properties [11]. Different studies have been reported to improve the mechanical properties of Galfenol for the purpose of promising applications. The composites

of NbC particles+dendrites/Fe₈₁Ga₁₉ showed substantial improvement due to the grains refinement strengthening and reinforcement of precipitates [12]. Previously, Deng et al. have also reported the magnetic induction property as a function of stress major loops of Galfenol and Terfenol. For Terfenol, the maximum piezomagnetic coefficient was 18.42 GP⁻¹ and for Galfenol, 19.53TGPa⁻¹[13].

Fe-Co has also been reported as good magnetostrictive material for some applications because of its low price as compared to Galfenol and Terfenol-D. Magnetostriction value of FeCo as compared to that of Terfenol and Galfenol has been shown below in table 2.1. The mechanical properties of FeCo alloys are considerably better than the Terfenol-D and Galfenol, making them better for device application which requires bending[14].

Table 2.1. Material properties of most Fe-Co alloys, Terfenol-D, and Galfenol.

	FeCo alloy	Terfenol-D	Galfenol
Magnetostriction (ppm)	~ 140	3000	300
Coercivity (A/m)	<200	300	3000
Saturation Flux density (T)	More than 2	1.0	1.5
Tensile Strength	600	30	400
Elongation (%)	<30	<1	1
Young's Modulus (GPa)	200	<100	<100
Curie Temperature (°C)	900	380	680

2.5 Spin orbit coupling

In magnetic bilayer thin film systems constituting heavy metal/ferromagnet or ferromagnet/oxide interfaces, spin orbit coupling may arise from the heavy metal layer or broken inversion symmetry. Spin orbit coupling (SOC) can be explained in layman language as a relativistic effect arising from the interaction of an electron's spin with its motion in orbit around the nucleus. For an electron moving with velocity $\vec{v}$ in an electric field $\vec{E}$ generating an effective magnetic field, which can be written as

$$\vec{B}_{eff} = \vec{E} \times \vec{v}/c^2$$

The engendered magnetic field precedes a Zeeman interaction between spin (S) of the electron and the effective magnetic field. The resultant Zeeman interaction depends on the momentum of an electron in motion and can be given by

$$H_{SO} = -\mu_B \left(\vec{E} \times \frac{\vec{v}}{c^2} \right) \cdot \vec{S}$$

The produced electric field can stem from the neighbouring ions, the nucleus of the same atom, or even by the broken inversion symmetry from an interface. This phenomenon subsequently generates various phenomena holding immense opportunities for spintronics applications.

SOC of atoms at interface of heavy metal and ferromagnet layer with broken inversion symmetry can induce a perpendicular magnetic anisotropy (PMA). The ground state magnetisation in such a system prefers to be perpendicular to the interface. The SOC and crystal field due to the neighbouring ions link the electron motion with the crystal lattice. Such motion results in a preferred alignment of electron moments. Hence, resulting in the formation of easy magnetisation and hard magnetisation axis.

2.5.1 Interfacial Dzyaloshinskii Moriya interaction

Interfacial Dzyaloshinskii Moriya interaction (DMI) is an antisymmetric exchange interaction proposed by Dzyaloshinskii in 1957 [15]. A few years later, a report by Moriya connected this interaction to the SOC [16]. SOC in the presence of broken inversion symmetry gives rise to a non-collinear arrangement of spins with a particular rotation. Functionally, it can be expressed as a resultant phenomenon of interaction between neighbouring spins $\vec{S}_i$ and $\vec{S}_j$ mediated through a third ion with high SOC [16]. This interaction can be written as

$$H_{DMI} = \vec{d}_{ij} \cdot (\vec{S}_i \times \vec{S}_j)$$

$\vec{d}_{ij}$ is the DMI vector shown in the figure 2.6. In a ferromagnetic material, the DMI competes with the exchange interaction, resulting in spin canting. The sign of $\vec{d}_{ij}$ depends on the

direction of rotation of spins from $\vec{S}_i$ to $\vec{S}_j$. The DMI vector $\vec{d}_{ij}$, shown in figure 2.6 is perpendicular to the plane depicted by the red arrow. $\vec{d}_{ij}$ can be given as $\vec{d}_{ij} = d_{ij} \cdot (\vec{r}_{ij} \times \vec{z})$, where $\vec{r}_{ij}$ represents the distance connecting vector of two spins and $\vec{z}$ is the direction normal to the film oriented from the heavy metal layer to the ferromagnetic thin film[17-20].

Figure 2.6. Interfacial DMI described by Fert-Levy as an interaction between two adjacent spins S_i and S_j of a ferromagnet (FM) mediated through a heavy metal (HM) atom with strong spin orbit coupling.

Remarkably, the existence of DMI can be found in bulk systems too, with B20 non-centrosymmetric crystals, e.g., FeGe and MnSi. On the other hand, DMI can also exist in thin films originating from the interface between a ferromagnetic material and a heavy metal. Due to its origin, DMI in thin films is typically called interfacial-DMI. Moreover, the existence of inversion asymmetry is natural in bi/tri-layer ultra-thin film systems[21, 22].

Two approaches can comprehend the interfacial DMI. The first approach is the Fert-Levy model, and the second type is due to the Rashba model.

Fert-Levy model

Fert-Levy model is the most commonly recognized model for DMI in heterostructures (structure used in thesis). In their indirect exchange model, the role of the mediator between

two spins of two magnetic ions is played by conduction electrons, which become spin-polarized in the proximity of these ions. The model relies on the presence of an impurity centre with high spin orbit interaction, which would perturb the spin state of the conduction electrons when it passes by[23].

DMI energy can be written on the basis of micromagnetic model as the following [24-26]:

$$E_{DMI} = t \int_S D \left[\left(m_x \frac{dm_z}{dx} - m_z \frac{dm_x}{dx} \right) + \left(m_y \frac{dm_z}{dy} - m_z \frac{dm_y}{dy} \right) \right] dS$$

where t is the thickness of ferromagnetic film, D is the effective DMI constant, $m_i = M_i/M_s$: normalized magnetisation. The relation between effective DMI D and d_{ij} can be given as $D = d/at$, here a is the lattice constant.

Ferromagnetic/heavy metal interfaces exhibit large interfacial DMI values. For example, in thin films of Pt/CoFeB stack, the DMI value can vary from 0.5 to 1.5 mJ/m² [27-29]. In PdFe/Ir(111) bilayers [30] and Pt/Co/Ir/Pt stacks [31], DMI values of 3.9 mJ/m² and 3.0 mJ/m² respectively, have been reported. The DMI, if large enough, can compete with the other magnetic properties like exchange coupling and interface anisotropy energy. This competition assists in stabilizing chiral Néel walls or skyrmion.

Rashba model

Rashba model of DMI is based on the Rashba effect, which occurs at the interface of ferromagnet and oxide layers [32, 33]. This phenomenon can be understood due to the breaking of inversion symmetry in ultra-thin films, which generates an electric field $E\hat{z}$ at the interface. In this case, the spin orbit coupling can be written as[34, 35]:

$$H_{RSOC} = \alpha_R \vec{\sigma} \cdot [\vec{k} \times \vec{z}]$$

here $\vec{k}$ is the electron momentum: which results in the splitting of energy bands for electron spin at the interface and α_R is Rashba parameter, which is proportional to the electric field as

$\alpha_R \propto -(\hbar^2 E / 2m_e^2 c^2)$. The effective magnetic field produced by Rashba effect can be written as[36]:

$$\vec{B}_R \propto [\vec{k} \times \vec{z}]$$

This Rashba effective magnetic field results in torques acting on magnetisation. In other words, the itinerant electrons of a ferromagnet at interface faces spin precession around the effective magnetic field $\vec{B}_R$.

Figure 2.7. Chiral magnetic configuration due to the Rashba effect at the ferromagnet(FM) and metal oxide (MO_x) interface. E_z indicates the intrinsic interfacial electric field, which leads to the Rashba effect.

Due to exchange interaction with the neighbouring magnetic moments, tilting of the spins with a specific rotation is observed, resulting in a chirality. This mechanism that arises due to antisymmetric exchange interaction is called Rashba-DMI. Hence, the Rashba DMI is capable of stabilizing the chiral spin textures.

DMI constant due to Rashba effect $D_{Rashba} = 2K_R A_{ex}$ [34], where K_R is the Rashba coefficient given by $K_R = 2\alpha_R m_e / \hbar^2$ and A_{ex} is the exchange constant. As the Rashba parameter is proportional to the interfacial electric field, one of the most interesting features of this phenomenon is that it can be sensitive to applied external electric field to facilitate manipulation of spins. This would lead to tuning of the interfacial DMI with applied electric field.

2.5.2 Spin orbit torque

Spin orbit torque (SOT) can originate from spin Hall effect (SHE) or the interfacial Rashba-Edelstein effect. SHE is an outcome of spin orbit coupling and the Rashba-Edelstein effect as the name suggests, is the outcome of the Rashba effect.

Spin Hall effect

The Spin Hall effect (SHE) can be construed as the conversion of unpolarized charge current into a pure spin current. SHE is a bulk spin orbit coupling effect that occurs in the heavy metal, causing separation of spin current of opposite polarity in the transverse direction of charge current. This effect was first predicted by D. Yákonov and V.I. Perel in 1971 [37, 38]. Later Hirsch and Zhang reapproached this phenomenon and renamed it the spin Hall effect [39, 40]. The SHE was first verified experimentally in GaAs semiconductor and Al or Pt metals [41-43]. SHE leads to converting a longitudinally applied charge current into deflection of spin-up and spin-down electrons in the transverse direction. The spin current produced by SHE is described as

$$j_s = \frac{\hbar}{2e} \theta_{SH} (j_c \times \sigma)$$

here j_s is the generated spin current, j_c is the applied charge current, σ is the polarization of spin current. $\hbar$ is the reduced Planck constant, θ_{SH} is the spin Hall angle (SHA) which is the conversion efficiency of spin current from charge current, and e is the elementary charge.

Figure 2.8. Schematic demonstration of Spin Hall Effect resulting a transverse spin current j_s (z-direction) with spins polarized ($j_c \times z$) in y-direction, in response to a charge current j_c (x-direction) in heavy metals (HM). This generates spin orbit torque, which manipulates the magnetisation m in the adjacent ferromagnetic (FM) layer.

The generated spin current from SHE enters the ferromagnet and passes on the spin to local magnetic moments via angular momentum transfer. These local spins get transfer to the magnetisation of the ferromagnetic layer via exchange interaction resulting in the switching of the magnetisation. The origin of SHE can be extrinsic and/or intrinsic. The three leading causes behind the SHE are listed below. The extrinsic contributions can be classified as skew scattering and side-jump, which arises by impurity scattering. In contrast, the intrinsic contribution originates from the crystal field.

Skew scattering: Mott discovered in 1929 that spin-carrying particles scattered asymmetrically by a central potential [44]. The magnetic field which acts on the electron is inhomogeneous, which engenders a spin-dependent force. The generated force is proportional to the gradient of Zeeman energy, which acts on the electron [45]. The direction of the force depends on the spin of the electrons, resulting in the deflection of opposite spins in opposite direction. The skew scattering leads to a conductivity perpendicular to the incoming particles.

Side-jump scattering: Side jump scattering originates from the anomalous form of the velocity operator in the SOC coupled system [46]. The semiclassical explanation by Berger interpreted the discontinuous sideways displacements of the centre of mass of the electron wave package upon collision. The shift in the direction depends on the spin orientation of the electron [47, 48]. These side-jumps are overlaid with the skew scattering and result in the conductivity perpendicular to the incoming particles.

Intrinsic scattering: The intrinsic scattering can be understood due to the Berry phase mechanism originating from anomalous velocity. It is dominant in heavy metal with significant spin orbit interaction. The anomalous contribution is linear to the applied electric field and it contains the Berry curvature, resulting in a generation of spin current perpendicular to the direction of the applied electric field [49].

Figure 2.9. Generation of Spin Current via spin Hall effect (a) Skew scattering at an impurity. (b) Side-jump scattering at an impurity, resulting in a spin-dependent displacement δ_{sj} . (c) Intrinsic mechanism. These effects led to the splitting of opposite spins in the transverse direction of the current [46, 50].

Interfacial Rashba-Edelstein effect

The Interfacial Rashba-Edelstein effect is another source of SOT in ultra-thin films, which arises at the interface of metal/ferromagnet. As the name suggests, it is based on the Rashba effect, which is of pure interfacial origin, as reported in the previous section. The Rashba effect leads to a homogenous net spin polarization resulting in a spin current perpendicular to the

applied charge current. The phenomenon is called the Edelstein effect/inverse spin Galvanic effect [51, 52]. The engendered spin current switches the magnetisation in the adjacent ferromagnet. The switching is done by the exertion of spin orbit torque, which has the exact symmetry as SHE. Rashba spin-orbit effect lifts the degeneracy between opposite spins of conduction electron at the interface of heavy metal/ferromagnetic interface. The spin state is strongly linked with momentum k . The spin state is opposite for opposite momenta on a Fermi surface for energy minimization. On application of current in the metal, the Fermi surface is shifted, which results in an uneven occupation of k and $-k$ momentum states. These uneven momentum states can lead to spin accumulation which generates spin torque.

Figure 2.10. (a) Spin split Fermi surface of the electron in the presence of Rashba spin orbit coupling with zero spin density. (b) When a current is applied, the Fermi contour shifts. Due to this shift, an additional magnetic field $\vec{B}_R$ is generated. This magnetic field causes all spins to tilt in the out of plane direction (shown by opaque arrows). The produced non-zero spin density diffuses as spin current into the adjacent ferromagnetic layer resulting SOT [53].

2.6 Domain walls in thin films

A magnetic domain is an area in ferromagnet where magnetic moments are aligned uniformly in one direction. Magnetic materials can consist of one or multiple domains, depending on their magnetic properties and size. Besides, the applied magnetic field also plays a vital role in deciding the number of domains in a sample. For example, when a demagnetisation field is

applied, the individual magnetisation of domains reduces the net magnetisation to zero. The number of domains observed in a demagnetized state is maximum. However, a fully magnetized state, on the other hand, has only one magnetic domain, which depicts the maximum net magnetisation of the system.

Different magnetic energies determine the structure of domains in a magnetic system. For example, dipolar energy facilitates the formation of multiple domains to minimize the surface charges. On the other hand, the exchange coupling energy favours uniformly aligned neighbouring magnetic domains. In contrast, the anisotropy energy prefers the orientation of magnetisation along a specific crystallographic direction, resulting in an easy axis/plane. Based on these energies, different numbers and shapes of domains are formed in a ferromagnetic film.

Figure 2.11. Schematics of two types of domain walls depicting their planes of rotation of spins: (a) Bloch type (b) Néel type.

A domain wall is the region of transition between two magnetic domains. In other words, a domain wall is a boundary between domains. The width of a domain wall depends on the different competing energies of a ferromagnetic system. In a ferromagnetic system, domain walls can be of two configurations – the first is called the Bloch wall, which rotates in the axis

perpendicular to the domain wall plane. The second one is called Néel wall, which rotates in the axis parallel to the domain wall plane[54, 55]. For ultra-thin films with thickness less than one nm, the surface anisotropy is substantially strong, which favours the out of plane magnetisation. In such ultra-thin films, all magnetic energies contribute to the type of domain wall. In these films, the magnetic energies remain the same, but the magnetostatic energy for the Bloch wall is lower than the Néel wall. Hence, Bloch wall is favoured in such films. However, in a heavy metal/ferromagnet/metal-oxide stack, the existence of DMI lowers the energy for the Néel wall and hence, favours the formation of Néel walls. The set of samples studied in the 6th chapter has a ferromagnetic film thickness of 0.6nm. The magnetic domains constitute in these films are of Néel wall configuration.

Rohart and Thiaville [26] have carried out comprehension of DMI contribution to the domain wall energy. In the explanation, the 1-D system was considered where z is the easy axis and the change in the magnetisation occurs along the x -axis. As the DMI vector $\vec{d}_{ij}$ is orthogonal to the vector connecting two spins $\vec{r}_{ij}$ and the axis of inversion asymmetry is z -axis, DMI favours rotation of the magnetisation in x - z plane with a fixed chirality. The angle θ can be used to describe the variation $m(x)$. Considering $\theta = \cos(\vec{m}, \vec{z})$, the total micromagnetic energy density can be written as

$$E[\theta(x)] = \int_{x_A}^{x_B} [A_{ex} \left(\frac{d\theta}{dx}\right)^2 - D \frac{d\theta}{dx} - K_{eff} \cos^2 \theta] dx$$

here, A_{ex} is the exchange constant, D is the effective DMI constant, x_A and x_B are the sample boundaries in x direction and K_{eff} is the effective magnetic anisotropy. Using standard variation calculus the function $\theta(x)$ can be written as for the following equations:

$$\frac{d^2\theta}{dx^2} = \frac{\sin\theta\cos\theta}{\Delta^2}, x_A < x < x_B$$

$$\frac{d\theta}{dx} = \frac{1}{\tau}, x = x_A \text{ or } x = x_B$$

here $\Delta = \sqrt{A_{ex}/K_{eff}}$ is Bloch-width parameter and $\tau = 2A_{ex}/D$ which is related to DMI by integrating equation:

$$\left(\frac{d\theta}{dx}\right)^2 = \frac{C + \sin^2 \theta}{\Delta^2},$$

where C is an integration constant. If D is relatively small (assuming an infinite system in x direction), domain wall energy is still positive. The integration constant C needs to be zero so that no cycloid develops. The above equation will have two solutions-one with uniform $\theta = 0$ or π . The second one refers to where θ is changing that is domain wall width and can be written as

$$\theta(x) = 2 \arctan \left[\exp \left(\pm \frac{x - x_0}{\Delta} \right) \right] + n\pi$$

here n is an integer, the $\pm$ sign determines chirality of domain wall and n enables type of wall ranging from 0 to $\pm\pi$ or from $\pm\pi$ to $\pm 2\pi$. The x_0 is the position of the domain wall. The domain wall energy can be written from equations as

$$\sigma_{DWE} = 4 \sqrt{A_{ex}K_{eff}} - \pi D$$

In the above equation D is considered positive. DMI lowers the energy of Néel wall by introducing the chirality, which favours the rotation of magnetisation in the domain wall in the x - z plane[56, 57].

Further σ_{DWE} , helps to understand the density of domains in magnetic samples based on magnetic properties. A low σ_{DWE} results in a high density state of magnetic domains (as shown in the chapter 6).

Figure 2.12. Different types of chirality in Néel Domain Wall. The index figure in hand points the direction of spin. (a) Right-handed chirality where the spins rotate clockwise. (b) Left-handed chirality where the spins rotate anticlockwise.

In the materials systems with large DMI, larger than a critical DMI value, the Néel type domain wall becomes more energetically favourable. Moreover, the sign of DMI determines the chirality of the Néel domain wall [21, 22, 58]. Noticeably, there might be discrepancies in conventions for the DMI sign and the DMI vector $\vec{d}_l$ defines the chirality. In the conventions of this thesis, negative DMI results in a clockwise rotation of magnetisation across the domain wall, referring to a right-handed chirality. On the other hand, a positive DMI results in an anticlockwise rotation of magnetisation across the domain wall, referring to left-handed chirality. Both types of chirality are shown in figure 2.12.

2.7 Magnetic skyrmions

The root of the name "skyrmion" goes back to the particle physicist Tony Hilton Royle Skyrme, who gave a theoretical model to define the existence of subatomic particles such as neutrons and protons as discrete entities emerging from a continuous nuclear field [59]. Interestingly, the name skyrmion can be given to any topological spatial texture (like the twist in a Möbius strip), which is preserved under continuous deformation. Since the work in this thesis is about the skyrmion in magnetic structures, henceforth literature review about the magnetic skyrmion has been carried out. The theoretical prediction of magnetic skyrmions can be tracked back to the 1980's [60, 61]. However, the first physical realization of magnetic skyrmion was reported in 2009 in the B20 compound [62].

A skyrmion is a topologically stabilized spin configuration, which behaves (moves and interacts) as a quasi-particle in a magnetic structure [63, 64]. The beauty of a skyrmion is the arrangement of spins: a circular whirl-like configuration that gradually transits from an up or down direction at the outer circumference to reach in the opposite direction at the centre of configuration. Noticeably, the spin direction at the centre of the configuration is opposite to the magnetisation direction of the magnetic films in which a magnetic skyrmion is observed. Based on the transition of spins from the outer circumference to the centre of skyrmion, there are two types of magnetic skyrmions: Néel skyrmion and Bloch skyrmion (figure 2.13).

Figure 2.13. Magnetic texture of skyrmions| (a) Néel-type and **b.** Bloch-type skyrmions with the magnetisation rotating from the down direction at the skyrmion's centre to the up direction of the external uniform magnetisation at the skyrmion's edge, as in a Néel or in a Bloch domain wall.

A way of differentiating magnetic skyrmions is by their topological winding number $\mathcal{S}$, which can be defined as below:

$$\mathcal{S} = \frac{1}{4\pi} \int m(r) \cdot \partial_x m(r) \times \partial_y m(r) dr = \pm 1$$

The topological number $\mathcal{S}$ characterizes the winding of the localized magnetisation $m(r)$ with respect to the position (r) [65]. This normalized magnetisation m can be mapped on a unit sphere, which covers the entirety of the sphere. The positive or negative value of $\mathcal{S}$ indicates the direction of spin magnetisation up or down at the centre of skyrmion, respectively. Remarkably, the topological number is fixed for a magnetic skyrmion, and the spin configuration cannot be twisted continuously to result in a new spin configuration giving rise to a different topological number. The chirality of a magnetic skyrmion texture, which defines the characteristics of a spin transition, is also fixed for a fixed topological number [66-68].

The physical existence of the first magnetic skyrmion was reported in a single B20 crystal of MnSi compound, which has helical phases [62]. However, this realization of magnetic

skyrmion was observed only at low temperatures. Another paper reported the realization of magnetic skyrmion one year later in the helical phase of FeCoSi thin film, which was also at low temperature [69]. Later, observations of skyrmions were reported in a monolayer of Fe or bilayer of FePd grown on Ir(111) [65, 70]. In these ultra-thin films, a considerable value of DMI arises due to the interface between Fe and Ir(111). However, the stabilization of the skyrmion's lattice phase at this interface required a high magnetic field along with a necessity of low temperature. The diameter of observed skyrmions in these ultra-thin films was minimal, extending to a few lattice parameters.

During the early stages, skyrmions were realized only at low temperatures. However, for any practical application, stabilization of such structures at room temperature is required.

2.7.1 Skyrmions at room temperature

A few years after the discovery of low-temperature skyrmions, compact magnetic skyrmions with a size of a few 100 nm were observed at zero field and room temperature [71, 72]. Larger skyrmions were observed where lower DMI allowed for different topologies in general [73]. The stabilization of these magnetic skyrmions at room temperature in films with interfacial perpendicular magnetic anisotropy was possible due to the existence of interfacial DMI arising at the interface of magnetic film/heavy metal [72, 74-76].

Advent of compact magnetic skyrmion at room temperature was reported in the multilayers of Pt/Co/Ir by Moreau-Luchaire et al [72]. The stabilization of magnetic skyrmion in this structure was achieved by the additive interfacial DMI from second heavy-metal and larger magnetic volume (achieved via increasing the repetitions of the ferromagnetic layer), improving the thermal stability of skyrmions. The increase in the magnetic volume anticipates to introduce the coupling of magnetic textures vertically throughout the multilayers, similar to what has been observed in coupled domain walls in Co/Pt multilayers reported by Baltz et al. [77].

Several other reports came out exhibiting the observation of stable magnetic skyrmions at room temperature in different multilayer stacks like Pt/Fe/Co/Ir, Pt/Co/Ta, and Ta/CoFeB/MgO structure, which is commonly used for memory applications [71, 78-80]. In the Pt/Fe/Co/Ir and Pt/Co/Ta multilayer structures, the stability is achieved by additive interfacial DMI and increased thermal stability, as explained before.

Several analytical and experimental methods have investigated the nucleation of skyrmions. These methods involve the nucleation of skyrmion using STT, SOT, magnetic field, electric field, or thermal excitation [70, 71, 75, 78, 79, 81-92]. To nucleate skyrmion, in a thin-film multi-layered structure sample with interfacial DMI, threshold energy (via thermal, magnetic field, or electric current) must be applied. This applied threshold energy transforms the magnetic domains from the labyrinthine stripe to circular skyrmions. Figure 2.14 shows such transformation (achieved by using simulations) of magnetic domains from the labyrinthine phase to the skyrmion phase by applying a magnetic field. The rate of skyrmion creation at a defined temperature (T) can be calculated by Arrhenius Law, which also relates the lifetime of skyrmion with the energy barrier [93-95]:

$$\tau \sim \exp^{\Delta K_u V / K_B T}, \quad (5)$$

where K_u is uniform anisotropy energy constant, V is the volume of the sample and K_B is the Boltzmann energy constant.

Figure 2.14. Effect of applied energy field at zero field on a magnetic state with labyrinthine stripes. Simulations results show the transformation of a labyrinthine stripes state of $(\text{Pt/CoFeB/MgO})_{12}$ to a skyrmion state under applied magnetic field at room temperature. Images are $1.5 \times 1.5 \mu\text{m}$ in size. This sample has been studied in chapter 6.

Once the magnetic skyrmions nucleate at a temperature, skyrmions can be stabilized even on removal of threshold energy, if the in-plane component of magnetisation is high enough to stabilize the skyrmions. However, to get the advantage of skyrmion structures for devices applications, stabilization of skyrmions at room temperature is required. For stabilizing the skyrmions at or above room temperature, distortion (or annihilation) of skyrmion structure due to thermal fluctuations of magnetisation needs to be avoided [88, 96]. For stabilizing the skyrmion at room temperature, the required stabilizing in-plane component of magnetisation can arise due to DMI which is higher or equal to the critical DMI (D_c) value above which total domain wall energy becomes zero [18, 97]. Besides the DMI, anisotropy energy ($K_u V$) also contributes a crucial role in stabilizing the skyrmion structure at room temperature. Various studies have been reported, in multi-repetition layered-structures, which signify the role of anisotropy energy (especially the volume of sample) in achieving stable skyrmions structure at

room temperature. Example of such structures includes (Pt/CoFeB/MgO)₁₅ [71], (Pt/Co/Ir)₁₀ [72], and (Ir/Fe/Co/Pt)₂₀ [78], where heavy metal layer induces a high interface DM interaction to the structures and repetition of layers induces thermal stability due to dipolar coupling.

2.7.2 Motion of skyrmions

The advent of magnetic skyrmion was exciting, not only for gaining the insight of physics behind skyrmion's structure, but also for the propitious application of skyrmion in devices. These skyrmion-based devices involve the motion of skyrmion on a track. The first few reports stating the movements of skyrmion by current were reported in MnSi (rotation of lattice) [98] and FeGe (motion in the flow regime) [99]. Further reports ascribing the use of spin transfer torque (STT) exerted by the spin-polarized current to move skyrmion were reported, which also exhibited the promising potential of using skyrmion in a racetrack for various applications [81, 100]. Besides the current induced motion, a ratchet motion of skyrmion crystal in MnSi and Cu₂SeO₃ through magnons was also reported in 2014, signifying the possibility of skyrmion motion through a spin wave [101]. The current induced displacement of skyrmion bubbles was first reported in thin-film structures by applying electric pulses [71, 73]. Later, multiple reports demonstrating the motion of skyrmion through spin transfer torque or current in plane were reported [86, 102-108]. Besides the skyrmion motion through current in-plane, skyrmion motion through spin current perpendicular to the plane approach, which relies on the spin Hall effect and inverse spin Hall effect, also has been reported. The spin current perpendicular to the plane gives rise to a high velocity of skyrmion as compared to the charge current in-plane.

The conversion efficiency of spin current through the spin Hall effect depends on the spin Hall angle of the heavy metal [109]. Hence, by using a heavy metal with a high spin Hall angle, the velocity of the skyrmion can be further improved. Besides the method of introducing torque (current in-plane or current perpendicular to plane) to the magnetic stack, the motion of skyrmion also depends on the magnetic stacks' morphological properties. A granular ferromagnetic layer structure (like Pt/Co/Ta) with grains will have variation in magnetic properties at the boundaries of grains. Such grain boundaries act as pinning sites to the skyrmion, causing a reduction in velocity or annihilation of skyrmion depending on the applied current density and relative size of skyrmions and grains. On the other hand, an amorphous ferromagnetic stack (like Pt/CoFeB/Ta) can mitigate these detrimental effects [71].

Interestingly, the motion of skyrmion in a track suffers from a Magnus force–type effect, where the skyrmion travels in the parallel as well as the transverse direction of the track (also known as the skyrmion Hall effect). This was experimentally first observed in Ref. [110-112]. The Magnus effect causes the skyrmion motion towards the edge of the track, which results in the annihilation of the skyrmion at the edge or bounce-back of the skyrmion from the edge depending on the velocity of the skyrmion. The Magnus force, which leads the unwanted transverse motion, is visible through the Thiele equation as follow [113, 114]:

$$\mathbf{G} \times \mathbf{v} + \alpha \mathbf{D} \mathbf{v} + \mathbf{F} = 0$$

where $\mathbf{F}$ is the force resulting due to the spin Hall effect combined with the repulsion by the track edges, $\mathbf{G}$ is a gyrovector oriented along the out-of-plane direction, and $\mathbf{D}$ is the dissipative tensor, which can be calculated from the parameters of the skyrmion [110, 113]. The first term on the left-hand side indicates the Magnus force, which causes the movement of the skyrmion away from the intended direction. In the above equation, $\mathbf{v}$ is the velocity of skyrmion, which has two components: longitudinal velocity (v_x) and transverse velocity (v_y). The latter

component of the skyrmion velocity is the one which leads to the annihilation of the skyrmion when the velocity of skyrmion is high [115-117].

Figure 2.15. Schematics of skyrmion motion in a track, depicting the skyrmion Hall effect. A transverse motion in the skyrmion leads to the annihilation of skyrmion at the edge of the track. (a) Track with a nucleated skyrmion. (b) Skyrmion under the effect of applied current, depicting the longitudinal and transversal motion. (c) Skyrmion at the edge of the track, under the process of annihilation. (d) Track after the skyrmion annihilation.

Many studies have been done to understand this effect to avoid or use this effect for any possible benefit [73, 111, 118-124]. To avoid the transverse motion due to magnus effect, several methods have been proposed. One of these methods uses synthetic antiferromagnetic layered structures, where the difference between the finite spin density in each layer is zero.

As a result, gyrovector $\mathbf{G}$ has no finite direction. Hence, the skyrmion moves in a straight line [125, 126]. Moreover, alternating theoretical schemes based on ferromagnetic skyrmionium also has been proposed, where the skyrmionium spin configuration nullifies the skyrmion number [127, 128]. In chapter 5, a computational study on restricting the skyrmion Hall effect by a potential barrier has been done.

2.8 Manipulation of magnetic properties via light ion irradiation

Ion Irradiation is a handy technique to introduce the displacement of atoms. Chappert et al [129], in 1998, first reported a paper demonstrating the switching of easy axis in Co-Pt multilayers. The carried out He^+ ion irradiation reduced the interface anisotropy, which resulted in in-plane magnetisation of the multilayers. Later, the same group used He ion irradiation to change the chemical order of FePt thin films [130]. More studies were carried out, which focussed on the magnetic media [131, 132].

The structure evolution of irradiation with light-ions irradiation is understand by a simple ballistic recoil mechanism model [133-137]. For He^+ ions radiation, where energies vary in the range of 5 to 150 keV, the collision cascades are absent, and modification of structures merely occurs in the vicinity of the ion path. In light irradiation, all ions settle deep into the substrate. Remarkably, the ion irradiation leads to recoil limited to one or two atomic distances, resulting in the substitution of $\text{Co} \rightarrow \text{Pt}$, $\text{Pt} \rightarrow \text{Co}$, $\text{Pt} \rightarrow \text{Pt}$, or $\text{Co} \rightarrow \text{Co}$. Due to such soft intermixing of atoms, the crystallographic structures are maintained. Moreover, reports have demonstrated that the irradiation of ions lacks any implantation into the magnetic layer [129, 132, 138], which sustains the stoichiometry intactness of the layer.

Figure 2.15. Schematics of Co-Pt atom layers are shown in purple and green colour spheres. Co atoms that move one atomic distance led to roughness at the interface. On the other hand, when the Co atom moves more than one atomic distance at high energies of irradiation, it becomes isolated from the ferromagnetic Co layer and results in Co-Pt alloying.

In ultra-thin films, irradiation might affect upper and lower interfaces differently [135]. For example, in the case of the Co/Pt/Co interface, the top Co interface goes through short-range intermixing leading to an increase in the roughness of the interface. On the other hand, the bottom interface goes through a higher range of mixing, leading to the formation of alloys. Moreover, the roughness amplitude increases linearly with the ion dose. Hence, by choosing the correct dose and energy, one can vary the intermixing of atoms at the interface. Therefore, the interface properties may be tweaked by changing the ion irradiation energy [139, 140].

Recently, studies have been carried out where the interface properties like interfacial magnetic anisotropy were reduced by irradiating He ions. In these studies, irradiation introduces the atomic displacement at the interface of the ferromagnetic layer and adjacent non-magnetic conducting layer. These displacements lead to the disordering of the interface, distorting the orbital hybridizations. Further, He⁺ ion irradiation can lead to DMI alternation[141]. As predicted, the intermixing of Co-Pt atoms at a rough Co/Pt interface may increase the DMI to

a certain extent [142]. These studies offer a promising route (via irradiation) to manipulate interfacial properties.

Interestingly, intrinsic magnetic properties such as Gilbert damping (α), saturation magnetisation and exchange coupling can also be manipulated by irradiation [140, 143]. Furthermore, the consequence of the ion irradiation induced variations has been demonstrated on the domain wall velocities [144], multi-level magnetisation switching [145] and domain wall transitions [146]. Moreover, for doses lower than 10^{20} ions/m², the roughness of the interface remains almost unaffected (varying <1.5%), and the morphology of layers does not show any blistering effect [135, 147]

More recently, ion irradiation has been used to nucleate skyrmions by introducing magnetic defects in materials with high magnetic anisotropy [148, 149]. The reduction in the anisotropy and magnetostatic energy in these samples showed promising potential for manipulating the magnetic properties of chiral samples. This route of manipulating magnetic properties has been explored in chapter 6 to tweak the physical behaviour of skyrmions.

2.9 Summary

In this chapter, different types of magnetic materials and magnetic energies were described. These magnetic energies assuage the stabilization of magnetic configuration in ultra-thin film systems. Further, different spin orbit interaction phenomena were discussed. The later part of the chapter has focused on the skyrmions. The nucleation, stability, and motion of skyrmions were discussed.

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3.1 Introduction

This chapter explains the details of sample deposition, patterning of microwires, imaging of magnetic textures, characterisation, and micromagnetic simulation. The first few sections emphasise the preparation of samples for experimental studies, viz. thin film deposition and lithography. Subsequently, characterisation techniques such as vibrating sample magnetometer (VSM) and magneto-optic Kerr measurements (MOKE), and alternative gradient field magnetometry (AGM) were explained. In the end, the details of micromagnetic simulations are provided.

3.2 Fabrication

3.2.1 Thin film deposition

The techniques to deposit thin films can be divided into two categories [1] (as shown in table 3.1.)

Table 3.1. Different Techniques for depositing thin films.

Technique	Deposition Type	Advantage	Disadvantage
Evaporation techniques	Physical	High Purity	Evaporating targets with high melting point
Molecular beam epitaxy	Physical	Achieve single crystal	Hard to control the alloying concentration
Sputtering	Physical	Co-sputtering: alloying, High Purity	Polycrystalline films, Inhomogeneities in films
Sol-gel	Chemical	Optical Transparency, chemical robustness	Poor brittleness
Plating	Chemical	Thin and adherent coating	Toxic Coating solutions, Subject to cracking and chipping
Chemical vapor deposition	Chemical	High deposition rate	Precursors evaporates at high temperature 600°C

A physical deposition is a generic term used to quantify thin film deposition processes that require condensation of the vaporised solid material on top of the surface of the solid material under a partial vacuum condition [2, 3]. The physical deposition is an atomistic deposition process in which there is a physical discharge of atoms or molecules and condensation and

nucleation of these atoms onto a substrate through a vacuum or low pressure gaseous or plasma environment. In the physical deposition, thin-film formation is caused by the condensation of atoms or molecules on the surface of the substrate due to evaporation, ion bombardment, or sputtering

Chemical Deposition is a generic term for deposition of the thin film via a series of chemical reactions. The process of material synthesis occurs when the constituent of the target in the vapour phase reacts via a chemical process near the surface or onto the surface of the substrate, leading to the growth of the thin film [4-7]. The chemical deposition is a thermodynamically complex process involving chemical reactions under specific conditions such as temperature, pressure, reaction rates, and momentum, mass, and energy transport.

I have chosen the sputter deposition process for my work, as it facilitates several advantages:

- a) Deposition of materials with high melting points.
- b) Co-sputtering of materials to form alloys.
- c) Control on the deposition rate and phases of the material.
- d) Deposition of high-quality thin films.
- e) Standard deposition tool in the industry.
- f) Availability in my lab.

Thin-film deposition of this thesis has been carried out using two types of DC magnetron sputtering systems. In the sputtering technique, the chamber is filled with a noble sputter gas (ultrahigh purity Ar in our case), and the target material is connected to a high negative voltage potential. After igniting a self-sustained plasma, the positive Ar^+ ions constantly bombard the target material, resulting in a release of the sputtered material, which then travels towards the substrate (and the chamber walls). Typically, in a magnetron sputtering, magnets are placed behind the target to produce a magnetic flux around the target. This magnetic flux leads to a higher ionisation of Ar atoms. High ion flux increases the bombardment of Ar^+ ions with the

target, resulting in a higher deposition rate [8, 9]. The deposition of Pt/CoFeB/MgO stack was done on a 1000 nm SiOx coated Si substrate. The deposition of the stack (used in chapter 6 for studying skyrmion) was carried out by the Singulus Timaris magnetron sputtering. Sputtering of metals was carried out by DC current. Deposition of MgO, which is an insulating material, was carried out by radiofrequency (RF) sputtering after matching the impedance with plasma. Whereas the deposition of thin films for energy harvesting work (chapter 3) has been carried by using facing targets sputtering (FTS) deposition system. FTS is a system with two magnetron sources positioned opposite to one another and at right angles to the substrate (shown in figure 3.1). The geometry of FTS can confine the plasma between the two sources. The direction of plasma can be used to set the direction of magnetisation of deposited material on the substrate[10, 11].

Figure 3.1. Schematics of facing target sputtering (FTS) deposition system. The magnetic flux is closed between two targets. A substrate is kept at a right angle to the targets [11].

Noticeably, the electron density produced in the FTS reaching the substrate is less than the conventional magnetron sputtering. The deposition rate can be varied by varying the distance

between the substrate and the plasma. The fringing field can be used to set the direction of easy axis magnetisation in the deposited soft magnetic materials. The stack used for study in chapter 4, CoNi/FeCo/CoNi, was grown at room temperature at an Ar pressure of about 0.24 Pa on two different substrates: glass substrates and 85 μm thick Kapton (a flexible polyimide film) membranes [11, 12].

3.2.2 Deposition of microwire patterns and devices for energy harvesting

Fabrication of the CoNi (3 nm)/FeCo (30 nm)/CoNi (3 nm) microwires was carried out with different widths (4 μm to 40 μm), and a constant length of 1 mm on a Kapton (a flexible polyimide) membrane. An optical – photolithography tool (NEOARK desktop maskless lithography) was used to fabricate the patterns [11, 12]. For this purpose, the optical light photoresist was spin-coated on the membrane and exposed to an optical light laser to etch trenches in the shape of microwires of defined dimensions. Then the trilayer magnetic stack was sputter deposited on the membrane using the facing targets sputtering. Finally, isolated microwires were achieved by treating the sample with a resist remover to wash away the unwanted deposited material.

A preliminary device was fabricated to exhibit the potential of our reported science as an alternating approach to replace the power source for IoT devices. Figure 3.2 shows the schematics of steps involved in the fabrication of the device. Spin coating of the photoresist was carried out on a polyimide membrane. The photolithography tool was used to make patterned trenches of the coil shown in figure 3.2a). Then sputter deposition of Ta(10 nm)/Cu(50 nm)/Ti(10 nm) on the sample was done. A lift-off process was carried out to form a patterned coil shape of one side (figure 3.2b). The photoresist was coated again and etched out using photolithography, exposing the inner area, where a 100 nm thick insulating layer of SiO₂ was deposited using RF sputtering (figure 3.2b). Subsequently, a lift-off process was

again carried out. Then the trilayer magnetic stack (figure 3.2c) and successively another layer of SiO₂ were deposited (figure 3.2d). Figure 3.2e shows the cross-section of the device fabricated so far. Finally, photoresist was coated on the sample and etched using photolithography to deposit the other side of the coil stack, as shown in figure 3.2e. The fabricated device was then connected to a 50 Ω resistive load and an oscilloscope for measuring the harvested voltage.

Figure 3.2. Schematics of steps involved in the fabrication of the preliminary device. (a) Coil-shaped trenches were formed using photolithography, and Ta/Cu/Ti was sputter-deposited consecutively. A lift-off process was carried out to get the final pattern. (b) An insulating layer of SiO₂ was deposited on the coil stack using photolithography. (c) Trilayer magnetic stack was deposited on the SiO₂ layer. (d) The second layer of SiO₂ was deposited atop the trilayer stack to prevent short-circuiting. (e) The final layer of the coil stack was deposited on the sample. Electrodes were connected to a resistive load and oscilloscope to measure the voltage [11].

3.3 Magnetometry

Magnetic hysteresis (MH) loops of a thin film sample can reveal essential details about the magnetic properties. The loops are used to extract critical magnetic properties such as uniform crystal anisotropy (K_u), saturation magnetisation (M_s) and coercivity (H_c), saturation field (H_s), and magnetic remanence (M_r). The shape of MH loop may help in getting information about the magnetic domain density and switching type of magnetisation. The MH loop measurement includes plotting magnetisation of thin-film samples as a function of a swept magnetic field. The following part explains different magnetometry techniques and methods used for calculating different properties

3.3.1 Vibrating sample magnetometry

The hysteresis loops measured for the skyrmion chapter (chapter 6) were carried out by using a vibration sample magnetometer (VSM) by Lakeshore (VSM 8600). In this technique, a sample is placed between two poles of a strong electromagnet. The probe attached to the sample vibrates at a specific frequency. The vibration of the sample introduces a time-varying flux Φ in the nearby pick coils. The change in magnetic flux induces a voltage $V = -d\Phi/dt$ in the pickup coil. The generated voltage is proportional to the magnetic moment of the sample. The absolute value of the sample's moment is calculated accurately with the help of calibration using a standard sample. In our VSM measurements, a Nickel sphere was used as the standard calibration. The measured magnetic moment of the sample is then divided by the volume of the magnetic material to calculate the magnetisation (which has units of A/m or emu/cc).

VSM is an accurate technique that can detect a magnetic moment $\approx 10^{-8}$ emu (VSM8600). To measure M_s of the samples accurately, the probe was set to its saddling point by optimising the position in all three axes. For multiple magnetic layers, the easy axis hysteresis loop rarely has

a square shape, and the hard axis is also not linear shape. Such a shape of the loop is understood by the splitting of a single domain into multiple domains. The switching of the magnetisation from one direction to another in such loops undergoes through the propagation of domain walls in [13].

In perpendicular magnetic anisotropy (PMA) films depicting multiple domains, the loops are butterfly shape. The effective anisotropy can be calculated by [13-15]:

$$K_{eff} = M_s \left[\int_0^1 B_{op}(m_{op}) dm_{op} - \int_0^1 B_{ip}(m_{ip}) dm_{ip} \right]$$

$$\text{where } M_s = M(B)|_{B > B_s}$$

where B_{op} and B_{ip} are the applied magnetic field and, m_{op} and m_{ip} are the normalised value of magnetisation in the out-of-plane and in-plane directions, respectively. For the equation to be valid the initial magnetisation should be demagnetised, and the final state should be kept saturated. The shape of the loop and value of the K_{eff} can be used as a control parameter for tracking “good” samples with a multidomain behaviour and PMA.

Measurement of K_{eff} was done by following the formula mentioned above. The magnetisation curves of out-of-plane and in-plane field directions were plotted for every sample (figure 3.3). The value of K_{eff} was measured by calculating the difference between areas of out of plane and in-plane magnetisation. As can be seen from figure 3.3, there are four chords: two for out of plane and two for in plane applied external field, which represent the measured magnetisation for different respective hysteric of the loop. An average of the difference in the area was calculated by measuring the areas between two out of plane and in plane chords. For this, a python code was written which calculates the area under the curve for every single combination of chords (total four combinations). These combinations are shown in figure 3.4.

Figure 3.3. The plot of MH loops (showing the value of magnetisation from zero to positive applied external field) of pristine sample used for skyrmion study (chapter 6). Curves 1 and 3 (2 and 4) represent the measured loop from positive to negative (negative to positive) applied external field in out of a plane and in-plane directions, respectively.

Figure 3.4. The magnetisation plots of different curve combinations of a pristine sample. The light green teal colour shows the area difference between out-of-plane and in-plane curves. An average of these areas was taken to estimate K_{eff} .

3.3.2 Magneto-optic Kerr (MOKE) magnetometry

Magneto-Optic Kerr effect (MOKE) magnetometer is based on an optical technique that can be used to obtain the hysteresis loop. Instead of magnetisation in the y-axis, the loop measured by MOKE will have a Kerr rotation-related parameter (angle or intensity). MOKE was used to measure MH loops for energy harvesting studies (chapter 4). The stress-induced variations in the MH loops were measured using the MOKE magnetometry (a custom-made MOKE) [16]. The MH loop measurements by MOKE are relatively fast and do not require any sample preparation. In this technique, a linearly polarised light hits the magnetic sample, which after reflecting from the sample, acquires change in the polarisation components (the technique is depicted in figure 3.5). A linearly polarised light experiences a rotation of the polarisation plane, which is known as Kerr rotation, and a phase difference appears between the electric field components perpendicular and parallel to the plane of the incidence (plane containing the incident light and the normal to the sample surface) which is defined by Kerr ellipticity [14]. By measuring the change in the polarisation of the reflected beam, the magnetisation state of the sample can be determined. This is achieved by using a polariser through which a plane-polarised laser beam is directed onto the magnetic sample. The reflected light from the sample then passes through an analyser to determine the Kerr rotation produced by the interaction of the laser with the local magnetisation. There are three types of Kerr effects depending on the used magneto-optic geometry (that is, the direction of the magnetisation with respect to the plane of incidence). In the polar Kerr effect configuration (figure 3.5a), the magnetisation M lies perpendicular to the sample surfaces but parallel to the plane of incidence. In the case of the longitudinal Kerr effect (3.5b), M lies parallel to the sample surface and the incidence plane. In the transverse configuration (figure 3.5c), M lies parallel to the sample surfaces and perpendicular to the plane of incidence.

Figure 3.5. (a) The schematics of the MOKE measurement. Various configurations for the magneto-optic Kerr effect, (b) polar, (c) longitudinal, and (d) transverse.

3.3.3 Alternating gradient field magnetometry

Alternating Gradient Field magnetometry (AGM) is a similar technique to VSM. But in AGM, an alternating field is exerted on the sample, which causes a force in the sample proportional to the applied gradient field and magnetic moment of the sample. The force is measured by the piezoelectric transducers at the tip of the probe. The detected force is converted to magnetic moment using calibration with standard samples. In this thesis, Micromag 2900 AGM was used to measure the MH loops and the first order reversal curves (FORC) of skyrmion samples (studied in chapter 6). Calibration of the system was done with a Ni sphere prior to measuring any sample. AGM has a noise level below 10 nemu even at high speed of measurements as compared to the VSM. Hence, the FORC measurements using AGM were comparatively less time-consuming. A script consisting of multiple field segments was used to perform FORC measurements. The sample was saturated at the high field before running every segment. In FORC, a set of measurements are acquired with a reversal field of H_r with regular intervals of field, leading to an irreversibility $\rho(H, H_r)$. ρ can be defined as[17-19]:

$$\rho(H, H_r) = \frac{-1}{2} \frac{\partial^2 M(H, H_r)}{\partial H \partial H_r}$$

Changed colour in the plot shows a finite value of ρ indicating an irreversible magnetic switching [20-22]. FORC contours were plotted using a python script. The irreversibility $\rho(H_r, H)$ was calculated using a second order polynomial surface fit [17]:

$$M(H, H_r) = a_1 + a_2 H_r + a_3 H + a_4 H_r^2 + a_5 H^2 + a_6 H_r H$$

In the above case, the magnitude of irreversibility can be determined from a_6 . The number of magnetisation points used for each fit is given by $(2 \times \text{smoothing factor} + 1)^2$ [20, 22, 23].

3.4 Imaging of magnetic samples

In this work, magnetic textures were captured using Bitter pattern and magnetic force microscopy (MFM). The size of imaged magnetic textures varies from a few micrometres and in sub-10 nanometre range.

3.4.1 Bitter pattern imaging

Bitter pattern imaging (BPI) is one of the oldest domain imaging techniques. In BPI, a colloidal suspension solution of magnetic particles is dropped on the surface of the magnetic thin film. The dropped solution particles agglomerate in regions with the sizeable stray field. The agglomeration of colloidal particles occurs typically on the domain wall. The highlighted patterns of the domain wall can be seen via an optical microscope. Figure 3.6 shows an example of a BPI of domains observed in CoNi/CoFe/CoNi microwire [11, 12].

The BPI technique solely depends on the stray field of the imaged sample. It can provide information about the shape and size of domains. However, BPI does not provide any accurate information about the direction of magnetisation or magnitude of magnetisation. Moreover, once the sample is imaged, it cannot be used for any structural characterisation. Nevertheless, BPI can provide quick information about the patterns of domains.

Figure 3.6. Bitter pattern image of microwires imaged by microscope. The magnetic domain walls are highlighted with dark edges [12].

3.4.2 Magnetic force microscopy

Magnetic force microscopy (MFM) is an imaging technique for magnetic textures based on scanning probe microscopy. In MFM, a tip with a magnetic coating is used to scan over the sample surface. Based on the stray field coming from magnetic samples, the tip receives a variation in the resonance, which is tracked through a laser. The vertical component of the force between the magnetic tip and sample is measured. The magnetic field interactions between the tip and the sample cause change in the magnetic force F which can be defined in proportion to integration over the total magnetic volume of the tip [24]:

$$F_z = \frac{\delta}{\delta z} \int dV \mu_0 (\vec{m}_{tip} \cdot \vec{H})$$

where H corresponds to the stray field coming from the sample.

The motion induced by stray field of magnetic sample in the cantilever can be written as a motion for a damped harmonic oscillator [25]:

$$m \frac{d^2 z}{dt^2} + \Gamma \frac{dz}{dt} + k(z - z_0) = F_{ext}(t) + F_z$$

where Γ is the damping coefficient, $F_{ext}(t)$ is the sinusoidal driving force, m is the mass of the cantilever. z is the distance between the magnetic surface and the magnetic tip. The last term

of the equation, spring constant k , can be replaced and redefined as $k_{effSpring} = k_{Spring} - dF_z/dz$:

$$m \frac{d^2 z}{dt^2} + \Gamma \frac{dz}{dt} + k_{effSpring}(z - z_0) = F_{ext}(t) + F_z(z = z_0)$$

Cantilever resonant frequency in the presence of stray field force can be written by:

$$f' = \frac{1}{2\pi} \sqrt{\frac{k_{eff}}{m}}$$

In limit where $\frac{dF_z}{dz} \ll k$, the right-hand side of the above equation can be simply written as:

$$\frac{\delta F_z}{\delta z} = \frac{2k}{f_0} \Delta f, \text{ where } \Delta f \equiv f' - f_0$$

where f_0 is the natural resonant frequency of the cantilever.

The minimum detectable force gradient, which can be detected by thermally limited measurement, can be defined by [24, 25] :

$$\frac{dF}{dz_{min}} = \frac{1}{Amp.} \sqrt{\frac{4kK_b T \omega'}{\omega_0 Q}}$$

$Amp.$ is the amplitude of oscillation of cantilever, ω' is the measurement bandwidth ($\omega' \equiv 2\pi f'$), ω_0 is natural bandwidth, which can be defined as $\omega_0 \equiv 2\pi f_0$, K_b is Boltzmann constant, T is the temperature in Kelvin, and Q is the quality factor of the cantilever.

For the study, Bruker's Dimension atomic force microscope was used. The SSS-MFM magnetic tips were bought from NANOSENSORS. The resonance frequency of the tips was ~ 80 kHz. The imaging of the samples was carried out diligently. The tip was magnetically saturated with a bar magnet before use. Similarly, the samples were saturated with the same pole of the magnet before every measurement. The measurement starts with the remanent ($H=0$) state (after removing the magnet) at first and subsequently at higher field (H) values.

3.5 Micromagnetic simulations

In this thesis, several investigations were performed using micromagnetic simulations to understand the behaviour of magnetisation, current-induced dynamics of domain walls, skyrmions, and to estimate the exchange coupling energy density. For micromagnetic simulations, mumax3 software was used. Mumax3 is a graphical processing unit (GPU) based accelerated simulation tool [26]. Mumax3 uses a finite difference discretisation method to simulate geometries with different dimensional areas. In mumax3, the magnetisation of given geometries with a defined set of magnetic parameters is brought to a low energy state using Landau-Lifshitz Gilbert (LLG) equation [27-30],

$$\frac{\partial \vec{m}}{\partial t} = -\gamma_0 \vec{m} \times \vec{H}_{eff} + \alpha (\vec{m} \times \frac{\partial \vec{m}}{\partial t}) + \gamma_0 H_{SO} (\vec{m} \times (\vec{\sigma} \times \vec{m}))$$

where $\vec{m}$ corresponds to the unit vector of local magnetisation, $\gamma_0 = \mu_0 |\gamma|$ in which γ is gyromagnetic ratio, H_{eff} effective magnetic field derived from the free energy density: $\vec{H}_{eff} = -\frac{1}{\mu_0 M_s} (\partial E / \partial \vec{m})$, which includes the effective field from exchange, magnetic anisotropy, demagnetisation, and DMI; α is the Gilbert damping. H_{SO} is the effective magnetic field due to SOT and is given by $H_{SO} = \hbar \theta_{SH} j_c / 2e M_s L_z$, where θ_{SH} is spin Hall angle, $\hbar$ is reduced Plank's constant, j_c is applied charge current density, L_z is thickness of magnetic film and $\vec{\sigma} = \vec{j}_c \times \vec{z}$, $\vec{j}_c$ and $\vec{z}$ are unit vectors in the charge current and out of plane.

To include finite-T in the LLG equation, an additional random effective field term that scales with the temperature is added to the effective field as [31]: $\vec{B}_{th} = \vec{n} (step) \sqrt{\frac{2\mu_0 \alpha k_B T}{B_{sat} \gamma \Delta V \Delta t}}$

k_B the Boltzmann constant, T the temperature, B_{sat} the saturation magnetisation expressed in Tesla, ΔV the cell volume, Δt the time step and $\vec{n} (step)$ a random vector from a standard normal distribution whose value is changed after every time step.

Before initialising the simulations, the size of the cell and the geometry is required. The cell size, as a rule of thumb, needs to be smaller than the magnetostatic exchange length as $l_{ex} = \sqrt{2A_{ex}/\mu_0 M_s^2}$, which usually is less than equal to 5 nm. Simulations used for domain wall and skyrmion dynamics (chapter four and five) were performed at 0 K, and the relaxation procedure follows the Dormand-Prince method (a member of the Runge-Kutta family) that uses six function evaluations per step of minimising the energy to resolve the LLG equation. In the case of estimating exchange coupling energy (chapter 6), the geometries of thin films were defined with periodic boundary conditions in the x and y direction, which mimics the stray field from a larger size thin film. The simulations were performed at 300 K. An effective medium model was used, replacing the trilayer repetition stack with an effective medium layer (shown in figure 3.7).

Figure 3.7. Schematics of used model for simulations. The thickness of one ferromagnet, heavy-metal, and oxide layer is replaced by an effective medium ferromagnetic layer, with a thickness of one repetition layer.

The magnetic properties of the simulated model were reduced by a factor λ for the effective medium model as following $\lambda = \frac{t_{fm}}{t_{rep}}$. The magnetic properties of an effective ferromagnetic

layer can be used with relation to actual magnetic properties as [32]:

$$\lambda = \frac{t_{fm}}{t_{rep}} = \frac{M_{seff}}{M_s} = \frac{A_{exeff}}{A_{ex}} = \frac{DMI_{eff}}{DMI} = \frac{K_{effeff}}{K_{eff}}$$

All the other quantities remained unchanged.

For minimising the energy of the system, Heun's method uses uniform steps that can deal with the discontinuous torques given by the random field model. A fixed-step time size was given with time = 1.5×10^{-13} s. The simulations were run for different time values (varying from 40 ns to 130 ns) to observe the difference in the final state. The error in the simulated last step was calculated to observe the final relaxed state. An error less than the order of 10^{-3} at 300 K was considered to be stable.

3.6 Application of stress on microwires for energy harvesting

Two different methods were used to apply stress to the samples studied in chapter 4.

Figure 3.8. Illustration of the method used to apply stress for observing the bitter pattern. The length of the Kapton membrane strip was constant throughout the whole experiment. δ was increased to increase the stress on the sample[11].

The formula used in this article to calculate stress $\sigma^{f'}$ is given by

$$\sigma^{f'} = \frac{E_s h_s^2 \delta}{3(1-\nu_s) h_f l^2}$$

where E_s (taken here as 2.5 e^9) is young's modulus of the substrate (Kapton membrane), h_s is the thickness of the substrate ($85 \text{ } \mu\text{m}$), δ is the lifted height from the free side of the strip, ν_s is Poisson's ratio (0.3), h_f is the thickness of the film (30 nm), and l is the length of the strip (2.03 cm). The stress value was increased by increasing the δ to observe the respective effect.

The experiment was designed to introduce non-uniform stress to the FeCo films, which imitates the stress-induced from ambient conditions.

Figure 3.9. Picture of curved surfaces (concave and convex) used to apply stress. The curved structures had different radii at different parts [11].

We used curved surfaces with radii of curvature of 16 mm and 18 mm for our experiment. The samples deposited on the Kapton membrane were pasted on these curved surfaces to induce the external stress during the measurement of hysteresis loops using micro-MOKE [11].

Calculation of stress:

The calculation of stress σ^f was carried out using Stoney's formula [33], which states as:

$$\sigma^f = \frac{E_s h_s^2}{6(1-\nu_s)h_f R}$$

where E_s (taken here as 2.5×10^9) is young's modulus of a substrate (Kapton membrane), h_s is the thickness of the substrate (85 μm), ν_s is Poisson's ratio (0.3), h_f is the thickness of the film (40 nm), and R is the radius of curvature (16 or 18 mm). The calculated stress value on the films for 18 mm and 16 mm are 5.9 GPa and 6.7 GPa, respectively.

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Chapter 4 Harvesting of Energy induced by Stress caused modification of Domain Walls

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4.1 Introduction

An IoT device typically consists of a processing unit, a sensor to observe the change or record the variable in data, a wireless connection to share data, and an energy source to power the device [1]. The energy source is vital for the IoT device, as continuous power is needed to reliably record the data and transfer the data to a database through a wireless internet connection. The conventional batteries, which need to be changed regularly, are not appropriate due to the inconvenience of changing the battery regularly. In addition, incapability to change the batteries can result in the loss of essential data. Therefore, IoT needs power sources that can self-harvest the energy from the ambient sources. The other necessary requirement of the power source for IoT devices is the small size (micrometer - nanometer) of the generator. There has been much research done to harvest energy from the ambience [2-6]. For such self-harvesting devices, the approach based on harvesting by solar cells seems obvious. Although solar cells hold many potentials, this approach confronts the failure to generate energy in areas lacking a light source. The second approach is based on the application of piezoelectric materials to harvest voltage. The use of piezoelectric-based sensors can be profoundly found in today's world. The piezoelectric generators hold the potential to provide high voltage. However, the piezoelectric generators suffer from low power generation due to the high internal impedance of piezoelectric materials. Moreover, piezoelectric materials like PZT and BaTiO₃ are ceramics and brittle. Continuous stressing and relaxing can decrease the longevity of device [7-12]

Other approaches based on ZnO nanorods have been researched intensively. However, ZnO-based approaches are not viable for nanogenerator applications. ZnO has a low piezoelectric constant, which implies that considerable stress needs to be applied to generate a high voltage [13, 14]. Secondly, a few theoretical models suggest the asymmetrical distribution of potential on a bent nanorod under applied stress. In addition, the synthesis of ZnO is done by the

chemical route and synthesizing a large amount of ZnO in a lab is not extendable for the mass production of devices [7, 15]. Hence, researchers are looking for other approaches which can be used to harvest voltage.

Researchers have explored approaches based on magnetic materials in the quest for new approaches for nanogenerator applications. A traditional method of generating electricity from magnetic materials uses Faraday's law, wherein a time-varying flux produces a voltage in the pick-up coil. Typically, the magnet or the coil is in relative motion to achieve a time-varying flux. While in IoT devices, this principle can be employed by using ambient vibrations. We have proposed a novel method of harvesting energy without a relative motion of coil and magnetic material.

In the field of magnetism, domain wall motion has been explored for various applications. Out of these applications, memory based on domain wall motion is the one that gained much interest from the magnetic society, in which the domains are used to store information and move in a track to read the stored information [16-19]. However, this domain wall motion can also be explored to harvest energy. So far, the motion of domain walls for harvesting energy has been reported for multiferroic structures, where a current is sent to the ferroelectric layer to generate strain on the magnetostrictive layer [20, 21]. However, employing a significant amount of electrical energy to generate voltage compromises the purpose of self-energy harvesting. In addition, ceramic ferroelectric layers, like PZT, require high-temperature deposition or post-deposition annealing ($>400\text{ }^{\circ}\text{C}$) under severe oxidation conditions, which inevitably degrade the magnetic properties of magnetostrictive material [22]. Such high temperatures also prevent the mass fabrication of IoT devices. Therefore, there is a need to explore a magnetism-based approach, which relies on the ambient source to induce stress on the magnetostrictive material.

Applying stress to a magnetostrictive material can lead to a change in the magnetisation direction. The induced change in the magnetisation results in a variation of magnetic flux. This change in the magnetic flux can be harvested into voltage using pick-up coils in the vicinity of the material.

In this chapter, I demonstrated the harvesting of energy using crystalline Fe₆₅Co₃₅ microwires. The chosen alloy composition of FeCo exhibits a high saturation magnetisation (which results in a higher flux) and reasonably high magnetostriction constant of 5.8×10^{-5} (which leads to a significant change in magnetisation for small values of stress) [23-27]. Studies have been carried out on the magnetic properties of FeCo thin films under stress. For harvesting energy, microwires were fabricated with magnetic anisotropy in the transverse direction to the microwire length. Magnetic anisotropy in the transverse direction to the length of the microwire results in the formation of multiple domains. Stress-regulated modification of domains has been observed in microwires. Eventually, a prototype device was fabricated, and we have demonstrated the production of energy by vibrating the sample through a piezoelectric stage.

4.2 Achieving in-plane magnetic anisotropy

Deposition of Fe₆₅Co₃₅(30 nm) thin films was carried out on a SiO_x substrate using a facing targets sputtering tool. Magnetic fringing fields from the sputtering guns can be used to set the direction of magnetic anisotropy during the deposition process. Magnetic properties of deposited FeCo films were measured using a Kerr magnetometer. Figure 4.1 shows the magnetic properties measured from the magnetic hysteresis (MH) loop of deposited FeCo film. Figure 4.1(b) shows the measured MH loops at 0° and 90° (shown in the schematics). The measured MH loops have identical properties with coercivity $H_c \sim 7$ to 8 mT. Hence, there is no magnetic anisotropy observed in the deposited FeCo films. Moreover, the magnetic anisotropy field $H_k \sim 9$ mT. Consequently, we hypothesized that the high coercivity of the

samples (which is similar to H_k) is the reason for not being able to observe a magnetic anisotropy.

Figure 4.1. (a) Schematics of magnetic field direction applied to FeCo films. 0° represents the direction of the easy axis set by the fringing magnetic field during deposition, and 90° refers to the direction transverse to the easy axis, both in the film plane. (b) Hysteresis loops of the single-layered FeCo. The loop does not exhibit in-plane magnetic anisotropy [28].

To introduce magnetic anisotropy in the FeCo films, we need to reduce the coercivity of deposited films to be considerably smaller than H_K . For thin films, coercivity is proportional to the grain size D_{grain} , as mentioned in the following equation [29]:

$$H_c \propto K_{eff} \approx K \left[\frac{K^4 D_{grain}^6}{A_{ex}^3} \right]$$

where A is the exchange energy, K is the anisotropy constant, and K_{eff} is the effective anisotropy. As explained by the random anisotropy model [29], effective anisotropy (also H_c) of material reduces when the grain size becomes smaller than the magnetostatic exchange length. Therefore, by reducing the grain size, one can decrease the value of H_c . To reduce the grain size, FeCo was deposited on different underlayers. We speculated that the lattice mismatching between the underlayer and FeCo would reduce grain size. Therefore, different underlayers were tried viz. Ta, Ti, Cu, Co, and Ni. Figure 4.2 shows the scanning electron microscope (SEM) images of the FeCo layer on various underlayers, depicting FeCo grains.

Figure 4.2. Scanning Electron Microscopy (SEM) images of FeCo layer deposited on various underlayers. (a) FeCo deposited on substrate (b) FeCo deposited on Ta. (c) FeCo deposited on Ti. Grain size of FeCo become larger after depositing FeCo layer on Ta and Ti. (d) FeCo deposited on Cu. (e) FeCo deposited on Co. (f) FeCo deposited on Ni. The grain size of FeCo become smaller after deposition on Cu, Co, and Ni. The thickness of FeCo and different underlayers was kept constant to 30 nm and 3 nm, respectively [28].

From figure 4.2, we measured the grain size of FeCo thin films deposited on different underlayers. The average grain size of FeCo was increased from 40 nm (without underlayer) to 50 nm for Ta and Ti underlayers, respectively. On the other hand, the average grain size of FeCo film deposited without any underlayer was ~40 nm, which became smaller when FeCo was deposited on Cu (~18 nm), Co (~13 nm), and Ni as underlayers (~16 nm) [Figure 4.2d–f, respectively]. From these observations, we chose Co₅₀Ni₅₀ (henceforth referred to as CoNi) as the underlayer to achieve a reduced grain size of FeCo.

Figure 4.3. (a) Schematics of deposited trilayer stack. A 2 nm CoNi layer was deposited on the top as a capping layer. (b) MH loop of FeCo after depositing it on CoNi. The coercivity of FeCo was reduced, and a magnetic anisotropy was introduced after the reduction in the grain size [28].

After depositing FeCo on top of 3 nm CoNi, a 2 nm capping layer of CoNi was deposited on top of FeCo. Deposition of the capping layer was done to avoid any possible corrosion. Figure 4.3a shows the schematic of the deposited trilayer. MH loops of deposited trilayer are shown in figure 4.3b. The loops were measured by applying a magnetic field in the easy axis direction (pink colour) and hard axis direction (purple colour). Coercivity along the easy axis of the trilayer was reduced to ~ 2 mT while the magnetic anisotropy field remained the same, around 9 mT. Moreover, we have noticed that the shape of loops is different in a different direction of the swept magnetic field. Hence, it is clear that the reduction of coercivity has led to the observation of magnetic anisotropy in the deposited film. One may argue that the seed layer (CoNi) itself has a magnetic anisotropy, and hence, the magnetic anisotropy of the seed layer induced anisotropy in FeCo film. To check the influence of CoNi on the achieved magnetic anisotropy of FeCo, a 3 nm CoNi layer was deposited on top of SiOx substrate. Figure 4.4 shows that MH loops of the CoNi layer. The loops are identical to each other, indicating no magnetic anisotropy observed in the 3nm CoNi underlayer. Therefore, we can confirm that the

magnetic properties of the bottom CoNi layer are not responsible for inducing the magnetic anisotropy in the FeCo layer.

Figure 4.4. MH loops of 3 nm CoNi layer deposited on SiOx. Magnetic loop plotted in pink color represents loop measured when magnetic field is applied in 0° direction and purple color represents the loop measured when magnetic field is applied in 90° direction (as shown in inset) [28, 30].

Since in the trilayer stack, FeCo was sandwiched between CoNi layers, there was a possibility to observe an exchange bias. However, MH loops of the trilayer stack (from figure 4.3) do not show any shift in the loop. Therefore, it can be inferred that the trilayer stack does not depict any effective exchange bias.

Further investigation on the microstructure of deposited layers was carried out by using X-rays diffraction (XRD). For XRD, CoNi and FeCo layers of 30 nm thickness were deposited on the substrate. The XRD patterns of different films are shown in figure 4.5. CoNi XRD depicts a diffraction peak at 2θ of 44°. This peak corresponds to the (111) face-centred cubic (fcc) phase of CoNi [31, 32]. For the FeCo layer, a diffraction peak of (002) for the body-centred cubic (bcc) phase of FeCo has been observed at 2θ of 65° [33, 34]. The trilayer CoNi(3 nm)/FeCo(30

nm)/CoNi(3 nm), shows a dominant peak of fcc(111) which has been shifted by 0.7° to the right. Noticeably, the FeCo structure has been transformed from bcc to fcc after depositing FeCo on CoNi. Hence, the XRD pattern in figure 4.5 depicts a change of resultant phase with the deposition of CoNi as an underlayer. We consider this change in crystallographic texture an essential contributor, along with the fringing field, for the induced anisotropy in the trilayer stack. This magnetic anisotropy, exhibited by the trilayer stack, plays a vital role in ensuring a considerable change in the magnetic properties of FeCo stack under external stress.

Figure 4.5. Crystallographic study of the CoNi, FeCo, and trilayer films exhibiting bcc (200) peak for FeCo, fcc(111) peak for CoNi, and the trilayer stack [28, 30].

4.3 Stress regulated properties of thin films

To study the effect of stress, trilayer thin-film stack was deposited on an $85\ \mu\text{m}$ thick flexible polyimide membrane (Kapton). The use of a flexible polyimide membrane facilitates the transfer of stress to the trilayer stack. Stress was applied by pasting the samples grown on the flexible membrane to curved surfaces. Concave or convex curved surfaces with radii of curvature of 16 and 18 mm were used to measure the effect of stress under different compressive and tensile stresses, respectively. Figure 4.6 shows the schematics of the curved surface used for applying stress.

The calculation of stress σ^f is done by using Stoney's formula [35], which states as:

$$\sigma^f = \frac{E_s h_s^2}{6(1-\nu_s)h_f R}$$

where E_s (taken here as 2.5 E^9) is young's modulus of the substrate (Kapton membrane), h_s is the thickness of the substrate ($85 \text{ }\mu\text{m}$), ν_s is poison's ratio (0.3), h_f is the thickness of the film (40 nm), and R is the radius of curvature (16 or 18 mm). The calculated value of stress on the films for 18 mm and 16 mm are 5.9 GPa and 6.7 GPa , respectively.

Figure 4.6. Schematics of curved surface used for applying stress on thin films. The radius of curvature for curved surface is 16 mm and 18 mm .

To observe the effect of externally applied stress, we deposited the trilayer stack (FeCo 30 nm) on the Kapton membrane and used the above method to observe Bitter patterns. We increased the value R (radius of curvature) to its maximum with the available apparatus and were unable to observe any change in the domain walls. Therefore, we deposited the trilayer stack of the same thickness on coverslip glass of 0.20 mm thickness. The Young's modulus value of the used coverslip glass is 7.29×10^{10} , which increased the value of applied stress on the trilayer stack by almost 29 times for the exact value of R . The resultant effect of changing the substrate is shown in figures 4.7b and c. However, the use of such a substrate is not appropriate for the application of energy harvesting from ambient sources.

Figure 4.7. Bitter Pattern of sandwiched FeCo thin film (30nm) on a cover slip glass. The insets show a zoomed-in view of the domains (a) shows the bitter pattern of the thin film under no stress. The magnetisation of domains in these are head-to-head. (b) and (c) are the bitter pattern under applied external compressive stress. (b) Deterioration of domain walls can be observed due to rotation in the magnetisation. (c) Bitter pattern when the applied stress is increased. The diminishing of the domain walls can be noticed to increase, forming small elliptical shape domains.

Tensile stress was applied to FeCo films, deposited on polyimide membrane. Similar to the effect of compressive stress on polyimide membrane, no change in magnetic domains were observed when the sample was pasted to the curved surface. Next, FeCo film with cover glass substrate was pasted onto the curved surface. Figure 4.8 shows the effect of tensile stress on domain walls. Unlike the compressive stress, domain walls did not diminish for tensile stress. At high-stress values, the domains rotated. We were not able to observe any further change in the magnetic domains on higher stress values.

From the observations that have been made so far, we speculated that the difference in the patterns of FeCo under compressive stress is due to the changes in the easy axis of the anisotropy directions. We measured hysteresis loops of trilayer thin-film stack under compressive and tensile stress using a MOKE magnetometer to confirm our speculations. In both cases, we measured the loops along the easy axis (direction set by the sputtering process) and the hard axis (shown by pink and purple arrows, respectively, in figure 4.9a and d).

Stress = 0

Stress = 58×10^9 Pa

Stress = 117×10^9 Pa

Figure 4.8. Bitter Pattern of trilayer FeCo thin film (30nm) on a cover slip glass. (a) Bitter pattern of the thin film under no stress. The magnetisation of domain in these are head-to-head. (b) and (c) are the bitter pattern under applied external tensile stress. No change in magnetic domain was observed for 58 GPa. For high stress magnetic domains were rotated.

Figures 4.9b and c show the MH loops of FeCo sandwiched stack for easy and hard axis under zero stress, where the FeCo layer was 40 nm thick. These loops resemble those plotted in figure 4.3 and validate the effectiveness of the MOKE magnetometer in measuring these properties. Stress was applied along the hard axis to study the effect of stress on the magnetisation curves. Figures 4.9c and f show the hysteresis loops of the FeCo stack under externally applied stress. An apparent change in the magnetisation direction from the easy axis (set by fringing field) to the hard axis was observed. We speculate the change in magnetic anisotropy is due to the change in the direction of magnetic anisotropy. The change in magnetisation direction resulted in a rotation of magnetisation in the FeCo stack under externally applied stress. These results indicate that the applied stress changes the anisotropy direction, leading to a change in magnetisation direction resulting in the rotation of domain walls, leading to a change in the magnetic flux.

Figure 4.9. Hysteresis loops of trilayer films measured using a MOKE. (a),(d) Color code for direction of the measured MOKE MH loops for zero stress and under stress. Pink represents the easy direction set by the magnetic field and purple, the transverse direction. (b),(c) Hysteresis loops of trilayer stack along easy axis and hard axis, respectively, without the application of any stress. (e),(f) Hysteresis loops of trilayer along easy axis and hard axis, respectively, under compressive stress. The bidirectional arrows in the plots represent the direction of applied external magnetic field [28].

We measured MH loops of the trilayer stack were measured under different types and values of applied stress using a micro-MOKE. Figure 4.10 shows the change in the hysteresis loops of the FeCo stack under compressive and tensile stress, along with their magnitudes. The changes in the anisotropy directions with compressed stress are clear from this figure. It must be noticed that the change in the direction of anisotropy results in a change in the coercivity of material. The extent of the change in coercivity varies with the value of applied external stress [36]. In the case of tensile stress, for unknown reasons, the easy axis remained unchanged. We did not see any significant changes in the coercivity for externally applied tensile stress. This result is in contrast with a drastic change reported in stress-annealed a-Fe₆₉B₁₂Si₁₄C₅ [37]. In compressive stress, the easy axis set by the sputtering process became the hard axis, and hence, the loops exhibited a lower remanence. A field of more than 100 Oe was required to bring the magnetisation along the applied field direction. Similar results exhibiting the interchange between the hard and easy axes are reported in some previous studies[37, 38].

Figure 4.10. Hysteresis loops of the sandwiched stack under different stresses, T: tensile stress, C: compressive stress and for different radii of curvature (16 and 18 mm). Stress was applied in the magnetic hard axis direction of the samples [28].

4.4 Micromagnetic simulations on study of stress in microwires

Micromagnetic simulations were carried out using MuMax3, which solves the Landau–Lifshitz–Gilbert (LLG) equation to minimize the system energy to its lowest [39]. One dimensional geometry (wire shape) was used to apply stress on FeCo for harvesting energy. The advantage of using a wire shape is that it can be easily wrapped with a pick-up coil, and such shape can be fabricated on a long flexible substrate that can pick vibrations from ambient sources. Hence, the simulation was carried out on microwires with $4500\text{ nm} \times 300\text{ nm} \times 30\text{ nm}$. The cell size was set as $3\text{ nm} \times 3\text{ nm} \times 3\text{ nm}$, which is less than the reported exchange coupling length [40]. Simulations parameters were chosen based on the material properties of $\text{Fe}_{65}\text{Co}_{35}$. The exchange constant and saturation magnetisation were fixed at 23 pJ/m and 1950 kA/m , respectively. The direction of easy-axis for uniaxial magnetic anisotropy was along the y-direction (0,1,0). The literature value of magnetic anisotropy for such material is 2.75 kJ/m^3 [28]. This value of anisotropy has been reported for magnetic films. It is better to have many domains in a transverse direction of the microwire to gain an advantage from the variation of flux induced by stress. Hence, we tried to introduce a higher value of magnetic anisotropy in transverse direction. In one dimensional geometry, the shape anisotropy will be prominent. Therefore, magnetic anisotropy must overcome the shape anisotropy to achieve magnetic domains in the transverse direction of the microwire.

4.4.1 Achieving magnetic domains in the transverse direction

In the absence of any magnetic anisotropy, the magnetisation of a one-dimensional microwire will form random-shape multiple domains or single domain with magnetisation along the length of wire. The number of domains will depend on the aspect ratio (ratio of length to width) of microwire (as shown in figure 4.11a). In the absence of any induced effect, the shape anisotropy will dominate in a microwire, causing the magnetisation of domains to lie along the

length of the microwire. However, if an anisotropy can be achieved along the transverse direction to the length of microwire, the magnetisation in the domains will be in the transverse direction. Therefore, at first, we carried out micromagnetic simulations to understand the role of transverse anisotropy. We started the initial condition from the random magnetisation state for these simulations, which was relaxed later to its minimum energy state. To get the magnetic domains in the transverse direction, we added the anisotropy in the transverse direction. Figures 4.11b-g show simulated images of microwire for various values of added magnetic anisotropy in the y-direction ($K_{transverse}$). It can be observed from figure 4.11 that an increase in the value of $K_{transverse}$ leads to an increase in the number of magnetic domains formed in the microwire. It must be noticed that the required value of $K_{transverse}$ to get densely-packed magnetic domains in the transverse direction will rely on the dimensions of the microwire. As the length of the microwire in comparison to the width of the microwire increases, the required $K_{transverse}$ to generate domains in the transverse direction will also increase. Figure 4.11g shows the simulated image of the microwire with magnetic domains in the transverse direction to the length of the microwire. The $K_{transverse}$ value used for this simulated condition was 100 times more than the value reported in the literature. We used this value of $K_{transverse}$ for further simulations to study the effect of stress.

Figure 4.11. Simulation images to estimate the value of magnetic anisotropy required to keep the domains transverse to the length of the wire. In-plane magnetisation map depicts the colors for different magnetisation directions. (a) Simulated image of microwire without any magnetic anisotropy. (b)-(g) Simulated images of microwires with different values of transverse magnetic anisotropy ($K_{transverse}$) in transverse direction [30].

4.4.2 Effect of stress-induced anisotropy on microwire

After achieving transverse magnetic anisotropy in microwires, micromagnetic simulations were carried out to study the effect of stress on the domain patterns. Magnetoelastic anisotropy was increased along the length of the microwire to simulate the effect of stress. The magnetoelastic anisotropy can be calculated from the relation $K_{MagnetoElastic} = 3\sigma_{stress}\lambda_{mstriction}/2$ [41, 42]. The symbols σ_{stress} and $\lambda_{mstriction}$ correspond to the applied

stress to the microwires and magnetostriction constant of $\text{Fe}_{65}\text{Co}_{35}$ (which is $\sim 5.8 \times 10^{-5}$), respectively [27].

Figure 4.12. Simulation images of microwires with different values of applied stress anisotropy (K_{stress}), which is equivalent to $K_{MagnetoElastic}$. (a) Simulated microwires with domains in the transverse direction. (b) The size of magnetic domains increases. (c) Tilt in domains when the applied stress is equivalent to 40% of transverse anisotropy. (d) Diminishing of magnetic domains at high stress. (e) Diminishing of domain walls at a high value of stress [30].

Stress was applied in the x-axis using the value of magnetic anisotropy (K_{stress}) along the x-direction (1,0,0) during simulations. The applied value of K_{stress} (which is equivalent to $K_{MagnetoElastic}$) was varied with respect to the value of the finally used $K_{transverse}$ (shown in figure 4.12). The applied value of K_{stress} was varied in the range of 27.5 kJ/m³ to 275 kJ/m³ (10% of $K_{transverse}$ to 100 % of $K_{transverse}$). Figure 4.12 shows the simulated images of domain patterns in microwires with different values of applied stress in the x-direction. Figure 4.12a exhibits the microwire without any applied value of K_{stress} (only $K_{transverse}$). The magnetic domains can be observed to be densely packed in the absence of any stress from the effect.

Figures 4.12b-e show the images of simulated microwires with different values of applied K_{stress} . Figure 4.12b shows the simulated image of K_{stress} with a 20% value of $K_{transverse}$. The size of magnetic domains was observed to increase with an increase in the value of applied stress (anisotropy). However, the domains can be observed to be tilting at a high value of applied stress (40% $K_{transverse}$).

Increasing the value of applied stress further demonstrated the motion of domain walls, resulting in larger size domains (shown in figure 4.12d). At a very high value of applied stress, all the domain walls were collapsed. Such a high value of stress changes the magnetic anisotropy direction from the y to x axis. This behaviour resulted in formation of a single domain, as shown in figure 4.12e. The outcome of the simulations showed a change in the direction of magnetic domains on applying stress due to the motion or collapse of domain walls. With such a change in the direction of magnetic domains, the magnetic flux around the microwire also changes. Such time-varying magnetic flux can be utilized to harvest energy by using a pick-up coil in the vicinity of a microwire. Therefore, we carried out the experimental investigations to achieve the results that we observed from simulations as a next step.

4.5 Observation of stress-induced effect on magnetic domains in microwires

To study the effect of applied stress on magnetic domains of microwires, we deposited the microwires of FeCo of 30 nm thickness. The length of deposited microwires was kept constant to 1 mm while the width was varied from 4 μm to 40 μm . The samples were deposited on the flexible substrate to induce stress. To observe the effect of applied external stress on the shape of magnetic domains, we used the Bitter pattern technique on the deposited microwire sample [43, 44].

Bitter pattern images in figure 4.13 show the domain structure of as-deposited FeCo microwires. The microwires were patterned using optical lithography to form trenches before depositing the magnetic stack. Later, a liftoff process was carried out to get the microwires. Figure 4.13a shows the microscope image of as-deposited microwires. The edges of the microwires were noticed to be smooth, lacking any mechanical roughness.

Figure 4.13. Microscopic images deposited microwires and Bitter-Patterns of deposited wire. (a) Microscope image of as-deposited microwires. No mechanical roughness was observed on the edges of microwires. (b) Domain walls in an as-deposited microwire show the orientation of domains transverse to the length of the wires. Roughness seen on edges of microwire is magnetic roughness arising due to Bitter pattern technique [28].

Figure 4.13b shows the Bitter patterns of as-deposited FeCo microwires. Inset of figure 4.13b shows the zoomed-in image of the Bitter pattern in a 40 μm wide microwire. The direction of magnetic domains can be seen to be set in the transverse direction to the length of the microwire.

Figure 4.14. Domain patterns under compressive stress. (a,b) Widening of domains under strain. (c) Diminishing of domain walls at 2.36 GPa applied stress. (d) Diminished domain walls in a microwire. The insets show zoomed-in views of the domains [28].

In order to apply stress on the deposited microwires, a stress applying apparatus was used (discussed in chapter 3). The formula used in this article to calculate stress $\sigma^{f'}$ is given by

$$\sigma^{f'} = \frac{E_s h_s^2 \delta}{3(1-\nu_s) h_f l^2}$$

where E_s (taken here as 2.5×10^9) is young's modulus of the substrate (Kapton membrane), h_s is the thickness of the substrate (85 μm), δ is the lifted height from the free side of the strip, ν_s is Poisson's ratio (0.3), h_f is the thickness of the film (30 nm), and l is the length of the strip (2.03 cm). The stress value was increased by increasing the δ to observe the respective effect. The experiment was designed to introduce non-uniform stress to the FeCo films, imitating the

stress induced by ambient conditions. The values of calculated stress are high due to the nanometre thickness of deposited thin films.

An expansion of domain size under externally applied compressive stress substantiates the results from simulations (shown in figure 4.14a). When higher stress was applied, the domain walls started diminishing (Figure 4.14c), similar to the observations in simulations. The diminished domain wall enhances further with higher stress, resulting into total diminishing of domain walls (figure 4.14d). Interestingly, the results are very similar to the simulated results (figure 4.12e). Unlike simulations, we did not observe any tilting of domains experimentally. In the experiments, the applied stress to the microwires during imaging was static and varied in steps. The step size cannot be reduced. Therefore, we speculated that the tilting of domains might have occurred in-between two values of applied stress.

In figure 4.15, the measured values of the average width of domains (for 25 μm wide microwire) as a function of externally applied compressive stress. Noticeably, the domain width increased with applied stress until about 2 GPa. From a stress of about 2.36 GPa, the domain walls started diminishing. This observation made the domain width immeasurable for high-stress values. We speculated the diminishing of domain walls with the increase of externally applied stress is due to the change in direction and strength of the magnetic anisotropy under the applied stress. This stress-induced anisotropy vanished when the applied stress was removed and the domains appeared. These observations indicate that the bending and unbending of the sample due to ambient vibration sources will result in domain wall motion. The resultant change in magnetic flux due to this domain wall motion can be converted into electrical voltage using pick-up coils. Devices based on this phenomenon can be built to harvest energy from ambience.

Figure 4.15. Plot of magnetic domain width varying as a function of applied stress [28].

As the next step, a comparison of magnetic domain wall width achieved from simulations and experiments was carried out. Figure 4.16 shows the trend of magnetic domain width. The length of magnetic microwires differed in simulations (4500 nm) and experimental (1 mm). Due to the difference in length of microwire, the width of magnetic domains at zero stress is different in simulations and experiments. The x-axis of the plot refers to the applied stress in terms of magnetic anisotropy. In simulations, the drop in width of domains at 40% refers to the tilt in magnetic domains observed, missing in experiments.

Interestingly, at 40% stress, the domain width did not change. At high-stress values, the increase in the domain width was caused due to diminishing of domain walls, resulting in submerging of adjacent domains. Conclusively, the trend observed in experiments and simulations is similar.

Figure 4.16. Comparison of magnetic domain width as a function of applied stress achieved from simulations and experiment [30].

4.6 Harvesting energy from stress application

The applied stress in the experiment mentioned in the previous section was static, resulting in a change in the direction of magnetic domains. The change in the directions of magnetic domains results due to conversion of the easy axis to the hard axis and vice versa under application of stress and removal of stress, respectively. Due to this conversion, the magnetic domains change their direction, resulting in the variation of magnetic flux [36-38, 45, 46]. Based on this phenomenon, the oscillating applied stress will result in an oscillating flux (time-varying flux). The time-varying flux can be harvested into voltage by using a pick-up coil in the vicinity of the sample, as defined by Faraday's law.

In order to show that the proposed method can be used to harvest energy, we investigated the energy harvesting performance of trilayer stack microwires deposited on the flexible substrate. To imitate the stress delivered by the ambient vibration from the environment, a piezoelectric stage was used to introduce vibrations. A waveform generator was used to introduce a voltage

pulse to the piezoelectric stage. A pulse with a 3.7 kHz frequency was used to produce vibrations. The waveform produced by the pick-up coil is shown in figure 4.17a. A sinusoidal waveform with a peak voltage of mV in a 50 Ω load was observed, which clearly shows the promise of such a device for energy harvesting applications. Figure 4.17b shows the energy harvested during a time frame of 350 μ s during this experiment. The total energy generated (with 50 Ω as the resistive load of the circuit) during this short time-scale was in the nJ scale. In energy harvesting devices based on this mechanism, the flux produced will be proportional to the $M_r t$ of the device, where M_r denotes the remanent magnetisation, and t denotes the thickness of the film. Therefore, in order to increase the output signal, it is necessary to increase the thickness. To study this aspect in detail, we carried out a study on FeCo thickness-dependent energy harvesting. We deposited trilayer stack thin films of 1 cm $\times$ 1 cm with different thicknesses of FeCo. Figure 4.17c shows the hysteresis of two film stacks with extreme thickness values of FeCo thin film used in this study, indicating that the required anisotropy is achieved at the studied thickness values. We mimicked the ambient vibration from the environment by using a piezoelectric stage to vibrate the samples at 3.7 kHz for this study. The samples were placed in the vicinity of a pick-up coil. The energy from stacks of different thicknesses, for an average time span of 50 μ s, has been shown in figure 4.17d. A linear increase in the harvested energy with the $M_r t$ was also observed. For this study, the thickness of FeCo film is only 50 nm and the microwires are not packed closely. Increasing the density of the fabricated microwires and the thickness of the films to an optimum value is expected to increase the energy to potentially higher values. Consequently, the reported results are useful in certain devices where solar panels cannot be used to harvest energy.

Figure 4.17. Harvesting energy from trilayer microwire. (a) Plot of waveforms obtained from the device over a duration of 50 μ s. (b) Plot of harvested energy from the trilayer stack microwires in the time span of 350 μ s (c) Hysteresis loop of the trilayer stack with different thicknesses of FeCo layer. (d) $M_r t$ of trilayer stack of different thicknesses and the total energy harvested in the time span of 50 μ s [28].

The tri-layer stack (CoNi/FeCo/CoNi) used for this investigation did not show any structure damages during the stress studies of the stack. However, the real device will go through ambient stress (of high frequency but of low amplitude) for a long time in its lifetime, which may introduce some minor fractures in the structure of the film. Such fractures will result in minor inhomogeneities in the shape of magnetic domains and hence, reducing a change in the magnetic flux ($\Delta\phi$). To systematically understand the effect of stress on the mechanical properties of FeCo thin film, thorough structural investigations need to be carried out where the device (or thin film) has already been undergone stress for a long time.

4.7 Summary

In conclusion, demonstration of energy harvesting by the movement of domain walls in magnetic microwires entirely by stress has been carried out. For this purpose, a trilayer magnetic stack was fabricated that exhibits stress-dependent magnetic easy axis direction. The easy axis was made to lie along the transverse direction of the microwires by using a fringing field from the magnetrons. A change in the microwire magnetisation axis from hard axis to easy axis was noticed by applying stress. As a result, domain wall motion or collapse of domain walls occurs entirely by stress-induced bending of the substrate, which can be harvested to energy. A comparison of our investigated method and different techniques-based energy harvesting devices has been shown in table 4.1.

Table 4.1. Performance comparison of energy harvesting devices [47].

	Energy Source	Characteristics	Relying Technology	Harvested Power
	Solar	Indoor Outdoor	Photovoltaic	10 $\mu\text{W}/\text{cm}^2$ 10 mW/cm^2
	Vibration Or Motion	Human	Piezoelectric Electrostatic	4 $\mu\text{W}/\text{cm}^2$
		Industry	Piezoelectric Electrostatic	100 $\mu\text{W}/\text{cm}^2$
	Thermal	Human Industry	Thermoelectric	30 $\mu\text{W}/\text{cm}^2$ 1-10 mW/cm^2
	Radio Frequency	WiFi	Electromagnetic Field	0.1 $\mu\text{W}/\text{cm}^2$
Investigated Method	Ambient Vibration	Indoor and Outdoor	Inverse Magnetostriction	200 $\mu\text{W}/\text{cm}^2$

Moreover, no damage to the samples was observed due to the induced stress. Furthermore, a demonstration of energy generation in the order of μJ was done. It is to be noted that in the above experiment, only a single microwire in the preliminary device to harvest energy. Further, by fabricating more microwires at an optimized distance, avoiding the magnetostatic interactions, a device with high-density microwires can be fabricated. Hence, in the future, by optimizing the layer stack, thickness, density of the microwires, and the circuitry to generate voltage, further gain in power density can be made. This provides an opportunity for more scientific investigations and potential use as a power source in IoT applications.

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Chapter 5 Exploring Applications of Energy Confined motion of Domain Walls and Skyrmion in 1-D Devices via Micromagnetic Simulations

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5.1 Introduction

This chapter explains the detailed results of micromagnetic simulations, which were carried out on restricted motion of domain walls and skyrmion. Here, energy confinement refers to using energy wells or energy barriers in a magnetic system to control current-induced dynamics. This chapter has been written in two parts: i) motion of domain wall to generate high amplitude microwave and ii) confined motion of skyrmions to suppress skyrmion Hall effect. The earlier part of this chapter details micromagnetic simulations on domain walls, which have generated a strong signal and frequency in GHz. The proposed microwave generation method has its niche application in telecommunication and promising use in emerging technologies like internet of things (IoT) devices. The latter part of this chapter gives details of energy confinement for a guided motion of skyrmion. The studied confinement method suppresses the unwanted motion of skyrmion in the transverse direction caused by the skyrmion Hall effect, which can be used for computing and synaptic devices.

5.2 Generation of microwaves

In this part of the chapter, generating a microwave with a strong signal via a domain wall motion has been explained. A magnetic nanowire with a sequence of oppositely magnetised domains in one half of the nanowire was simulated to generate the microwave. The domain walls were moved by applying spin orbit torque to the nanowire. Generation of the microwaves was carried out by detecting the changes in magnetisation value, which can be done using a magnetic tunnel junction (MTJ) in an actual device. The oscillating frequency of magnetisation was found to be in GHz. Moreover, a method to demonstrate the tuning of frequency was also defined. The outcome of this work promises the application of the reported method to generate microwaves in nanoscale devices of the future.

Figure 5.1. Schematics of the microwave oscillator device based on domain wall motion. Opposite magnetised domains were defined periodically before moving back and forth in nanowire [1].

5.2.1 Initial investigations of microwave generation

A nanotrack was defined with a dimension of 2000 nm x 100 nm x 1 nm to generate microwaves. The used magnetic parameters were taken from previous reports [2-6] as follow: saturation magnetisation $M_s = 5.8 \times 10^5 \text{ Am}^{-1}$; exchange stiffness $A_{ex} = 15 \times 10^{-12} \text{ Jm}^{-1}$; the DMI constant $D = 3 \times 10^{-3} \text{ Jm}^{-2}$; the damping parameter $\alpha = 0.3$; the spin Hall angle $\theta_{SH} = 0.04$, and the perpendicular anisotropy constant $K_u = 7 \times 10^5 \text{ Jm}^{-3}$. We have used MuMax3 for simulations, which provides induced DMI for thin films with out-of-plane symmetry breaking [7], yielding an effective field term, included in the effective field.

To move the domain walls, we have mimicked the spin orbit torque mechanism, where we have considered that the charge current is applied in the x-axis of heavy metal. Therefore, the polarisation of spin will be along the transverse to the direction of applied charge current (y-axis), and spin current injection (the spin current density equals the product of spin hall angle and charge current) will be along the z-axis. It must be noticed that the used value of $\theta_{SH} = 0.04$ is for Pt. However, by using other materials as an electrode, one can achieve a higher value of θ_{SH} for example, 0.15 (for Ta) [8] or 0.62 (for W) [9], resulting in the reduction of applied current density. The simulated nanowire was patterned with periodic domains of opposite magnetisations. The dimension of introduced opposite domains is 20 nm x 100 nm.

Figure 5.2. Simulated images of nanowires for generation of the microwave. (a) Oppositely magnetised domains in a nanowire. Zoomed-in image shows the random width of domains, which occurs due to the expansion of domain during magnetisation relaxation. (b) Moved domain walls to the right side of nanowire. Domain width shows stochastic expansion [1].

The oscillating magnetisation of the domains during the motion of domain walls was measured to mimic the MTJ output signal. This data was read from a reading region of dimension 20 nm x 100 nm defined in the middle of the nanotrack (as shown in figure 5.1). Fast Fourier transform (FFT) of the oscillating magnetisation was carried out to calculate the microwave's parameters like frequency, amplitude, and line width.

The computed width of magnetic domains was set to be 20 nm (figure 5.2). However, the relaxation of magnetisation to the minimum energy state resulted in an unwanted expansion of domains, which increased the width of domains. Zoomed-in image of figure 5.2a exhibits non-uniform distribution in the width of domains after the evolution of magnetisation to the minimum energy state. Moreover, the irregularity in the domain width was increased during the motion of domain wall when a current was applied. Figure 5.2b shows the simulated image of nanotrack after applying a current density $j = 1.4 \times 10^{12} \text{ A/m}^2$ for a time of 6 ns. The width of domains became irregular after the motion of domain walls. The irregularity in domain walls will increase further when domains are moved back and forth. Such irregularity in the size of domains will increase the bandwidth of generated microwaves.

Figure 5.3. Generation of microwave from an unconfined motion of domain walls. (a) Oscillating magnetisation of the domains detected during the motion of domain walls. (b) Fast Fourier transform of the oscillating magnetisation in a nanowire without pinning sites. The output signal frequency of 1.7 GHz was observed [1].

Figure 5.3a shows the oscillating magnetisation in the nanowires. The corresponding FFT is shown in figure 5.3b. The observed frequency of oscillation was calculated to be 1.7 GHz. The linewidth of the signal peak was calculated by fitting the peak to a Lorentzian function and measuring the full width at half maximum. The linewidth was measured to be 3.08×10^2 MHz. The time axis of figure 5.3a is around 6 ns, as this was the time taken to move the domains from left to right of the nanowire with a current density of 1.4×10^{12} A/m². It must be mentioned here that the time limit of 6 ns in this study is due to computational limitations in the simulated long wire. In a practical device, the wire can be longer (about 100 μ m). Therefore, although the proposed concept has potential, the randomness in the width of the domain will result in a broader line width of the signal.

5.2.2 Introducing energy confined geometries as pinning sites

To solve the repercussion of an unconfined motion of domain walls, geometries of different effective anisotropy energy were introduced. Figure 5.4a shows the schematics of the nanowire with pinning sites. Yellow-colored regions depict the pinning sites with a width of 10 nm in the nanowire. The pinning sites were introduced to the nanowire by introducing regions of a lower uniform anisotropy energy density K_u (95% of K_u of the rest of the region). Such a reduction in perpendicular magnetic anisotropy can be achieved experimentally by inter-diffusion with non-magnetic elements [10], implanting ions [11], or by fluctuating the domain wall surface energy [12] in the specific regions. The pinned regions have the dimension of 10 nm x 100 nm. Oppositely magnetised domains with a width of 20 nm were introduced. The total length of the nanowire is 3000 nm. After introducing such pinning sites, the magnetisation of the nanowire was relaxed to a minimum energy state. As a result, a uniform-shaped magnetic domain was observed.

Fig. 5.4. Images of simulated nanowires with pinning sites. (a) Nanowires with pinning sites are represented by yellow regions, which have different perpendicular anisotropy than the rest of the nanowire. (b) Oppositely magnetised domains in a nanowire with pinning sites. Zoomed in the image shows more uniform domains due to the introduction of pinning sites [1].

Figure 5.4b shows the image of the simulated nanowire after introducing the pinning sites. Zoomed-in image of the simulated nanowire depicts the uniform width of domains after the

evolution of magnetisation to the minimum energy state. The random distribution of pinning sites (due to material inhomogeneity) can introduce a stochastic motion of domain walls in the experiments [13], which have been ignored in this study.

To observe the effect of pinning sites on continuity of adjacent domain, the K_u was varied at pinning sites by two different values (10% or 5%). Figure 5.5 shows the simulated images of the nanowire with a 10 % variation of K_u for pinning sites. A uniform magnetic domain structure can be observed at $t = 0$, under zero applied current. Figure 5b-d show the simulation images when current is passed through the nanowire with current density $J_c = 3.0 \times 10^{11}$ A/m² at different simulation time. As simulation time increases, it can be noticed that the first oppositely magnetised domain collapses during the motion of the domain from the pinning site. This behavior continues for the rest of the domains, which can be observed from figure 5.5c and (d).

Figure 5.5. (a) Simulated image of nanowire with synthetic pinned sites (with 10 % variation of K_u). (b)-(d) Simulations images when current is passed with current density $J_c = 3.0 \times 10^{11}$ at different time. Nanowire with 10% of variation of anisotropy at pinned sites, show a lag in the motion of domains which result in the collapse of domain.

The observed collapse in magnetisation domains will result in inconsistency of generated frequency from microwave. To cope with this issue, the confinement energy of pinning sites was reduced with the variation of K_u around 5%. Figure 5.6 shows the simulated images of domain wall motion in a nanowire with synthetic pinned sites (5 % variation of K_u). Unlike the domain wall motion in the previous case, which suffered from the collapse of domains, the nanowire with 5 % variation of K_u at pinning sites shows the non-collapsing motion of domains through the pinning sites. The used value of anisotropy at pinning sites for the rest of the study is $K_u = 6.65 \times 10^5 \text{ Jm}^{-3}$.

Figure 5.6. (a) Simulated image of nanowire with synthetic pinned sites (with 5 % variation of K_u). (b)-(d) Simulations images when current is passed with current density $J_c = 3.0 \times 10^{11}$ at different times. Nanowire with 5% of the variation of anisotropy at pinned sites show a lag in the motion of domains which result in the collapse of the domain.

5.2.3 Microwave generation using domain wall motion

To generate a microwave, the motion of domain walls was carried out in a nanowire with pinning sites. In this case, the observed frequency of generated microwave was calculated to be 2.1 GHz, which is higher than the frequency of microwave observed in nanowire without pinning sites. The applied current density, in this case, was the same as the previous case ($j =$

$1.4 \times 10^{12} \text{ A/m}^2$). The time taken to move domains from the left to right side of the nanowire was 10 ns. The increase in the time taken by the domains to move to the right is due to the increased length of nanowire for pinning sites (3000 nm). Hence, to compare the properties of two nanowires, we used the data from the same period (6 ns). The increase in the observed frequency of the generated microwave in nanowire with pinning sites can be explained because of the increased uniform width of moving domains. As the size of domains remains uniform during the motion of the domain wall, a more significant number of domains passed through the reading region than in the previous case (nanowire with no pinning sites).

Figure 5.7. Generation of microwave from a confined motion of domain walls. (a) The oscillating magnetisation of the domains detected during the motion of domain walls. (b) Fast Fourier transform of the oscillating magnetisation in a nanowire without pinning sites. The output signal frequency of 2.1 GHz was observed [1].

Figure 5.7a exhibits the oscillating magnetisation of the domains detected during the motion of domain walls for the same time span as for nanowire without pinning sites (6 ns). The number of peaks in the inset indicates the number of domains passed through the reading region. By comparing the number of peaks (from figure 5.3a and 5.7a), it can be noticed that the number of domains passing through the reading region in the case of the nanowire with pinning site is higher than the nanowire without pinning sites. The line width of the signal was measured

to be 1.72×10^2 MHz, which is almost half of the line width of the signal measured in the case of nanowire without pinning sites. The reduction in the broadness of the line width can be explained due to the reduction in the randomness of the width of domains. Noticeably, the result of this investigation infers the role of domain size and domain uniformity on the frequency and pulse of the generated microwave. It can be concluded that by varying the domain size, the frequency of microwaves can be tuned. One may need to notice that the introduction of MTJ to the magnetic wire can influence the homogeneity of the magnetic properties of the wire and thus cause a significant domain wall pinning. This consequence of introducing the MTJ has not been considered in this reported approach. Secondly, we also have concern that the stray field from MTJ's reference layer can result in pinning of domain wall in the vicinity of MTJ [14]. However, we believe this deteriorating effect can be circumvented using synthetic anti-ferromagnetic coupled layers as a reference layer in the MTJ [15].

5.2.4 Effect of varying DMI on the generated microwave

Figure 5.8. Manoeuvring the frequency of generated microwaves. (a) Tuning the frequency of generated microwave by varying the current density. (b) Increased range of generated frequency with variation in DMI [1].

Furthermore, the effect of DMI was investigated on the properties of the generated microwave.

The value of DMI was varied in a range from $D = 1.0 \times 10^{-3}$ Jm⁻² to $D = 3.0 \times 10^{-3}$ Jm⁻²

to observe the impact on signal frequency, line width, or amplitude of the generated frequency. However, there were no noticeable consequences of varying the DMI on the velocity of domain wall motion at lower values of applied current densities. The frequency of the generated microwaves remained the same for all values of D at lower current densities. Nonetheless, as the current density values were increased, higher velocity values were observed, as also reported by Zhang et al. [16]. The calculated frequency for different values of D is shown in figure 5.8b, which shows maximum frequency of 5.18 GHz. Moreover, there was not any difference noticed in the domain width, which is a promising sign for application. The amplitude of the generated microwaves was almost similar and did not show any specific trend for the variation of DMI. The line width of generated microwaves of different DMI values also remained in the range of 10^2 MHz, and we did not observe any specific trend between values of line width for different DMIs.

5.2.5 Narrowing the bandwidth of signal

The broadness in the bandwidth of generated signal arises from the slight inclined variation in the shape of domains (due to the pinning from edges) during the domain wall motion. The pinning effect from the edges is inevitable. However, the adverse effect of edge pinning on the bandwidth can be mitigated in different ways. We have carried out studies to narrow the bandwidth of generated signals by varying the size of the reading region, the width of the microwire, or the length of the microwire. The effect of varying these dimensions on the bandwidth of the generated signal is summarised below.

Figure 5.9. Plot depicting the variation of bandwidth as a function of reading region width. A small change was observed in the bandwidth as the width of the reading region is reduced [1].

Varying the width of the reading region

The size of the reading region used for earlier studies is 20 nm x 100 nm. The bandwidth of the output generated signal for $j = 1.40 \times 10^{12} \text{ A/m}^2$ with 10 ns simulation time was 1.069×10^2 MHz. The width of the reading region was varied from 24 nm to 16 nm while keeping the height constant as 100 nm. By varying the width of the reading region, a maximum reduction of bandwidth by 3 % was observed (figure 5.9). We believed that this change in the bandwidth is not significant, but this study facilitated the possibility of reducing bandwidth's broadness. Therefore, to further reduce the broadness in the bandwidth, we varied the width of the microwire as the next step.

Figure 5.10. Plot depicting the variation of bandwidth as a function of reading region width. A small change was observed in the bandwidth as the width of the reading region is reduced [1].

Varying the width of microwire

We have varied the width of the microwire from 400 nm to 50 nm while keeping the length constant. Figure 5.10 exhibits the variation of bandwidth as the function of nanowire's width. Noticeably, the broadness in the bandwidth was reduced as compared to the previously studied nanotrack of width as 100 nm (bandwidth of 1.069×10^2 MHz for $j = 1.40 \times 10^{12}$ A/m² with 10 ns simulation). The main purpose behind studying a nanotrack with increased width was to reduce inclination in the 'V' shape of domains during the motion domains, as shown in figure 5.11b. However, it can be noticed from the plot in figure 5.10 that the bandwidth did not prominently reduce with increasing the width. The bandwidth of the signal was reduced a bit for 200 nm width of the nanowire. However, on further increasing the width of the nanowire (400 nm), the bandwidth of the signal was increased. This increase in the bandwidth arises due to curviness in the shape of domains when the width is increased, as shown in figure 5.11a.

Figure 5.11. Simulated images of nanowires with different widths. (a) Image of the nanowire with a width of 400 nm. Yellow circles show the domains with a curve shape, which causes an increase in the broadness of bandwidth. (b) Simulated image of the nanowire with a width of 100 nm. The zoomed-in image shows the ‘V’ shape of domains. (c) Simulated image of the nanowire with a width of 50 nm. Zoomed in the image shows the uniformly deformed domains, which reduced the broadness of the bandwidth of the generated signal [1].

Remarkably, a substantial decrease in signal bandwidth was noticed when the width of the nanowire was reduced to 50 nm. This decrease in the bandwidth was arisen due to the modification of domains’ shape from rectangular to a ‘V’ shape (shown in figure 5.11c), which eventually increased the uniformity in the shape of domains. We speculate the formation of ‘V’ shape magnetic domains might have arisen from increased spin density in the microwire, which may have sustained the uniformity of domains. Further, we increased the number of domains crossing under the reading region by increasing the simulation time for fixed-length or increasing the nanowire length, which increased the number of oppositely magnetised domains in a track.

Varying the number of domains passing through the reading region

Figure 5.12. Plot depicting the variation of bandwidth as a function of domains passing through the reading region [1].

As the number of domains passing through the reading region increases, the uniformity in domain shape increases, the bandwidth of the domain was reduced. The maximum number of domains passed through the reading region was 96, which showed the maximum decrease in the broadness of bandwidth. Unfortunately, we could not increase the number of domains further due to computational limitations. As we can not the introduction more new regions in the simulations. However, we would like to articulate that the bandwidth will be reduced by further increasing the number of domains.

5.3 A guided skyrmion motion through energy confinement

A new approach to restrain the SkHE by introducing Dzyaloshinskii-Moriya interaction (DMI) energy wells on the edges of nanotrack has been studied by us. The benefit of this method is that DMI can be manipulated by slightly varying interface of a ferromagnet.

5.3.1 Restricting the skyrmion Hall effect using the DMI energy well

First, we studied the motion of skyrmion in a normal nanotrack. The nanotrack was simulated with a cell size of 1 nm^3 , with a dimension of $500 \text{ nm} \times 50 \text{ nm} \times 1 \text{ nm}$. The magnetic parameters were similar to those in previous articles [2-4, 17]: exchange stiffness $A_{ex} = 15 \times 10^{-12} \text{ Jm}^{-1}$; the damping parameter $\alpha = 0.3$; saturation magnetisation $M_s = 5.8 \times 10^5 \text{ Am}^{-1}$; perpendicular anisotropy constant $K_u = 8 \times 10^5 \text{ Jm}^{-3}$, and spin Hall angle $\theta_{SH} = 0.40$. The value of DMI constant D for nanotrack was set to $3.0 \times 10^{-3} \text{ Jm}^{-2}$, and for the DMI energy wells, D was varied from 0.0 Jm^{-2} to $3.0 \times 10^{-3} \text{ Jm}^{-2}$. The skyrmions were moved in the nanotrack by mimicking the spin orbit torque mechanism.

Figure 5.13a shows the simulated image of the skyrmion in a magnetic nanotrack with constant $D = 3.0 \times 10^{-3} \text{ Jm}^{-2}$. The skyrmion was created at 70 nm from the left of the track. Figure 5.13b depicts the motion of skyrmion in both longitudinal and transverse direction of the nanotrack at $t = 0.60 \text{ ns}$ under an applied current density of $j = 20 \times 10^{10} \text{ Am}^{-2}$. Figure 5.13c shows the annihilation of skyrmion at the edge of the track at $t = 1.00 \text{ ns}$. During the annihilation, the calculated topological charge of the skyrmion was reduced from -0.99 (almost 1) to 0.06 , indicating the destruction of the topologically stable structure. Figure 5.13d shows a track without any skyrmion at $t = 1.05 \text{ ns}$.

Figure 5.13. (a)-(d) Simulated images depicting the skyrmion Hall effect. (a) Skyrmion in a magnetic nanotrack nucleated at 70 nm from the left of the track. (b) The motion of skyrmion to the upper side of the track after time $t = 0.60$ ns. (c) Annihilation of skyrmion at the edge of the track at $t = 1.00$ ns. (d) Image of the skyrmion less track at $t = 1.05$ ns, after the annihilation of the skyrmion. (e)-(h) Introducing the modified DMI energy regions on a nanotrack to confine the skyrmion Hall effect (the coloured/shaded areas show the regions with lower DMI energy). (e) Schematics of a nanotrack with modified DMI. (f) Simulated image of a nanotrack with skyrmion confined between two DMI energy wells at $t = 0.00$ ns. (g) Skyrmion motion along the boundary of the upper energy well after $t = 2.65$ ns ($j = 20 \times 10^{10} \text{ Am}^{-2}$). (h) The successful motion of the skyrmion towards the right end of nanotrack [18].

The observed transverse motion of skyrmion in the nanotrack is the consequence of spintronic version of Magnus force, as explained in chapter 2 [19-21].

To overcome the annihilation of skyrmion, we have proposed the use of locally modified DMI energy wells. DMI can be modified experimentally by irradiating ions or diffusing non-magnetic elements at the interface of heavy metal/ferromagnet [10]. Here, the simulated DMI energy wells along the edges of the nanotrack as shown in figure 5.13e. The coloured areas represent the modified DMI energy regions with $D = 0.0 \text{ Jm}^{-2}$ and the remaining region has a value of $D = 3.0 \times 10^{-3} \text{ Jm}^{-2}$. The width and length of the modified regions are 10 nm and 500 nm, respectively. The distance (d) between the two DMI energy modified regions is 20 nm

unless specified otherwise. Figure 5.13f shows a track with nucleated skyrmion between two DMI energy wells at time $t = 0.00$ ns. Due to the introduction of DMI energy wells, the skyrmion diameter was reduced slightly, from 21.5 nm (without any energy well) to 21 nm (with energy wells). After sending a current of $j = 20 \times 10^{10} \text{ Am}^{-2}$, skyrmion was observed to move along the boundary of the top energy well (figure 5.13g). The skyrmion drags along the interface between the modified DMI region and track. Figure 5.13h shows the simulated image of nanotrack after skyrmion travelled to the right end. The total simulation time was 6.00 ns. However, the skyrmion reached the end of the track at $t = 5.30$ ns and stayed there for the remaining simulation time. The simulation results demonstrated the successful motion of a skyrmion along the nanotrack. The velocity of the skyrmion motion was observed to be 79 ms^{-1} .

Moreover, the skyrmion total energy also increases when D is reduced, which can be calculated from the following formula [22]

$$E(R, W) = 4\pi d' \left[A_{ex} \left(\frac{R}{W} + \frac{W}{R} \right) - \frac{\pi}{2} DR + K_{eff} WR + M_s B \left(\frac{R^2}{2} + \frac{\pi^2}{24} W^2 \right) \right]$$

where $R = \pi D \sqrt{\frac{A_{ex}}{16A_{ex}K_{eff}^2 - \pi^2 D^2 K_{eff}}}$, $W = \frac{\pi D}{4K_{eff}}$ and d' is the thickness of the ferromagnetic layer. The first term of the above equation refers to the exchange energy, the second term refers to the DMI energy, the third term refers to the anisotropy energy and the fourth term refers to Zeeman energy associated with skyrmion. Since, the applied magnetic field B in our simulation is zero, the Zeeman energy associated with skyrmion resultingly becomes zero. Figure 5.14a and 5.14b exhibit the changes of different energies with D . The increase in total skyrmion energy with reducing D (Figure 5.14a) directs the confinement of skyrmion inside the DMI modified regions.

Figure 5.14. Variation of energies as a function of varying D , exhibiting the formation of potential energy barrier at low D . (a) Change in the total skyrmion energy with varying D . Domain wall energy varies linearly with D . The skyrmion energy is presented as a factor of $K_B T$, where K_B is Boltzmann constant and T is the temperature of the system in Kelvin. $T = 300$ K was used to represent the energy of the system at room temperature. (b) Variation of different energies associated with skyrmion as a function of D [18].

5.3.2 Measuring the threshold of DMI energy well

The SkHE can appear again if the difference between the DMI energy of the track and the energy well decreases. To determine this threshold value of DMI energy of well (D_{th}), which can resist the SkHE, the difference between the DMI energy values of the wells and track was varied. For this study, the DMI energy of the track was kept constant as $3.0 \times 10^{-3} \text{ Jm}^{-2}$. The DMI energy of the wells was varied from 0.0 Jm^{-2} to $3.0 \times 10^{-3} \text{ Jm}^{-2}$. The distance between the energy wells (d) and the applied current density were also kept constant as 20 nm and $20 \times 10^{10} \text{ Am}^{-2}$, respectively. Using these parameters, the results indicated that the DMI energy value (D) up to $2.80 \times 10^{-3} \text{ Jm}^{-2}$ confined the motion of skyrmion along the boundary. However, for a D value of $2.85 \times 10^{-3} \text{ Jm}^{-2}$ and above, annihilation of skyrmion occurs at the upper edge of track. Hence, $D = 2.80 \times 10^{-3} \text{ Jm}^{-2}$ can be considered as the threshold value of DMI (D_{th}), which can resist the transverse motion of skyrmion towards the upper edge of the track.

Figure 5.15. Annihilation behaviour of skyrmion for different D of the energy wells (a) Simulated image of nanotrack exhibiting the skyrmion in relaxed state at $t = 0.00 \text{ ns}$ without any applied current. (b)-(e) are the simulation results showing the annihilation of skyrmion at different points of time due to different values of D . (f) Dependence of skyrmion velocity and threshold DMI on the current density [18].

Figure 5.15a shows the position of a skyrmion in the track at $t = 0.00 \text{ ns}$. Figure 5.15b-e show the simulated images of tracks depicting the annihilation of skyrmion after the transverse motion of skyrmion due to SkHE for $D = 2.85 \times 10^{-3} \text{ Jm}^{-2}$, $2.90 \times 10^{-3} \text{ Jm}^{-2}$, $2.95 \times 10^{-3} \text{ Jm}^{-2}$ and $3.00 \times 10^{-3} \text{ Jm}^{-2}$, respectively. It can be noticed from the simulation images that the time at which the annihilation of skyrmion occurs decreases as D of the energy wells approaches the D of the track ($3.00 \times 10^{-3} \text{ Jm}^{-2}$). Interestingly, the skyrmion velocity was observed to be maximum at the threshold D value of the energy barrier. Moreover, the skyrmion has the minimum velocity (79 m/s) for $D = 0.0 \text{ Jm}^{-2}$ and the maximum velocity ($\sim 87 \text{ m/s}$) at the threshold value of D , which is $2.80 \times 10^{-3} \text{ Jm}^{-2}$. Further, an increase in skyrmion velocity was observed as the value of D increases from 0.0 Jm^{-2} to the threshold.

We speculate that the threshold DMI (D_{th}) will be different for different applied current densities. We varied the value D_{th} for different applied current densities and further

investigated the maximum and minimum velocities to confirm this speculation. The applied current density was varied from $10 \times 10^{10} \text{ Am}^{-2}$ to $80 \times 10^{10} \text{ Am}^{-2}$. To calculate the maximum and minimum velocities of skyrmion and D_{th} for energy wells, D of energy wells was varied. Figure 5.15f shows the plot of skyrmion velocities and D_{th} vs. current density. By increasing the current density, an increase in both minimum velocity (which was at $D = 0.0 \text{ Jm}^{-2}$) and maximum velocity (which was near to the D_{th} of energy wells) was realised. Noticeably, the D_{th} value of the energy wells was found to decrease with an increase in the current density. This observation inferred that a more substantial energy barrier (DMI energy difference between the track and barriers need to be high) is required to restrict SkHE for higher current densities.

5.3.3 Effect of restricted geometries on the skyrmion

The magnetic properties define the diameter of skyrmion in a material system. Tuning the skyrmion diameter (besides varying the materials parameters) is a valuable knob to get skyrmion for high-density storage and logic devices. Therefore, the distance between the energy wells (d) and DMI energy constant D of the tracks was varied to see if the skyrmion size can be reduced using the proposed design. The current density was kept constant as $20 \times 10^{10} \text{ Am}^{-2}$. Figure 5.16a and b show the simulation images of confined skyrmion between energy wells with $d = 5 \text{ nm}$ and a D of $2.0 \times 10^{-3} \text{ Jm}^{-2}$. Above this value of D , SkHE (skyrmion moved over the DMI modified region to reach the top edge of track) and the subsequent annihilation of skyrmion was observed. Below this value of D , a stable skyrmion could not be nucleated. Figure 5.16c summarises the effect of varying d and D on the velocity and diameter of skyrmion. It can be noticed that the diameter of the skyrmion (blue colour font in summary) shrinks with a reduction in the value of d , as predicted. However, this effect is more pronounced for smaller values of D . The velocity of skyrmion (red colour font in summary) showed a

similar trend as the diameter of skyrmion, viz., the velocity was smaller for smaller values of d and D . Hence, the velocity and diameter of skyrmion can be manipulated by modifying the energy of DMI energy wells and the distance between the DMI energy wells.

Figure 5.16. Effect of varying the distance between the energy wells on diameter and velocity of skyrmion. (a) Simulation image of nanotrack depicting a reduced-sized skyrmion confined between energy wells with distance $d = 5 \text{ nm}$. (b) Simulation image of nanotrack depicting the motion of skyrmion after $t = 6.00 \text{ ns}$. (c) Summary of the distance between the energy wells on diameter and velocity of skyrmion [18].

5.3.4 Magnetostatic interaction between the skyrmions

Figure 5.17. Effect of magnetostatic interaction between multiple skyrmions. (a) Observed position of an isolated skyrmion at $t = 6.0 \text{ ns}$, under applied current density $j = 10 \times 10^{10} \text{ Am}^{-2}$. (b)-(d) Observed positions of skyrmions in parallel tracks ($t = 6.0 \text{ ns}$, $j = 10 \times 10^{10} \text{ Am}^{-2}$). The increase in magnetostatic interaction, as effected by a reduction in the spacing between the tracks, leads to an increase in the lagged displacement of the middle skyrmion and gain in the displacement for the skyrmions at the edges. (e) Skyrmion positions (for track-pitch = 25 nm), at $t = 0.00 \text{ ns}$ and $t = 0.05 \text{ ns}$. A strange negative displacement of skyrmion at the upper track ($t = 0.05 \text{ ns}$) and an eventual collapse (not shown) of skyrmions at outer tracks due to increased magnetostatic interaction were observed [18].

To use skyrmions for high-density storage or logic applications, skyrmions need to be moved in parallel track. In a closed pack skyrmion state, skyrmions will encounter magnetostatic interactions. Hence, it is essential to estimate the effect of magnetostatic interaction on closely packed skyrmions. Therefore, we studied the effect of magnetostatic interactions between three skyrmions in neighbouring tracks. For this study, centre-to-centre distance (track-pitch) was varied between skyrmion tracks (by varying the width of energy wells) and observed the behaviour of skyrmion under various applied current densities. The DMI of track and the energy wells were kept constant as $3.0 \times 10^{-3} \text{ Jm}^{-2}$ and 0.0 Jm^{-2} , respectively. In such a scheme, the skyrmion in the middle track has two neighbours compared to the skyrmions at the edge tracks (which have only one neighbour). Hence, the presence of magnetostatic interaction, if any, will significantly affect the motion of the skyrmion in the middle track.

Figure 5.17a shows the position of a single skyrmion without any magnetostatic interaction at $t = 6.0 \text{ ns}$ under current density $j = 10 \times 10^{10} \text{ Am}^{-2}$. Figure 5.17b-d depicts the position of skyrmions in parallel tracks for the same t and j . A reduction in w (track-pitch) leads to an increase in magnetostatic interaction. A lag in the distance travelled by the middle skyrmion was observed for figure 5.17b-d (for a certain magnetostatic interaction). The lag in the distance of the centre skyrmion increases as the magnetostatic interactions between skyrmions increases. Interestingly, this also manifests in to gain in the displacement for the skyrmions at the outer tracks.

When the track pitch (w) was reduced to 25 nm , a decrease in the size of the middle skyrmion at $t = 0.0 \text{ ns}$ was observed. When a current was sent, we observed a diagonal motion of skyrmions in outer tracks to the upper and lower edges of the tracks. Figure 5.17e shows that the top skyrmion moved backward to the upper left of the edge while the bottom skyrmion moved forward to the lower right of the edge before annihilating at the edges of the track.

Figure 5.18 shows the lag in the distance travelled by skyrmion for different values of track pitches and various current densities. A decrease in the lagged distance was observed when the applied current density was increased. This behaviour indicates that the effective field on skyrmions arising from a high current density can counter the repulsive magnetostatic field between skyrmions and keep the skyrmions closer to each other. The effect of magnetostatic force was negligible when track pitch was increased to 50 nm. These results indicate that the magnetostatic interaction between skyrmions in closely spaced adjacent tracks would cause an increase in energy required to move the skyrmions around.

Figure 5.18. Effect of magnetostatic interactions between skyrmion vs. applied current densities [18].

5.4 Summary

The chapter showed the potential application of energetically confined geometries. The domain wall motion as a prospective application for generating microwaves was explored through micromagnetic simulations. Computationally, a maximum value of 5.18 GHz microwave was generated. Nanowires with pinning sites have advantages over a nanowire without pinning sites. Microwave generated in a nanowire with pinning sites has a higher frequency and narrower linewidth than in a nanowire without pinning sites. This study on the nanowire also inferred that by varying the width of domains, the frequency of microwaves could be tuned.

Furthermore, the effect of varying current density, which resulted in varying domain wall velocity, offers a knob to tune the frequency of microwave frequency.

The dynamics of skyrmion confined between the DMI energy wells were studied. The introduction of the DMI energy wells in a nanotrack assuages overcoming the skyrmion Hall effect and confine the motion of skyrmion. Furthermore, the skyrmion diameter can be reduced to a certain extent using DMI energy wells. Finally, the effect of magnetostatic interactions on closed pack skyrmions was studied, which depicted that a certain pitch length is required to have a negligible effect on magnetostatic interaction. The outcome of this study shows a new scheme to manipulate the motion, velocity, and diameter of skyrmion.

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Chapter 6 Enhancing the Density of Skyrmions realized via manoeuvring Atomic Intermixing at Interface

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6.1 Introduction

The existence of skyrmion in thin films and the promising nature of skyrmions to move in a nanotrack offer a vast opportunity for skyrmion application in computing and memory devices. Such applications require a high skyrmion density, which depends on the material stack's magnetic properties. Precisely, a high ratio of Dzyaloshinskii Moriya interaction (DMI) to effective magnetic anisotropy and exchange coupling energies is required for achieving high skyrmion density. In this chapter, an increase in skyrmion density has been achieved via intermixing atoms at the interface. Two methods of atoms intermixing at the interface were explored. The first method was demonstrated via annealing, which causes the diffusion of atoms at the interface. The second method was ion irradiation, a comparatively high-energy method than annealing, which induces atoms intermix at the interface. The effect of these two treatments is different on the skyrmion density. This chapter reports the effect of the treatments on the magnetic and skyrmion properties of the sample.

6.2 Optimization of the pristine sample

It was important to choose a stack that generates skyrmion to carry out the study on interface modification. Since the future aim of this work is to realize the skyrmion motion in a nanotrack, an amorphous CoFeB stack was chosen, which has been reported previously for unhindered skyrmion motion [1]. To start with, a stack of Ta(5)/[Pt(3)/CoFeB(0.45-0.95)/MgO(1.5)]_x5/Pt(3) was deposited with different stoichiometries of Co and Fe as Co₄₀Fe₄₀B₂₀, Co₅₀Fe₃₀B₂₀, and Co₆₀Fe₂₀B₂₀. The number in parentheses represents the thickness of the films in nm. The magnetic properties of the stacks with different stoichiometries were measured by a vibrating sample magnetometer (VSM). Figure 6.1 shows the measured effective magnetic anisotropy energy density (K_{eff}) of different samples. The sign of K_{eff} depicts the magnetisation

direction of sample [2-4]. A positive K_{eff} represents a perpendicular magnetic anisotropy, while a negative value represents in-plane magnetisation direction.

Figure 6.1. Effective magnetic anisotropy energy (K_{eff}) of different stoichiometries of Co and Fe. A reduction in the value of K_{eff} represents a decrease in the perpendicular magnetic anisotropy energy.

The value of K_{eff} was seen to be reduced with an increase in the thickness of samples, as explained in chapter 2. The decrease in the K_{eff} represents a reduction in the perpendicular magnetic anisotropy. The 0.6 nm thick $Co_{40}Fe_{40}B_{20}$ and $Co_{50}Fe_{30}B_{20}$ samples show an in-plane magnetisation. However, the 0.6 nm thick $Co_{60}Fe_{20}B_{20}$ sample depicted a weak perpendicular magnetic anisotropy. As explained in chapter 2, DMI in these samples is generated from the interface of Co and Pt due to spin orbit coupling between the Pt (heavy metal) and Co atoms [5, 6]. A high concentration of Co will generate a high value of DMI. Hence, $Co_{60}Fe_{20}B_{20}$ stoichiometry was chosen for further studies.

As the next step, the number of repetitions for the $Co_{60}Fe_{20}B_{20}$ sample was varied. The repetition of the stack is crucial, providing dipolar coupling between the layers and stabilising the skyrmions at room temperature. For this, different repetitions of $Co_{60}Fe_{20}B_{20}$ were deposited, as shown in figure 6.2a. The plot of K_{eff} for different repetitions has been shown in figure 6.2b. The K_{eff} results of 0.5 nm thick samples show a perpendicular magnetic

anisotropy. However, for 0.6 nm thickness, only 5 repetitions sample showed a positive K_{eff} , whereas 0.6 nm sample with 8 repetitions showed a nearly zero K_{eff} indicating lack of any preferred direction of magnetic anisotropy. On the other hand, a 0.6 nm thick sample with 12 repetitions has a negative K_{eff} value which indicates an in-plane magnetisation. The decrease in the K_{eff} with increasing thickness was identical in samples with different stoichiometries.

Figure 6.2. The stack structure of the pristine samples and the effective anisotropy for various values of repetition of CoFeB stack and thickness.

Further, to select suitable samples for further investigations, magnetic force microscopy (MFM) images of 0.5 nm and 0.6 nm thick samples were collected to see the effect of repetitions on the magnetic domain structures in these samples. Figure 6.3 shows MFM images with calculated periodicity for different repetitions.

A high periodicity value indicates a high domain wall energy [7], reflecting a larger skyrmion size. Moreover, a lower repetition sample stack (5x) shows limited dipolar coupling, restricting the stability of skyrmions and poor contrast in MFM imaging. The periodicity of magnetic stripes increases with an increase in repetitions depicting a reduction in the density of magnetic stripes. With this perspective, 0.6 nm thick sample sounds useful. However, to achieve a high density with a small diameter of skyrmion, the ratio of DMI over K_{eff} needs to be high. However, a material (0.6 nm thick) with negative K_{eff} leads to a larger diameter of skyrmions. Moreover, this study aimed to reduce the K_{eff} without compromising DMI. Treatment of

samples with 0.6 nm thickness, which have negative K_{eff} , will lead to a further reduction in the K_{eff} . This reduction in the value will lead to unwanted magnetic texture sizes. Hence, it is better to choose a sample with positive K_{eff} .

Figure 6.3. MFM images of (a) 0.5 nm and (b) 0.6 nm thick samples for various repetitions. The periodicity of magnetic stripes shows an increase in stripe domain density with an increase in the number of repetitions.

6.3 Properties of the pristine sample

Based on the optimization of the pristine sample, which was investigated in the previous sections – 0.5 nm thick $\text{Co}_{60}\text{Fe}_{20}\text{B}_{20}$ sample was chosen. The magnetic properties, namely saturation magnetisation (M_s) and effective magnetic anisotropy density (K_{eff}) of this sample were measured by VSM (also confirmed by alternating gradient field magnetometer (AGM)).

DMI was measured by Brillouin light spectroscopy (BLS). The hysteresis loops of pristine samples are shown in figure 6.4.

Figure 6.4. Magnetic Hysteresis loop of the pristine sample. (a) Out of plane and in-plane and (b) Measured zoomed-in plot at small field range.

The magnetic properties of pristine sample were measured and calculated to be: $M_s = 921$ kA/m, $K_{eff} = 0.135$ MJ/m³, and DMI = 1.25 mJ/m². Before proceeding further, magnetic textures of the pristine sample were imaged using magnetic force microscopy (MFM).

Figure 6.5. MFM images of pristine sample imaged at the different static fields, applied in perpendicular direction. Saturation field H_S was estimated from the FORC plot.

Magnetic force microscopy imaging for various applied fields was done on the pristine sample, and the results are shown in figure 6.5. The field value is calculated with respect to the saturation field (H_S), which was measured from the FORC plot. The dense stripes domains (observed at 0 mT) were broken into smaller stripes at a field of 20 mT ($0.32 H_S$). The density of stripes was further reduced at higher fields. However, all stripe domains did not transform into skyrmion bubbles. Further, increasing the field leads to a smaller skyrmion structure. The density of skyrmions can be seen diminishing with an increase in the magnetic field, and magnetic textures of pristine samples were thoroughly diminished at the field of 48 mT (0.78

H_S). From magnetic images, it can be concluded that the 0.5 nm pristine sample has a low-dense state of skyrmion.

6.4 Annealing induced diffusion of atoms at the interface

To induce the diffusion of atoms at the interface, magnetic annealing was carried out at different temperatures. The annealing of samples was performed at 150°C, 200°C, 250°C and 300°C for an hour in a vacuum oven. Magnetic properties of annealed samples were measured using VSM. Figure 6.6 shows the MH loops of annealed samples measured in the out of plane direction. The shape of the loops can be seen to become squarer with an increase in the annealing temperature. The increase in the squareness of loops infers an increase in the perpendicular magnetic anisotropy. The kink observed in MH loops after annealing at high temperature may arise due to different phases in multi-repetition ferromagnetic layers.

Figure 6.6. Magnetic Hysteresis loops of samples annealed at different temperatures.

The M_s of the samples was increased after annealing, reaching the highest value for 150°C (shown in figure 6.7). The increase in the M_s can be explained based on the migration of B atoms from the CoFeB layer, which results in lesser dilution in magnetisation.

Figure 6.7. Remanence magnetisation (M_r) and Saturation Magnetisation (M_s) of samples annealed at different temperatures.

The increase in the M_r indicates that the density of up and down magnetised domains is not equal. Further, the coercivity of the sample was also increased, as shown in figure 6.8. The increase in the magnetic coercivity of annealed samples is also inferred to an increased perpendicular magnetic anisotropy.

Figure 6.8. Coercivity of annealed samples.

To verify the effect of annealing on the perpendicular anisotropy, effective magnetic anisotropy energy was calculated from out of plane and in-plane loops of the annealed samples. Figure 6.9 shows an increase in the K_{eff} with an increase in the annealing temperature. The value increased from 0.13 MJ/m³ at room temperature to 0.55 MJ/m³ for a 300°C annealed sample.

Figure 6.9. Effective magnetic anisotropy energy (K_{eff}) of annealed samples.

To achieve a high density of skyrmions, K_{eff} needs to be lower than the DMI, which was the aim of this study. However, after annealing, K_{eff} has been found to increase, which supports a decrease in the domain wall density. Moreover, an increase in the M_r , which depicts a change in the proportion of up and down magnetic domains, was noticed. Magnetic samples annealed at 250°C and 300°C showed $M_r \sim M_s$. Therefore, there will not be any magnetic texture to be observed at this field. The increased values of K_{eff} and M_r infer a reduced density of magnetic textures. Hence, to confirm the effect on the density of stripes – MFM imaging of annealed samples was done. Figure 6.11 shows the MFM images at measured at zero fields.

Figure 6.10. MFM images of magnetic domains at zero field for annealed samples.

MFM images at zero field for annealed samples show a decrease in the domain density as compared to the pristine sample. The periodicity of magnetic domains at zero field was measured using 2 dimensional fast Fourier transform (FFT) [7, 8]. Figure 6.11 shows the periodicity of magnetic textures observed for samples annealed at different temperatures.

Figure 6.11. Periodicity of magnetic textures observed at zero field for annealed samples. High temperature annealed samples lack magnetic textures at zero field.

The periodicity of stripes domains varies from 168 nm (pristine sample) to 420 nm for 200°C. No magnetic textures were observed for higher annealing temperatures. The lack of any magnetic textures at zero field for higher annealed temperatures was corroborated by the squareness of the magnetic loops at these temperatures. The decrease in the stripe density of magnetic textures indicates a reduction in the skyrmion density too. Hence, magnetic imaging of samples at a high static magnetic field was carried out. Figure 6.12 shows the MFM images of samples measured at high static magnetic fields. The density of skyrmion was reduced after annealing. The K_{eff} increases due to improvement of interface quality (between CoFeB and Pt atoms) and possibly due to the hybridization between Oxygen (from MgO) and Fe atoms [9]. The enhanced perpendicular anisotropy enhances domain wall energy [10, 11] and hence, the density of magnetic textures was reduced. Therefore, an annealing treatment is not a viable method to increase the skyrmion density.

Figure 6.12. MFM images of annealed samples, measured at the high static magnetic field. The skyrmion density was reduced after annealing.

6.5 Ion irradiation-induced intermixing of atoms at interfaces

To induce intermixing of atoms at the interface, ion irradiation was carried out with He^+ ions. The irradiation energy was kept constant at 20 keV, which is high enough to settle ions in the substrate.

Figure 6.13. TRIM simulations for predicting the effect of ion irradiation on atomic displacement. (a) Final distribution of He^+ ions in the target sample. The simulated ion range is 120 nm. (b) The final position of displaced atoms due to collisions. (c) Zoomed in image of first tri-layer, depicting final positions of displaced atoms.

6.5.1 Monte Carlo simulations for ion irradiation

Monte Carlo simulations using transport range ions in matter (TRIM) were performed to understand the effect of irradiation on the atomic displacements. A sample size of 100k ions was chosen for higher accuracy.

Figure 6.13a shows the final distribution of ions settled into the sample. The plot y-coordinate has units: $(\text{atoms}/\text{cm}^3)/(\text{atoms}/\text{cm}^2)$, representing the number of ions distribution per unit depth. Noticeably, the estimated ion range is around 120 nm. Whereas the thickness of deposited layers is 48 nm. Therefore, most of the ions are reaching the depth of 120 nm and settling into the substrate. Figure 6.13b shows the final position of atoms displaced, measured for a sample up to 1000 ions for the first few layers (a higher number of ions for a sample space increases the number of dots, which is hard to deconvolute). Source represents the ion irradiation source, which is on the top surface of the sample. Figure 6.13c is a zoomed-in image showing the first three layers. Different coloured dots represent the position of different atoms after displacement

due to collision with He^+ ions and local atoms. We have noticed the displacement of ferromagnetic atoms (Co and Fe) into the Pt layer. The migration of ferromagnetic atoms into the Pt layer can lead to alloying with Pt and changes in the interfacial properties.

Moreover, B atoms (shown by orange dots) can also be seen migrating into Pt. Furthermore, Mg and O atoms (shown by pink and green dots, respectively) have moved into the ferromagnetic layer. Migration of non-magnetic atoms (Mg and O) can dilute the magnetisation of the ferromagnetic layer. Besides the migration of atoms into the adjacent layers, migration of atoms in the same layer has also been observed. Further, the probability distribution of displaced atoms was calculated, using a sample space of 100000 ions.

Figure 6.14. (a) Plot of atom displacement distribution calculated for large sample space. (b) Plot for ferromagnetic atoms migration into the Pt layer. Intermixing of ferromagnetic atoms increases linearly with the ion dose (c) Displacement of B in Pt layer vs. FM (CoFeB) FM layer.

Figure 6.14a shows the plot for atoms displacement distribution (zoomed for first tri-layer). Migration of Co, Fe, and B into Pt layers can be noticed in the plot. Moreover, displacement

of atoms is higher at the interface, inferring a possibility of interface intermixing. One can achieve the atomic displacement density after multiplying the atomic displacement per depth to the irradiated dose. Figure 6.14b shows the displacement of ferromagnetic atoms into the underneath Pt layer. The displacement of atoms increased linearly with ion dose. Moreover, we have noticed a higher migration of Co into Pt than that of Fe and B atoms. This can be explained due to the higher stoichiometry of Co in the ferromagnet layer as compared to other atoms. Furthermore, we have seen a considerably higher displacement of atoms within the same layer than a movement to the adjacent layer (also inferred from Figure 6.14a). Since the ferromagnetic layer is amorphous, the replacement of Co and Fe atoms with each other would not affect the structural and magnetic properties. However, the B atom - smaller in size than the Co and Fe atom - can migrate at interstitial sites (figure 6.14c). This migration to interstitial sites can affect the bulk properties like magnetisation. Hence, the TRIM results insinuate modification of bulk as well as interfacial properties.

6.5.2 Structure variation in irradiated samples

To understand the effect of irradiation on the structure of pristine samples, a cross-section transmission electron microscope (TEM) was carried out. The cross-section images of pristine samples are shown in figure 6.15a. The deposited thickness of CoFeB is 0.5 nm, which was not visible in the TEM images. Figure 6.15b shows the zoomed-in image of the cross-section TEM of the pristine sample. An indistinctly thin CoFeB layer (marked by teal colour dotted line) can be seen between the MgO and Pt.

Figure 6.15. Cross-section transmission electron microscopy (TEM) of Pristine and irradiated samples. Energy Dispersive X-rays spectroscopy of samples showed displacement of Co atoms away from layers (marked in red dashed circles)

Energy dispersive X-rays (EDX) spectroscopy of the cross-sectional regions was carried out to gain further insight. Figure 6.15c shows the Co mapping of pristine samples and irradiated samples. The lower irradiation dose (4×10^{18} ions/m²) resulted in a slight migration of Co atoms from the layer. For a higher dose, carried out at 8×10^{18} ions/m², atoms can be seen missing out from the actual ferromagnetic layer and moving to the adjacent layers. Hence, increasing the irradiation dose can increase the displacement of Co atoms from the layer.

Figure 6.16. Energy Dispersive X-rays spectroscopy of Pt map. No Pt displacement was seen.

Furthermore, from figure 6.16, no displacement of Pt atoms was seen for the same irradiation doses. This inferred that the used irradiation doses could displace Co atoms away from the ferromagnetic layer. However, for the Pt layer, which consists of heavy atoms Pt, displacement of atoms was lacking. Nevertheless, it can be concluded that by varying the irradiation dose, the migration of atoms at the interface can be varied. Hence, the intermixing of at interface will increase with an increase in the irradiation dose.

6.5.3 Magnetic properties of irradiated samples

Figure 6.17 shows the MH loop plots of irradiated samples with different ion irradiation doses. Compared to the pristine sample, irradiation has led to an increase in the saturation field of samples. Moreover, the sheerness of the loop was reduced after the irradiation. The coercivity of irradiated samples is shown in figure 6.18. The coercivity of samples after irradiation remained similar to the pristine sample.

Figure 6.17. MH loops of irradiated samples, measured in out of plane direction.

Figure 6.18. Coercivity of irradiated samples, measured from out of plane MH loops.

Further, M_r and M_s of irradiated samples were measured using VSM and AGM. Figure 6.19 shows the variation in magnetisation values due to different irradiation doses. The M_r of the sample remained low, inferring the presence of stripe domains at a high density. Unlike the annealed samples, MH loops of the irradiated samples still have a low value of M_r . The low value of M_r indicates equal area occupied by the up and down magnetized domains (which was observed in the pristine sample also). The M_s of irradiated samples showed an increase in the value after irradiation. A high value of M_s was achieved until $8\times$ (here after, $n\times$ indicates the n multiple of 10^{18} ions/m²) sample, and the value was reduced for higher irradiation dose ($12\times$). The increased value of M_s for irradiated sample is similar to the M_s value achieved for 150°C annealed sample.

Figure 6.19. Magnetic Remanence (M_r) and Saturation Magnetisation (M_s) of irradiated samples.

To see the effect of irradiation dose on perpendicular magnetic anisotropy, K_{eff} of the irradiated samples was measured. Figure 6.20 shows the calculated K_{eff} from out of plane and in-plane magnetic loops of the irradiated samples.

Figure 6.20. Effect of irradiation dose on the effective magnetic anisotropy energy (K_{eff}).

A decrease in the K_{eff} shows reduction in the perpendicular magnetic anisotropy energy density. The reduction of K_{eff} with increase of irradiation dose is a direct repercussion of enhanced disordering at the interface between ferromagnetic layer and Pt [12, 13], as corroborated by the structural studies. Noticeably, the value of K_{eff} remained positive for the used irradiation dose. The reduction in K_{eff} infers a lower periodicity of domains (assuming that the DMI and exchange energy coupling remains constant). Further, uniform anisotropy magnetic energy density K_u was calculated using the formula [14, 15] $K_u = K_{eff} + \frac{\mu_0}{2} M_s^2$, where $\frac{\mu_0}{2} M_s^2$ represents the magnetostatic energy density. Figure 6.21a shows the variation of K_u as function of irradiation dose. Magnetostatic energy of irradiation doses is also plotted in figure 6.21a. A substantial contribution of magnetostatic energy density can be noticed for calculated K_u . However, for annealed sample, K_u does not show such substantial contribution from magnetostatic energy density (figure 6.21b). The difference in the magnetic uniform energy densities of irradiated and annealed samples can be understood due to a weak K_{eff} in irradiated sample, which was increased with annealing treatment of samples.

Figure 6.21. Uniform anisotropy energy density (K_u) and Magnetostatic energy density of irradiated (a) and annealed samples (b).

Further, DMI was measured using Brillouin light scattering (BLS) spectroscopy, an essential property for skyrmion [16, 17]. An increase in the DMI will give rise to a high density of

skyrmion and facilitate higher thermal stability. The difference in stoke and anti-stoke frequencies (Δf) measured for a wavevector of $16.7\mu\text{m}^{-1}$, which gives a ratio of DMI over M_s , is shown in figure 6.22a. The Δf stayed almost constant until $8\times$ dose and was noticed to be reduced for the highest dose $12\times$. The measurement of DMI (D), based on formula taken from references [17-19] was used as $\Delta f = \frac{2\gamma D k_{sw}}{\pi M_s}$, where k_{sw} is spin wave vector and γ is the gyromagnetic ratio.

Figure 6.22. Measurement of DMI using BLS. (a) Frequency difference in stoke and anti-stoke frequencies (Δf) as a function of irradiated dose. Red dotted lines show the range of error in the measured value. (b) Measured DMI for different irradiated doses.

In place of M_s , the in-plane saturation magnetisation of irradiated samples was used in the formula to calculate D . Noticeably, a maximum value of DMI was found for the $8\times$ dose sample to be 1.94 mJ/m^2 (figure 6.22b). However, one might argue that the effective thickness might vary due to intermixing of Co and Fe atoms in Pt, causing discrepancies in the calculated values of M_s and affect the calculated D values. Therefore, interfacial DMI (D_{int}) was also calculated by using $M_s \times t$ in the DMI formula [20, 21]. Noticeably that the trend of D_{int} is similar to the trend of D , showing a maximum value of 0.97 pJ/m for the $8\times$ irradiated dose samples. Similar behavior of D or D_{int} due to interface intermixing has been reported in previous reports [21-23]. Noticeably, we have noticed that the D_{int} trend in our irradiated

samples is following the trend of $M_s \times t$, which is different from the previous report [21]. The changes in magnetic properties induced by the intermixing will affect the properties of magnetic textures.

6.5.4 Effect of irradiation on magnetic textures

To understand the effect of irradiation on magnetic textures, MFM was carried out. Samples were saturated with a magnetic field of 300 mT before imaging the samples. Figure 6.24 shows the MFM image of samples at remanence, which depicts an increase in the density of stripe domains with irradiation dose. The periodicity of magnetic stripes was calculated using 2-dimensional FFT.

Figure 6.23. MFM images of irradiated samples measured at zero field.

The calculated periodicity of the irradiated sample is shown in figure 6.25. The value of periodicity was decreased by 51 nm from 168 nm to 117 nm. Unlike the annealing treatment, irradiation has led to an increase in the density of stripes. The decrease in periodicity is a promising indicator for the increased density of skyrmions. Hence, the variation in properties

viz. K_{eff} (decreased with irradiation) and DMI (increased with irradiation) has led to the desired state of dense stripe domains.

Figure 6.24. Periodicity trend of irradiation samples as a function of dose

Figure 6.25. FORC of various irradiated samples. Saturation Field $\mu_0 H_s$ is highlighted by the yellow dotted lines [24]. Changed colour in the plot shows a finite value of ρ indicating an irreversible magnetic switching [25-27] (a) Pristine sample has $\mu_0 H_s$ of 61.3 mT (b) Sample with 4×10^{18} ions/m² irradiation dose has $\mu_0 H_s$ of 88.4 mT. (c) Sample with 8×10^{18} ions/m² irradiation dose has $\mu_0 H_s$ of 91.7 mT. (d) Sample with 12×10^{18} ions/m² irradiation dose has $\mu_0 H_s$ of 96.3 mT.

The MFM images of pristine and irradiated samples are shown in figure 6.26. Samples were saturated out of plane direction first and held at an applied magnetic field (H_s) during imaging. This step was repeated for every single measurement.

A high skyrmion density was observed for irradiated samples. However, the field of highest skyrmion density achieved was different. Images with the red dotted lines represent the highest density of skyrmions observed in their respective irradiation dose. It can be noted that the field at which the highest density of skyrmions was observed increases with the irradiation dose. From these images, other properties of skyrmions, viz. density, diameter, and eccentricity were also measured.

Figure 6.27. Skyrmion properties of irradiated samples. (a) Skyrmion density of irradiated samples. (b) Skyrmion diameter of irradiated samples. (c) Eccentricity of skyrmions after irradiation.

Figure 6.27 shows various physical properties of skyrmions measured from the selected images. Skyrmions were filtered free of artefacts, and the properties were measured by fitting every skyrmion with a 2-dimensional Gaussian. The skyrmion density was found to increase by almost five-fold, reaching up to $51 \text{ skyrmion}/\mu\text{m}^2$ (shown in figure 6.27a). Interestingly,

skyrmion diameter decreased by 69 nm, reaching from 148 nm to 79 nm (figure 6.27b). The eccentricity of skyrmion was measured by $e = \sqrt{1 - \frac{b^2}{a^2}}$, where a represents the major axis and b represents the minor axis. The eccentricity was reduced after irradiation, representing an increase in skyrmions' circularity at higher doses(figure 6.27c).

6.5.5 Effect of irradiation on domain wall energies

The increase in the skyrmion density is a promising result achieved by tweaking the interfacial properties via interface intermixing. We investigated the effect of irradiation dose on domain wall energy to get more insight into the phenomenon causing such an increase in the skyrmion density. To calculate the exchange coupling energy density (A_{ex}), micromagnetic simulations were used. An effective model of eight repetition layers was performed [28]. Magnetic properties of respective samples measured from VSM, AGM, and BLS were used. The ratio of DMI over A_{ex} was varied to achieve a relaxed magnetic state at zero field [29].

Figure 6.28. Magnetisation profiles of the pristine sample with various values of A_{ex} . The size of the simulation window is $1.5 \mu\text{m} \times 1.5 \mu\text{m}$.

The density of magnetic textures shown in figure 6.28 decreases with an increase in the A_{ex} value. The trend of periodicity calculated from the simulated magnetisation profiles (shown in the figure 6.29a) was fit by a binomial function and interpolated to estimate the value of A_{ex} for which experimental periodicity is equal to the simulated periodicity. Figure 6.29b shows the variation of A_{ex} for different irradiation doses. Noticeably, the trend of A_{ex} is similar to the trend of M_s , which is also followed by DMI (insight on the trend of M_s is given in the next section).

Figure 6.29. The plot of periodicity trend for different A_{ex} values of pristine and irradiated samples. (b) Estimated exchange coupling energy A_{ex} values for different irradiated samples. The coloured yellow area shows the range of error in the estimated value.

Once the value of A_{ex} is estimated, we proceeded to calculate the domain wall energy, using the formula $\sigma = 4\sqrt{K_{eff}A_{ex}} - \pi D$ [15]. Figure 6.30 shows that the domain wall energy decreases with an increased irradiation dose. The decrease in the domain wall energy with higher doses supports the formation of more domain walls. Since the number of domain walls formed at lower domain wall energy increases, the density of skyrmions also increases.

Figure 6.30. Variation in domain wall energy due to irradiation at different doses.

6.5.6 Magnetisation trend due to irradiation induced structure changes

The trend of magnetisation (M_s) has been seen to influence the trends of DMI, A_{ex} , and K_u . Therefore, it is vital to understand the reason behind the observed behaviour of M_s . To achieve an understanding of the trend of M_s , Time of flight Secondary Ion Mass Spectroscopy (ToF-SIMS) was carried out on the irradiated samples.

Figure 6.31 shows the ToF-SIMS plots of irradiated 8× and 12× samples. The first row of figure 6.31 compares the elemental intensities of reference and 8× dose sample. Intensities of B, Co, and Fe for pristine and 8× dose samples are shown in figures 6.31a,b, and c. Dotted lines in the plots depict interfaces of MgO-CoFeB and CoFeB-Pt. Intensities of plots are expressed in arbitrary units. No difference in the intensities of Co, Fe, and B was noticed. Negligible change in the peak of Co and Fe infers the intactness of these atoms in the ferromagnetic layer. Figure 6.31d shows the normalized intensities of Co, Fe, and Mg.

Intensities of pristine samples are shown with dotted lines. The peak position of Mg in the irradiated 8× sample is the same as the peak position of Mg in the pristine sample. Hence, we concluded that Mg does not intermix into the CoFeB layer. Further, a 12× sample was also analyzed. Like an 8× dose sample, no difference in the intensity of the B peak was observed (figure 6.31e). The constant B peak intensities in 8× and 12× irradiated samples infer a possibility of B migration at interstitial sites of Co and Fe, which can also increase the magnetisation of the ferromagnetic layer. However, intensities of Co and Fe have been reduced (figure 6.31f,g), inferring the migration of Co and Fe from the ferromagnetic layer to the adjacent layers. Moreover, we have seen a shift in the peak of Mg in the 12× dose sample. The Mg peaks were fit for the pristine and 12× sample. A shift of Mg peak by ~0.1 nm towards CoFeB was found (figure 6.31h). This shift infers the mixing of Mg peaks into the ferromagnetic layer. The mixing of a non-magnetic (Mg) atom into a ferromagnetic layer can be understood as the reason for the decrease in the M_s of 12× sample.

Figure 6. 31. Time of Flight Secondary ions mass spectroscopy (ToF-SIMS) profiles of B, Co Fe, relative intensities of 8x and 12x irradiated samples compared with respective intensities of pristine sample.

Figure 6.32. Time of Flight Secondary ion mass spectroscopy (ToF-SIMS) profiles of B, Co Fe in 300°C annealed samples .

As the ToF-SIMS did not show any significant change in the intensities of B, we decided to check if our ToF-SIMS tool can detect the variation of B. To answer this question, ToF-SIMS of the sample annealed at 300°C was carried out. In these samples, annealing gave rise to the migration of B (resulting in crystallization) and a corresponding increase of M_s . Hence, the ability of ToF-SIMS to detect B in this sample will confirm the efficiency of the technique to detect B. ToF-SIMS did show a reduction in the intensity of annealed sample B was observed (figure 6.32a), inferring the ability of our ToF-SIMS tool to detect the changes in the B. The reduction in the B peak can be explained due to the migration of B away from the ferromagnetic layer [30]. Moreover, a reduction in Co and Fe peaks (figure 6.32b,c), which can be seen shifting into Pt, was also observed. In addition, a shift of Mg peak into the ferromagnetic layer

was observed for the 300°C annealed sample (figure 6.32d). This shift of Mg peak into ferromagnetic layer might be the reason that 300°C annealed sample has a lower M_s .

6.6 Summary

In summary, two different methods were explored to alter the atomic arrangements and vary the density of skyrmions. Although the energy of annealing (ranges in meV) is different from irradiation (keV), the two methods introduce displacements of atoms at the interface. The results of the two used methods were strikingly different for effective magnetic anisotropy energy density. The magnetic properties showed a different trend for the two methods. Resultingly, the change in density of skyrmions due to these two methods of treatments was different.

Nevertheless, irradiation of the sample has led to an increase in the density by almost five folds and a reduction in the skyrmion size by 70 nm. Hence, the irradiation method can be used to increase the density of skyrmions. A high dense skyrmion state is promising for achieving skyrmion-based racetrack memories and computing devices.

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Chapter 7 Perspective: what is next?

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This chapter gives a summary and future perspective of the studies reported in this thesis. The primary purpose of this chapter is to take readers of this thesis through the key points and our outlook on the carried-out studies. Besides this, I have given different future investigations, which the reported studies can persuade.

7.1 Summary

One-dimensional devices have been widely used in rigid electronics to flexible electronics to soft stretchable electronics. These devices are not only limited to electronics but also has been extended to spintronics devices. In spintronics, one-dimensional devices have been proposed for various applications. This thesis emphasizes three such one-dimensional devices which can be used for various applications like energy harvesting, microwave generation, and computing, using micromagnetic simulations and experimental investigations. These studies have been reported in three chapters.

In **chapter 4**, a study on energy harvesting has been done. In this study, we have used an FeCo alloy, which exhibits a high magnetostriction behaviour. External mechanical stress changes the direction of magnetic stress, leading to a variation in the magnetic properties. In a specially prepared FeCo trilayer stack, we have noticed the rotation of the easy axis to the hard axis and the hard axis to the easy axis on the application of stress. This change was also visible in the magnetic domains, where the direction of magnetic domains was changed under stress. We fabricated FeCo microwires on flexible polyimide films. The deposition of these microwires was carried out by a facing-targets sputtering, which introduced fringing field during the deposition. Using this fringing field, the direction of magnetic domains was set transverse to the direction of the length of microwires. The formation of magnetic domains in the transverse direction maximizes the number of domains in the microwire. We have fabricated a pick-up coil around the microwire. To mimic the ambient vibration, we induced vibrations in the waveform. We have demonstrated energy generation in the order of μJ in a time span of $50\ \mu\text{s}$.

In **chapter 5**, a computational demonstration of motion of domain structures through energy barrier constrained nanowire has been done. This chapter reports two studies- one with magnetic domains and the second with magnetic skyrmions. For both studies, magnetic properties of a Co/Pt-based material stack were used. The first study was done on the motion

of magnetic domains through a nanowire with pinning sites. The pinning sites were required to sustain a controlled motion of domain walls in the wire. In this study, we have shown that moving oppositely magnetized domains back and forth through a reading sensor can generate signals with frequency in GHz. The frequency of generated microwaves can be tweaked by varying the velocity of domain walls and width of domains. The second study in this chapter offers a technique to guide the motion of magnetic skyrmion in a nanowire. Our proposed method is based on introducing energy barriers at the edges of the nanowire. The introduction of an energy barrier was carried out by reducing the value of DMI, which resultingly increases the domain wall energy of the regions. Moreover, we have noticed that the skyrmion Hall effect, which results in the annihilation of skyrmions, was suppressed due to this energy barrier. Noticeably, after introducing the energy barriers, skyrmions moved along the energy barrier. Furthermore, in this study, we have demonstrated a motion of multiple skyrmions in parallel tracks. The energy barriers separated these tracks. In the end, we have carried out a study on the magnetostatic repulsion of skyrmions in a parallel track. Out of the two investigations in this chapter, we decided that the motion of skyrmions in such DMI modified tracks is worth investigating. Therefore, we proceeded to carry out experimental investigations on the modification of magnetic properties in Skyrmion systems.

In **chapter 6**, we have reported a study that emphasizes the modification of magnetic properties and, consequently, physical properties of skyrmions using interface intermixing. This study used a Pt/CoFeB/MgO stack, which could show an unhindered skyrmion motion due to its amorphous nature. To carry out interface intermixing, we have chosen to irradiate He^+ ions. The irradiation of light element ions induces roughening of the interface and intermixing of atoms at the interface. As the intermixing varies linearly with the ion dose, we have varied the dose of ion irradiation. On carrying out the structural characterization, we have confirmed the intermixing of atoms at the interface. This intermixing has exciting outcomes in the obtained

results. The interface magnetic properties like effective anisotropy and DMI have a direct influence from interface intermixing. Besides the interface properties, bulk magnetic properties like saturation magnetisation and exchange coupling energy were also modified. The alloying of CoPt and Boron migration led to an increase in the saturation magnetisation.

Interestingly, the exchange coupling and DMI followed the trend of saturation magnetisation. The changes in the magnetic properties led to a decrease in the domain wall energy, and hence, the physical properties of magnetic texture were also changed. We have noticed a reduction in the periodicity by 50 nm and the skyrmion size by 70 nm after irradiation. Interestingly, the skyrmion density was also increased by almost five-fold, reaching up to 50 skyrmions per μm^2 .

7.2 Outlook and Future direction

A continuation of this thesis implies working on achieving the improvements in the reported studies. Here, I have listed out the stance on the application and future investigation for carried-out studies. These suggestions will be beneficial for the expansion of reported studies.

Self-Energy Harvesting IoT devices: IoT devices sense and record/transmit the information continuously or intermittently. These devices may be placed in inclement conditions or other places, where reaching them consistently is inconvenient. The power required to run these devices comes from a storage battery that needs to be recharged or replaced regularly. Therefore, it would be irrational to depend on conventional batteries. Such IoT devices should be self-powered. In this connection, we have demonstrated that FeCo based microwire can be used for harvesting energy. We were able to harvest energy in order of μJ in a mere span of a few μs . If we have an array of such devices in cascades, it will enhance the total energy generated in the same time span. The advantage of our proposed device is that it harvests energy

from ambient vibrations. Therefore, it can also work during nighttime and daytime when the weather is cloudy, which does not happen if a device is hooked to solar cells.

- Shape-dependent effect of stress: The sample studied in the investigation consists of a long straight patterned microwire. The effect of externally applied stress will vary depending on the shape of the material. Hence, different shapes will lead to a difference in the rotation of domains. We propose that studying different shapes may provide a better insight into how the shape-dependent effect of stress and variation in the magnetic flux can be enhanced. A more significant change in the magnetic flux will lead to the generation of high energy.
- Effect of long-term stress: The proposed device for energy harvesting in an actual condition will go through stress million times. Hence, there is a possibility that the structure properties might change after being under stress for a long time. The changes in the structure will lead to altering the stress-induced behaviour of the magnetic properties. Hence, there is a need to study the effect of long-term stress-induced effect. Long-term stress can be applied by keeping the sample under vibrations for a long time (a day). We propose to carry out a structured study using the X-Rays diffraction characterization technique. Consequently, magnetic properties can be investigated after long-term stress.
- Increasing the thickness of ferromagnet: Increasing the thickness of ferromagnet leads to an increase in the magnetic flux. In our study, we have investigated the effect of thickness till a particular value. The flux of magnetic films will further increase with the increase in thickness until the thickness becomes large enough to favour the formation of vortex magnetic domains. Therefore, a study investigating the limit of maximum thickness of the ferromagnetic film is essential. An optimized thickness of

the film will generate a maximum difference in the flux under stress. Hence, the output efficiency of devices will be enhanced.

Generation of microwaves with GHz frequency: Microwave works as a line-of-sight communication technology. Microwaves provide a high wireless speed that can be used for sending data. Microwave links are slight wavelength radiation, pointing directly in narrow bandwidth at a receiving antenna. Our study demonstrated that high-frequency microwaves could be generated by moving domain walls back and forth in a nanowire. The generated frequency of microwaves can be calculated as $frequency = v/2 \times w$ (here v is the velocity of domain wall motion and w is the width of domains). Hence, by increasing the velocity, we have achieved a high frequency of 5.18 GHz. Any further increase in frequency is restricted by Walter breakdown, limiting the velocity of the domain wall to a certain number. Besides this, we have observed a broad bandwidth of MHz. Such broad bandwidth is a drawback for communication as it leads to interference of other signals. On investigation, we have found that the broadness in the bandwidth is due to oscillations in the domain walls during motion. These oscillations are inevitable, and hence, the proposed method fell short in providing a successful route for generating microwaves.

- Reduction of the bandwidth: The method used to generate microwaves in the study gave a higher amplitude value for the generated microwave than reported magnetism-based methods. However, the bandwidth of generated microwave was in the range of MHz. The required bandwidth of generated microwave is very narrow (in the range of kHz). Therefore, reducing the bandwidth of microwaves to match the need for narrow bandwidth for IoT applications. The broadness in the width of generated microwave

arises due to the randomness in the size of domains during the motion of domain walls. Hence, to reduce the bandwidth of generated microwaves, we must control the randomness in the size of domains during the domains wall motion. We have tried different studies related to the physical properties of nanowires and domains. However, we were unable to notice any decrease in the bandwidth of generated microwave. As a next step, we propose to perform the study on other materials. The focus of the study would be on the reduction of bandwidth for generated microwave signals.

- Using a circular wire geometry, we used a one-dimensional wire in our proposed design, where magnetic domains were moved back and forth. A spin current was applied in both directions to achieve bi-directional motion of domain walls, which led to the inhomogeneity in the shape of domains, resulting in increased bandwidth. To cope with this, I propose the employment of a circular wire. In the circular geometry, current will be needed to apply in one direction. The unidirectional current will reduce the inhomogeneity in the shape of the domain wall.

The guided motion of multiple skyrmions: Skyrmions, due to their small size and capability of circumventing defects during motion, are popular candidates for memory and computing devices. Unlike domain walls, the current-induced motion of skyrmions results in an unwanted transverse motion and annihilation (caused by the skyrmion Hall effect). This effect is famously known as the skyrmion Hall effect due to a resemblance to the charge in the Hall effect. We have reported a method to suppress the skyrmion Hall effect by introducing energy barriers on the edge of the nanowire. These energy barriers prevent the skyrmions from reaching the edge and cause skyrmion motion along the barrier. Hence, our study confirmed that the energy barrier could control the direction of skyrmion motion. Besides, we also proposed that the size

of the skyrmion can be shrunk by squeezing the skyrmion in between an energy barrier. However, the motion of a skyrmion in squeezed state reduces. Nevertheless, introducing multiple skyrmion energy barriers with a specific pitch and nucleating skyrmions between these barriers have provided a route to move multiple skyrmions in the same geometry. We have demonstrated multiple simultaneous motions of skyrmions in these geometries, which in our point of view has potential for dense devices.

- The motion of skyrmions in a grained nanowire: In the simulation study on the guided motion of skyrmions, we did not account for the effect of magnetic grains on the dynamics and size of skyrmions. Since most of the films used in the experimental study are polycrystalline, small grains and grain boundaries are prevalent. These grain boundaries possess different magnetic properties than the grains. Therefore, the change in the magnetic properties at grain boundaries affects the size and dynamics of skyrmion. In addition, the size of grains with respect to the skyrmion's size also influences the skyrmion's behaviour in track. These effects, which will be prominent in the experiments, will influence the behaviour of skyrmions. As a next step, we would like to include the effect of grains in the studies.
- The effect of varying all magnetic properties: In our micromagnetic simulation, we have only varied the DMI, resulting in the variation of domain wall energy. The difference in energy acts as a barrier that abstains skyrmion from the skyrmion Hall effect. An experimental technique to achieve such variation in the properties will not reduce DMI only. Other magnetic properties such as K_u and M_s will also change. Hence, it is crucial to study the variation of DMI in combination with other magnetic properties. The change in the magnetic properties will also affect the skyrmion dynamics and physical properties of skyrmions. Therefore, an extensive study needs to be done.

Formation of a dense skyrmion nucleation site: A dense skyrmion state is a favourable state for high-density memory and synaptic applications. We have demonstrated that a highly dense skyrmion state can be achieved by varying the interfacial properties in a specific ratio. The increase in skyrmion density can be construed due to a reduction in the domain wall energy. The combination of these properties can create a high-density nucleation region at the beginning of a nanowire. An application of current in such nanowire can move skyrmion. Secondly, the difference in the domain wall energy, due to the variation of magnetic properties, can be used as a route to creating energy barriers on the edges of the nanowire. Such an energy barrier can be created by irradiating ions at the edges of a nanowire. The decrease in the effective anisotropy and increase in the DMI achieved after irradiation will lead to a higher domain wall energy on the edges. Hence, the proposed scheme of guided skyrmion motion can be achieved.

- Energy-dependent irradiation study: The achieved high skyrmion state was persuaded by irradiation He^+ ions. In the study, we have chosen the irradiation energy, which settles the ion deep into the substrate. The depth of ions is 120 nm which is quite profound. However, the total thickness of the films stack is 48 nm, implying that the energy of irradiation can be reduced. By varying the energy, the displacement of atoms can be varied, which comes from a primary collision. Hence, we propose to investigate energy-dependent irradiation. The effect on magnetic properties will be different; hence, the properties of skyrmions would also vary accordingly.

Cohesive applications of the reported studies:

Here, I have tried to discuss a broader context in which the reported works can be overarched:

Stress-induced nucleation and annihilation of skyrmions: The FeCo alloy, which shows good magnetostriction properties (demonstrated in chapter 4), can also be studied for skyrmion nucleation. Depositing a set of layers, which includes a seed layer/thin FeCo layer/a heavy-metal layer (Pt), on the top of flexible polyimide film, would be a rational point to start this study. In any case, the application of stress will introduce an in-plane magnetoelastic anisotropy. The in-plane component can help to stabilize the skyrmion at room temperature, as mentioned in chapter 2. Hence, the application and removal of stress to such film will introduce and diminish in-plane components, respectively. Therefore, the stress can control the stabilization and annihilation of skyrmion.

Use of irradiation modified regions to demonstrate the suppression of the skyrmion Hall effect: A mask can be used to select the region of interest, which can be exposed to the irradiation. The irradiated region will have modified magnetic properties, as demonstrated in chapter 6. The irradiated region next to the non-irradiated region will give rise to an energy barrier. Such an energy barrier will be used to suppress the skyrmion Hall effect (shown in chapter 5). The irradiation dose and energy can be varied at the irradiated regions. The current-driven dynamics of skyrmions can be carried out once the energy barriers are generated. The study on the suppression of the skyrmion Hall effect will be promising for the computing and memory applications of skyrmions.