Dynamic response of magnetic skyrmions in multilayer heterostructures

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DYNAMIC RESPONSE OF MAGNETIC SKYRMIONS IN MULTILAYER HETEROSTRUCTURES

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SCHOOL OF PHYSICAL AND MATHEMATICAL SCIENCES

2020

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SCHOOL OF PHYSICAL AND MATHEMATICAL SCIENCES

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

2020

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 a professional qualification to any other university or institution. I declare that this 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.

23-08-2019

Bhartendu Satywali

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23-08-2019

Prof. Christos Panagopoulos

Authorship Attribution Statement

This thesis contains material from 1 paper published in the arxiv, where I am listed as an author. Several of the results presented in chapter 4 are included in ‘Gyrotropic Resonance of individual Néel skyrmions in Ir/Fe/Co/Pt multilayers’, arXiv:1802.03979v1, 2018.

The contributions of the co-authors are as follows:

- Prof. Christos Panagopoulos conceived and coordinated the project.
- Dr. M.Raju prepared the samples.
- I performed the magnetometry and FMR experiments. For FMR experiments I received assistance from Dr. Shikun He.
- Mr. Nghiep Khoan Duong and I performed the MFM imaging experiments. I analysed the images.
- Dr. Fusheng Ma performed micromagnetic simulations.
- Dr. Volodymyr Kravchuk and Dr. Markus Garst performed the analytical calculations.
- I analysed all the experimental data. For simulations and analytical calculations of skyrmion eigenmodes, I received inputs from Dr. Fusheng Ma and Dr. Volodymyr Kravchuk.
- I and Dr. Fusheng Ma wrote the initial versions of the manuscript. The submitted version contains input from all the co-authors.

23-08-2019

Bhartendu Satywali

Abstract

Topological effects in condensed matter physics have been explored for fundamental and technological reasons in a wide variety of materials. Recently, magnetic skyrmions - circularly-shaped twisted spin textures - have aroused interest due to their enhanced stability and efficient coupling with electric currents. Consequently, they can be harnessed for data storage and logic operations. Two key requirements for skyrmion formation are (a) an absence of spatial inversion symmetry and (b) a large spin-orbit coupling in the material. Metallic multilayers featuring ferromagnet-heavy metal (FM-HM) interfaces satisfy both conditions and can hence exhibit nano-skyrmions at room temperature (RT). In this thesis, we nucleate sub-50 nm skyrmions at RT in engineered Ir/Fe/Co/Pt multilayers, where the size and order (lattice or isolated) of the skyrmions can be tuned by the relative thickness of the Fe/Co layers. We employ magnetic force microscopy (MFM) to image skyrmions at RT. A significant variation is observed in the size and shape of skyrmions in different regions of the film, suggesting local inhomogeneities in the magnetic parameters for our films.

A detailed analysis of the microwave excitation modes of skyrmions is of profound importance for understanding their current and field-induced dynamics. In this thesis, we report the first experimental detection of magnetic resonance from Néel skyrmions in multilayers from 300K-100K, measured with a home-built broadband ferromagnetic resonance (FMR) probe. The field and frequency-sweep FMR measurements reveal two distinct resonant modes originating from skyrmions: a low frequency (LF) resonance exhibiting a steep proportionality to the external out-of-plane magnetic field (H), and a high frequency (HF) resonance which is less sensitive to variations in H . Combining micromagnetic simulations and analytical calculations, we identify the counterclockwise (CCW) skyrmion gyrations to be responsible for the LF mode. The HF mode is caused by the scattering of magnons from the uniformly-precessing ferromagnetic background off skyrmions. Furthermore, we present a method of tuning the skyrmion excitation frequencies by varying the thickness of the non-magnetic Pt/Ir spacer layers in our heterostructures, which modulates the dipolar coupling between the FM layers.

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Abbreviations

FM-HM	Ferromagnet-Heavy Metal
MFM	Magnetic Force Microscopy
FMR	Ferromagnetic Resonance
LF	Low Frequency
HF	High Frequency
CW	Clockwise
CCW	Counterclockwise
CMOS	Complementary Metal-Oxide-Semiconductor
EUV-IL	Extreme Ultraviolet-Interference Lithography
OP	Out-of-Plane
CGS	Centimetre-Gram-Second
SOC	Spin-Orbit Coupling
PMA	Perpendicular Magnetic Anisotropy
DW	Domain Wall
DMI	Dzyaloshinskii-Moriya Interaction
SP-STM	Spin-Polarised Scanning Tunnelling Spectroscopy
AHE	Anomalous Hall Effect
LTEM	Lorentz Transmission Electron Microscopy
GMR	Giant Magnetoresistance
HDD	Hard Disk Drive
SSD	Solid State Drive
NCMR	Non-Collinear Magnetoresistance
STT	Spin Transfer Torque
SHE	Spin Hall Effect
LLG	Landau-Lifshitz-Gilbert

YIG	Yttrium Iron Garnet
VNA	Vector Network Analyser
CPW	Co-Planar Waveguide
TL	Transmission Line
RF	Radio Frequency
DUT	Device Under Test
FWHM	Full-width half maximum
ELC	End Launch Connectors
SANS	Small Angle Neutron Scattering
IP	In-Plane
BLS	Brillouin Light Spectroscopy
DE	Damon-Eshbach
MPMS	Magnetic Property Measurement System
MTXM	Magnetic Transmission X-ray Microscopy
BR	Breathing
NDD	Nonreciprocal Directional Dichroism
STTD	Spin-Transfer Torque Diode
MTJ	Magnetic Tunnel Junction
PBC	Periodic Boundary Condition
PVD	Physical Vapour Deposition

Symbols

H	Magnetic field
J	Heisenberg exchange coupling parameter
S	Spin moment
A	Exchange stiffness
M	Magnetization
T	Temperature
H_{SO}	Spin-Orbit hamiltonian
g_e	Electron g factor
e	Electron charge
m_e	Electron mass
c	Velocity of light in vacuum
V_n	Electric potential from atomic nucleus
λ_{nl}	Spin-orbit coupling strength
K	Uniaxial anisotropy constant
$\hbar$	Reduced Planck's constant
N	Demagnetization facotr
H^d	Demagnetization field
E^d	Demagnetization energy
μ	Permeability of material
μ_0	Permeability of free space
N	Demagnetization tensor
H^e	Applied magnetic field
D	Dzyaloshinskii-Moriya interaction parameter
E_{DM}	Dzyaloshinskii-Moriya interaction energy
E_F	Fermi energy

k_F	Fermi wave vector
λ_d	Spin-orbit coupling strength for conduction electrons
Z_d	Atomic number
w	Generalised form of DMI
P	Lifshitz invariant
E	Energy density
m	Normalised magnetisation
Δ	Domain wall width
τ	Decay parameter
k_B	Boltzmann Constant
L	Orbital angular momentum
S	Spin angular momentum
γ	Gyromagnetic ratio
χ	Magnetic susceptibility
α	Damping constant
g	Landé g factor
f	Frequency
ω	Angular frequency
ϵ_{eff}	Effective dielectric constant
Z	Impedance
T	Transmission coefficient
q_T	Topological charge density
Q	Topological number
G	Gyroscopic vector

Chapter 1

Introduction

The demand for data storage and information processing has grown in diverse ways over the past decade. While cloud technologies require large capacity data storage devices with ‘always-on’ status for continuous synchronisation, mobile devices demand energy efficient operation in a compact package.

Computational operations can be broadly divided into two categories [1]: (a) microprocessors to perform arithmetic and logical operations, and (b) memory units to store the data bits.

The majority of microprocessors are based on complementary metal-oxide-semiconductor (CMOS) technology developed for silicon. To date, the rapid advancement of silicon-based technology has relied on the miniaturisation of devices. This is supported by parallel advances in lithography techniques. For example, extreme ultra violet-interference lithography (EUV-IL) allows the fabrication of nanostructures with sub-10 nanometer (nm) spatial resolution (figure 1.1a), enabling denser transistor assemblies (≈ 100 MTr/mm²) in electronic devices.

The annual rise in transistor density is approximated by Moore’s law [2], which predicts a two-fold increase in transistor density after every two years. Following Moore’s law, the size of the transistors will be reduced to 2-3 nm in the next few years. Electron motion at this length scale is governed by quantum uncertainties (such as electron leakage through an insulating barrier), rendering such small devices unreliable for operations. An additional challenge emerges from the Joule heating of devices, which is intrinsically associated with electron motion. For chips containing millions of transistors this can produce a significant amount of

heat. Heating is an undesirable side effect as the performance of silicon degrades at higher temperatures. Another undesirable outcome of Joule heating is the restriction which it imposes on the clock speed of the processor. In the past few years, processing speeds have remained roughly constant at 1.5-4 GHz (figure 1.1b). Therefore, even though existing miniaturisation supports a large transistor density, the heating problem limits the maximum performance of CMOS devices.

The solution to increasing demand for computational power is to develop new paradigms which can support the scaling of electronic devices beyond the predictions of Moore's law, while minimising heat dissipation. Spintronic technology harnesses the charge and spin of an electron to provide a viable alternative to conventional charge-based silicon electronics. Computation in spintronics relies on flipping of spin orientations among the bistable ground states. Spin reversal can be achieved by electrical techniques (spin torques or Oersted field) and/or magnetic mechanisms (spin-waves and spin currents).

The maximum heat dissipation occurs during the transfer of data bits from micro-processor to memory unit [3]. Current-controlled manipulation of domain walls in racetracks [4] has been proposed as a fast and energy efficient alternative to existing technologies. However, the domain walls are sensitive to defects and impurities in the magnetic material, resulting in pinning of domain walls at defect sites. Consequently, a higher critical current is required for domain wall manipulation, raising the energy cost of the information handling process.

FIGURE 1.1: Current status of semiconductor electronics. (a) The 5 nm transistor unveiled by IBM in 2017 [5]. (b) The steady increase in transistor density (red) over the last few decades. The clock frequencies (blue) have remained roughly constant due to large heat dissipation [6] at higher clock speeds.

Magnetic skyrmions are circular quasiparticles with non-trivial topological properties [7, 8]. Their sub-100 nm size [9–11], stability against material defects and efficient coupling to electric currents [12, 13] make skyrmions excellent candidates for next-generation data storage and logic applications [14–17]. Magnetic skyrmions are formed due to competition between spin-aligning (Heisenberg exchange, uniaxial anisotropy etc.) and spin-canting (Dzyaloshinskii-Moriya, magnetic frustration etc.) interactions in ferromagnets. In the next sections, we will detail the magnetic interactions leading to the formation of skyrmions with distinct internal magnetizations, and the unique ‘topological’ properties of the skyrmions.

1.1 Magnetic Interactions in Ferromagnets

Heisenberg Exchange Interaction

The Heisenberg exchange interaction is a quantum mechanical phenomenon originating from the interaction of localised electrons in neighbouring atoms, which promotes collinear magnetic order. Quantitatively, the exchange interaction between a pair of spins S_i and S_j separated by a distance r can be expressed as

$$E_{ex} = - \sum_{\langle i,j \rangle} J(r)(\mathbf{S}_i \cdot \mathbf{S}_j) \quad (1.1)$$

$J(r)$ is the exchange coupling parameter, which in general decays rapidly beyond nearest neighbours. The neighbouring spins are arranged in parallel (ferromagnet) or antiparallel configurations depending on the sign of $J(r)$. A dot product in equation 1.1 indicates the isotropic nature of the interaction, where the energy scales with the relative orientation of the spins. Therefore, the exchange interaction does not support any preferential orientation of the magnetization. The functional form of the exchange interaction is given by $A(\nabla \mathbf{M})^2$ where A is exchange stiffness, which depends on the atomic structure of the ferromagnet. Typically, A is of the order of 10^{-11} Jm^{-1} .

Spin-Orbit Coupling

The spin-orbit interaction is a relativistic phenomenon which couples the spin and orbital motion of an electron. Electrons with high orbital velocity experience the electric field of the nucleus as a magnetic field in the electron’s frame of reference,

which acts on the electron spin. The corresponding Hamiltonian is [18]

$$H_{so} \approx \frac{g_e}{2} \frac{e}{m^2 c^2} \frac{1}{r} \frac{\partial V(r)}{\partial r} \mathbf{s} \cdot \mathbf{l} \quad (1.2)$$

where $g_e \approx 2$ is the electron g factor. e and m are the charge and mass of an isolated electron respectively, $V(r)$ is the electric potential from the nucleus, s and l are the spin and orbital momenta of the electron. The spin-orbit coupling for a given orbital is given by the matrix element of the Hamiltonian and the radial wave function of the orbital $\Phi_{nl}(r)$: it is estimated by

$$\lambda_{nl} \approx \frac{g_e}{2} \frac{l}{2} \frac{\hbar^2 e}{m^2 c^2} \int_0^\infty dr \Phi_{nl}^2(r) \frac{1}{r} \frac{\partial V(r)}{\partial r} \quad (1.3)$$

λ_{nl} is the spin-orbit coupling strength for orbital s with quantum numbers n and l . The nuclear potential $\frac{\partial V(r)}{\partial r}$ increases with the atomic number, and hence heavier atoms tend to exhibit larger spin-orbit effects.

Magnetostatic Interactions

Short range exchange interactions favour a collinear orientation of all the atomic moments in a ferromagnet. On longer length scales (distances longer than a few atomic sites), magnetic moments interact via ‘magnetostatic’ interactions, which often stabilise canted structures like spin helices and bubbles. In nanostructures, magnetostatic interactions emerge as a demagnetization energy, originating from the magnetic charges on the surface. For example, in an out-of-plane (OP) magnetized thin film, opposite charges appear on the top and bottom surfaces (figure 1.2a). The magnetic field due to the surface charges is oriented opposite to the applied field and is referred to as the ‘demagnetization field’ H^d . H^d can be correlated with the magnetization by $H^d = \mu N \mathbf{M}$, where μ is the permeability of material, N is the demagnetization tensor and $\mathbf{M}$ is the net magnetization. The precise form of N depends on the geometrical shape of the material. In symmetric geometries, the off-diagonal elements of N are reduced to zero. For example:

- Plane: $N_x = N_y = N_z = 1$
- Sphere: $N_x = N_y = N_z = \frac{1}{3}$
- Infinite circular cylinder: $N_x = N_y = 1/2, N_z = 0$

FIGURE 1.2: (a) Origin of the demagnetization field $\mathbf{H}^d$, due to opposite surface charges on the top and bottom surfaces of a magnetic film. $\mathbf{M}$ is the net OP magnetization induced by an external field $\mathbf{H}^e$. (b) A and B are magnetic atoms and C is the bisection point of $\overrightarrow{AB}$. The plane passing through C and perpendicular to line $\overrightarrow{AB}$ is a symmetry plane for A and B.

For a thin film, the demagnetization energy is given by

$$E^d = -\frac{\mu_0}{2V} \int \mathbf{M} \cdot \mathbf{H}^d dv$$

where μ_0 is the permeability of free space and E^d is the anisotropy energy per unit volume. Solving with $N_z = 1$, $N_y = N_x = 0$ (for a perpendicularly magnetized film), $E^d = \frac{1}{2}\mu_0 M_s^2 \cos^2 \theta$, where the magnetization is assumed to be constant over the concerned volume ν_0 and aligned at an angle θ to the surface normal.

Magnetic Anisotropy

Magnetic anisotropy eliminates the spherical symmetry of magnetic order (induced by Heisenberg exchange) and imposes a directional preference based on the structural symmetry. The anisotropy can have diverse origins, ranging from the crystal structure to the geometry of the test material. The most ubiquitous form of magnetocrystalline anisotropy correlates the magnetization with the high symmetry axis of a crystal. In a solid, the charge distribution of an electron fluctuates as a function of the relative position and coordination number of the host atom. The spin-orbit interaction couples the electronic charge distribution with the magnetization. Consequently, the magnetization is bound to the crystal structure. This correlation between the magnetization and crystal symmetry imposes restrictions on the orientation of the magnetization.

In thin ferromagnets, shape anisotropy favours in-plane orientation of the magnetization, which minimises the induced surface charges, and thereby, reduces the

demagnetization field. However, it has been demonstrated that for thin ferromagnetic films in proximity to a heavy metal, the easy axis is oriented perpendicular to the film surface. This form of anisotropy is induced by a large spin-orbit coupling from the heavy metal and is referred to as perpendicular magnetic anisotropy (PMA).

The interplay of the isotropic exchange interaction, magnetic anisotropy, and long range magnetostatic energies leads to the formation of magnetic domains. Adjacent domains with distinct magnetization directions are separated by domain walls (DWs). DWs are non-collinear magnetic textures, where spins gradually rotate between the magnetization directions of connected domains. The precise nature of the spin rotation and the dimensions of the DWs are restricted by the relative strength of the various magnetic interactions. Typically, in bulk magnets and thick films, domains are separated by a ‘Bloch wall’ in which the magnetization rotates in a spiral fashion between the two ends of the wall. This form of magnetization rotation does not generate surface charges: $\nabla M = 0$, resulting in the absence of a demagnetization field.

For thin-film magnets in proximity to heavy metals, DW spins rotate in a cycloidal fashion. Such walls are known as Néel walls. A cycloidal rotation of the magnetization results in surface charges, generating stray fields. Therefore, Néel walls are energetically less stable than Bloch walls [19]. However, in the presence of magnetic interactions favouring a cycloidal rotation of spins, a Néel DW can be stabilised over a Bloch wall, depending on the relative strength of the magnetostatic and spin canting interactions. For example, the Dzyaloshinskii-Moriya interaction (DMI) at ferromagnet-heavy metal interfaces favours Néel walls and Néel-type skyrmions.

Dzyaloshinskii-Moriya Interaction (DMI)

The Dzyaloshinskii-Moriya Interaction (DMI) is a spin-canting interaction which was introduced as a phenomenological term [20] to understand the weak ferromagnetism in low symmetry crystals, such as α -Fe₂O₃ and carbonates of Mn and Co. The mathematical form was formulated on the basis of crystal symmetry and is written as

$$E_{DM} = - \sum_{\langle i,j \rangle} \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) \quad (1.4)$$

where i and j are neighbouring spins and $\mathbf{D}$ is the DMI vector.

As expected from the cross product in equation 1.4, the minimum energy configuration is achieved by an orthogonal alignment of neighbouring spins rotating around the vector $\mathbf{D}$. However, such an arrangement leads to a large exchange energy. Consequently, balancing the DMI with the exchange interaction stabilises magnetic textures where the magnetic moments of neighbouring atoms display a gradual angular deviation from the parallel axis. The magnitude and the direction of rotation are related to the orientation of $\mathbf{D}$, which in turn is controlled by the crystal symmetry.

Physical insights into the origin of the DMI were developed by Moriya in a two-body interaction model. Moriya [21] explained the origin of the anti-symmetric form of the DMI by including spin-orbit coupling in the theory of anisotropic superexchange interaction. A significant contribution of Moriya's work was the correlation of $\mathbf{D}$ with the structural symmetry of the material. The rules for determining the direction of $\mathbf{D}$ for two magnetic spins located at A and B, where C bisects $\overrightarrow{AB}$ (see figure 1.2b), are as follows:

1. When the centre of inversion is located at C, $\mathbf{D} = 0$
2. When the mirror plane perpendicular to $\overrightarrow{AB}$ passes through C, $\mathbf{D} \parallel$ to the mirror plane or $\mathbf{D} \perp \overrightarrow{AB}$
3. When there is a mirror plane including A and B, $\mathbf{D} \perp$ to the mirror plane
4. When a two-fold rotation axis perpendicular to $\overrightarrow{AB}$ passes through C, $\mathbf{D} \perp$ to the two-fold axis
5. When there is a n -fold axis ($n > 2$) along the $\overrightarrow{AB}$, $\mathbf{D} \parallel \overrightarrow{AB}$

In certain materials such as thin film heterostructures, the DMI can also be explained by a three-body interaction between two magnetic atoms and a nonmagnetic heavy metal, as proposed by Fert and Levy [22]. The presence of heavy metal impurities breaks the inversion symmetry between two spin sites, and spin-orbit scattering of conduction electrons by the nonmagnetic ion couples space and spin coordinates. The DMI between two spins at $\mathbf{R}_A$ and $\mathbf{R}_B$, mediated by the heavy metal (positioned at the origin, $\mathbf{R} = 0$) is calculated as

$$H_{DM} = -V_1 \frac{\sin[k_F(R_A + R_B + R_{AB}) + (\pi/10)Z_d]\hat{R}_A \cdot \hat{R}_B}{R_A R_B R_{AB}} (\hat{R}_A \times \hat{R}_B) \cdot (S_A \times S_B) \quad (1.5)$$

with

$$V_1 = (135\pi/32)(\lambda_d\tau^2/E_F^2k_F^2)\sin[(\pi/10)Z_d] \quad (1.6)$$

where λ_d is the spin-orbit coupling strength for the conduction electrons, E_F and k_F are the Fermi energy and Fermi wave vector, Z_d is the atomic number of the transition metal and $\mathbf{R}_{AB}$ is the separation between two spins. The interaction depends on the relative orientation of the spin cross product $\mathbf{S}_A \times \mathbf{S}_B$ to the direction of $\hat{\mathbf{R}}_A \times \hat{\mathbf{R}}_B$, and is responsible for the anisotropic nature of the DMI.

The relative orientation of $\mathbf{D}$ and $\mathbf{R}_{AB}$ limits the spin rotation to two directions. For $\mathbf{R}_{AB} \parallel \mathbf{D}$, the neighbouring spins rotate in a helical manner, where both in-plane and out-of-plane magnetization components rotate along the length of the structure. This is identical to the magnetization variation in Bloch domain walls. This form of DMI is observed in non-centrosymmetric magnets such as MnSi and FeGe. In the other case, $\mathbf{R}_{AB} \perp \mathbf{D}$, the out-of-plane magnetization of the neighbouring spins twists in a cycloidal fashion, while the direction of in-plane magnetization remains constant throughout the structure. The resulting structures are similar to Néel domain walls. Typically, this form of DMI originates at ferromagnet-heavy metal interfaces, and is hence sometimes referred to as ‘interfacial DMI (i-DMI)’.

Crystal Class	C_1	C_2	C_3
C _{nv}	1	0	0
D _{nv}	0	1	1
D _{2d}	0	0	0

TABLE 1.1: DMI coefficients for various symmetry classes in crystals.

A generalised form of the DMI which incorporates the crystal symmetry can be written as [23]:

$$w_D = C_1w_1 + C_2w_2 + C_3w_3 \quad (1.7)$$

where

$$w_1 = P_{zx}^x + P_{zy}^y = m_z \frac{\partial m_x}{\partial x} - m_x \frac{\partial m_z}{\partial x} + m_z \frac{\partial m_y}{\partial y} - m_y \frac{\partial m_z}{\partial y} \quad (1.8)$$

$$w_2 = P_{zx}^z - P_{zy}^z = m_z \frac{\partial m_x}{\partial y} - m_x \frac{\partial m_z}{\partial y} - m_z \frac{\partial m_y}{\partial x} + m_y \frac{\partial m_z}{\partial x} \quad (1.9)$$

$$w_3 = P_{xy} = m_x \frac{\partial m_y}{\partial z} - m_y \frac{\partial m_x}{\partial z} \quad (1.10)$$

The quantity

$$P_{ij}^k = m_i \frac{\partial m_j}{\partial x_k} - m_j \frac{\partial m_i}{\partial x_k}$$

is called a ‘Lifshitz invariant’ and denotes rotation about the $\hat{x}_k$ direction. Lifshitz variants exist in ferromagnets without inversion symmetry and describe an anti-symmetric form of spin coupling. Such interactions also exist in frustrated magnets and can be introduced by nonmagnetic impurities with strong spin-orbit coupling. The values of the coefficients C_1 , C_2 and C_3 are related to the symmetry of a material (table 1.1). For C_{nv} symmetry, $C_2 = C_3 = 0$ and the DMI induces a Néel rotation of spins, while for D_n symmetry, $C_1 = C_3 = 0$ and the DMI favours Bloch wall-like rotation.

1.2 Non-collinear Spin Textures: Helices, Cones and Skyrmions

Non-collinear magnetic structures in non-centrosymmetric magnets such as MnSi [24–26] and FeGe [27] have been known for a long time. In this case, the absence of spatial inversion symmetry and the spin-orbit coupling result in a finite DMI. Typically, in this symmetry class of magnets, the zero field ground state displays spin helices which evolve into cones under an applied field (figure 1.3a and b). The symmetry axis of cones is aligned along the applied field. At large fields, all spins align to form a field polarised magnetization state. After theoretical predictions of magnetic skyrmions around 20 years ago [23, 28, 29], a skyrmion lattice was imaged in the non-centrosymmetric magnet FeCoSi [30] at low temperatures in 2010. This was followed by the observation of skyrmions in various other non-centrosymmetric magnets: FeGe [13, 31–33], MnGe [34, 35], Cu₂OSeO₃ [36, 37], Co-Zn-Mn alloy [38, 39] etc..

Ferromagnetic films in proximity with heavy metals are another class of materials hosting non-collinear magnetic order. The absence of spatial inversion symmetry and strong spin-orbit coupling from the heavy metal generate a finite DMI at the ferromagnet-heavy metal (FM-HM) interface. A DMI in such heterostructures has been demonstrated by spin-polarised surface tunnelling microscopy (SP-STM) [40, 41], asymmetric domain wall motion [42] and spin-wave dispersion [43] experiments. In the absence of DMI, the magnetostatic interactions favour Bloch walls which

lack surface magnetic charges and hence have no stray magnetic field. A large DMI stabilises chiral Néel domain walls, where the spins rotate in a cycloidal manner. The chirality (i.e. the sense of rotation of the spins) of Néel walls is governed by the orientation of the DMI and can exist in clockwise or counterclockwise forms (figure 1.3(c) and (d)). Néel walls have been explored for racetrack applications due to their efficient coupling with electric currents [44, 45] via spin-orbit torque. More recently, DMI-induced magnetic skyrmions have been reported in FM-HM films [10, 11, 46, 47].

FIGURE 1.3: Non-collinear spin structures. (a) Spin helix: the arrow represents the direction of the propagation vector k of the helix. (b) Transformation from helical to conical structure under external magnetic field $\mathbf{H}$. (c) and (d) are Néel domain walls of opposite chirality.

The skyrmion ground state can be calculated by minimising the total energy of system in a model comprising exchange, DMI, uniaxial anisotropy, Zeeman and demagnetization terms. The total energy density is written as [48]

$$E = E_{exc} + E_{DMI} + E_{ani} + E_{Zeeman} + E_{demag} \quad (1.11)$$

The explicit form of the individual contributions to the energy is determined by the geometry and the details of the material structure.

$$E = A(\nabla \mathbf{m})^2 + w_D(\mathbf{m}) - K(\mathbf{m} \cdot \hat{\mathbf{n}})^2 - \mu_0 M H^e \mathbf{m} \cdot \hat{\mathbf{n}} - \mu_0 M \mathbf{m} \cdot \mathbf{H}^d \quad (1.12)$$

Here $\mathbf{M}$ is the magnetization and $\hat{\mathbf{n}}$ is the unit vector perpendicular to the film surface. The unit vector $\mathbf{m}$ is the reduced magnetization and is defined (in cylindrical coordinates) as

$$\mathbf{m} = \mathbf{M}/M = \sin \Theta \cos \Phi \hat{r} + \sin \Theta \sin \Phi \hat{\phi} + \cos \Theta \hat{z} \quad (1.13)$$

w_D is the energy density of the DMI: its precise form depends on the crystal symmetry (see table 1.1). A and K are the Heisenberg exchange parameter and

uniaxial anisotropy constant, respectively. H^e and $\mathbf{H}^d$ are the external (out-of-plane direction) and demagnetization fields. The magnitude and direction of $\mathbf{H}^d$ vary as a function of the local magnetization and can vanish for certain non-collinear magnetic structures, such as Bloch DWs. The above form is well suited to represent the axisymmetric structure of skyrmions and includes a translational symmetry along the $\hat{z}$ direction. Both Θ and Φ are functions of the spatial coordinates r and ϕ . The magnetization direction at the centre and perimeter of a skyrmion are antiparallel, resulting in boundary conditions $\Theta = \pi$ for $r = 0$ and $\Theta = 0$ for $r = \infty$.

For symmetry breaking in the $\hat{z}$ direction (which is the case for the FM-HM interface), the DMI expression is given by [49]

$$w_D = L_{zx} + L_{zy} = M_z(\nabla \cdot \mathbf{M}) - (\mathbf{M} \cdot \nabla)M_z \quad (1.14)$$

$$= \hat{y} \cdot \left(\mathbf{m} \times \frac{\partial \mathbf{m}}{\partial x} \right) - \hat{x} \cdot \left(\mathbf{m} \times \frac{\partial \mathbf{m}}{\partial y} \right) \quad (1.15)$$

Upon simplifying the various terms of the energy density equation, the exchange potential reduces to

$$(\nabla \mathbf{m})^2 = \mathbf{m} \cdot \nabla^2 \mathbf{m} \quad (1.16)$$

In polar coordinates

$$\mathbf{m} \cdot \nabla^2 \mathbf{m} = -\Theta_r^2 - \frac{\sin^2 \Theta}{r^2}, \quad (1.17)$$

$$\mathbf{m} \cdot (\nabla \times \mathbf{m}) = \sin \Phi \left(\Theta_r + \frac{\sin \Theta \cos \Theta}{r} \right) \quad (1.18)$$

The magnetic anisotropy along $\hat{z}$ can be written as

$$K(1 - m_z^2) = K \cos^2 \Theta \quad (1.19)$$

and the Zeeman term is

$$\mu_0 M H^e \mathbf{m} \cdot \hat{\mathbf{n}} = \mu_0 M H^e \cos \Theta \quad (1.20)$$

The net energy density in polar coordinates is [48]

$$E = A \left[\Theta_r^2 + \frac{1}{r^2} \Theta_\phi^2 + \sin^2 \Theta \left(\Phi_r^2 + \frac{1}{r^2} \Phi_\phi^2 \right) \right] + w_D - K \cos^2 \Theta - \mu_0 M H^e \cos \Theta - \mu_0 M \mathbf{m} \cdot \frac{\mathbf{H}^d}{2} \quad (1.21)$$

The above equation includes the rotationally symmetric solutions

$$\Theta = \Theta(r), \Phi = \Phi(\varphi), \mathbf{H}^d = \mathbf{H}^d(r) \quad (1.22)$$

The in-plane component of the magnetization Φ does not change with r . The general solution for Φ is given by

$$\Phi(\varphi) = n\varphi + \beta \quad (1.23)$$

β is defined as the helicity of the phase and indicates the sense of rotation for the in-plane magnetization. The values of n and β depend on the form of the DMI, whose characteristics for various crystal structures are provided in table 1.1. For example, $\Phi = \varphi$ (for Néel skyrmions) and $\Phi = \frac{\pi}{2} + \varphi$ (for Bloch skyrmion). Integrating the Φ component of energy density, we are left with the functional containing only the out-of-plane component of magnetization Θ

$$F = 2\pi \int_0^\infty f(\Theta, r) r dr \quad (1.24)$$

Here, $f(\Theta, r) = E(\Theta, r) - E(0)$ is the difference in energy density between the skyrmion and saturated phases. The energy density of the saturated state is given by $E(0) = -K - \mu_0 m_z H$ [48, 50]. $f(\Theta, r)$ can be expanded as

$$f(\Theta, r) = A \left(\Theta_r^2 + \frac{1}{r^2} \sin^2 \Theta \right) - D \left(\Theta_r + \frac{1}{r} \sin \Theta \cos \Theta \right) - K \sin^2 \Theta + \mu_0 H (1 - \cos \Theta) \quad (1.25)$$

In the above equation $H = H^e - H^{d(sk)}$, i.e the applied field minus the demagnetization field generated from an isolated axisymmetric skyrmion. The contribution of $H^{d(sk)}$ to the energy functional and the resulting magnetization structure varies with the ratio of magnetic layer thickness d to the skyrmion radius r_s . The skyrmion radius r_s is defined as the radial distance from the skyrmion centre to the point at which the out-of-plane magnetization is zero. For skyrmion sizes less than the film thickness, i.e $r_s \leq d$, the demagnetization field can be approximated by $\mu_0 M$. For films with larger skyrmions, i.e. $r_s \geq d$, the demagnetization contribution is negligibly small. Since typically observed skyrmion sizes are much larger than the film thickness, we will ignore the effect of the demagnetization field.

The Euler equation corresponding to the above functional is:

$$f(\Theta, r) = A \left(\Theta_{rr} + \frac{\Theta_r}{r} - \frac{1}{r^2} \sin \Theta \cos \Theta \right) + \frac{D}{r} \sin^2 \Theta - K \sin \Theta \cos \Theta - \mu_0 M H \sin \Theta = 0 \quad (1.26)$$

where subscripts r and rr represent the first and second order derivatives with respect to r . The above equation is valid for all rotationally symmetric solutions where the magnetization has an anti-parallel orientation at the centre and perimeter. Stable solutions exist only when Θ decreases with r . The boundary conditions for solution are therefore $\Theta(0) = \pi, \Theta(\infty) = 0$.

A convenient method for solving the Euler equation is to employ dimensionless units:

$$\rho = \frac{2\pi r}{L_D}, \quad h = \frac{H}{H_S}, \quad k = \frac{K}{K_0} \quad (1.27)$$

where $L_D = \frac{4\pi A}{|D|}$ is the period of a helix at zero field and zero anisotropy, $\mu_0 H_S = \frac{D^2}{2AM}$ is the saturation field and $K_0 = \frac{D^2}{4A}$ is the critical anisotropy. With the above substitutions, equation 1.25 reduces to a simpler form

$$\Theta_{\rho\rho} + \frac{1}{\rho} \Theta_\rho - \frac{1}{\rho^2} \sin 2\Theta + \frac{2}{\rho} \sin^2 \Theta - \frac{k}{2} \sin 2\Theta - h \sin \Theta = 0 \quad (1.28)$$

where subscripts ρ and $\rho\rho$ represent the first and second order derivatives with respect to ρ . The above equation provides a generalised description of localised inhomogeneous magnetic order in two dimensional thin films. The quantities A , D and K (or their dimensionless forms) govern the stability and distribution of various states such as helices, isolated skyrmions or a skyrmion lattice as a function of field and temperature.

For Néel skyrmions

$$\mathbf{m} = \cos \Theta(r) \hat{e}_z + \sin \Theta(r) \hat{e}_r \quad (1.29)$$

Here, the spin variation from the core of the skyrmion to the perimeter resembles that of a Néel domain wall (figure 1.4a). Néel skyrmions are typically stabilised at ferromagnet-heavy metal (FM-HM) interfaces, where an in-plane DMI favours Néel wall type rotation of spins. A detailed description of DMI at FM-HM interfaces and the resulting Néel skyrmion formation is provided in chapter 3. For Bloch skyrmions, where the DMI form is $w_D = D\mathbf{M} \cdot (\nabla \times \mathbf{M})$ [30], the skyrmion

solutions are

$$\mathbf{m} = \cos \Theta(r) \hat{e}_z + \sin \Theta(r) \hat{e}_\varphi \quad (1.30)$$

where the spin variation from skyrmion centre to perimeter resembles a Bloch wall (figure 1.4b). Different forms of spin rotation lead to different requirements regarding the DMI for Bloch and Néel skyrmions. While the Bloch skyrmions can be stabilised in zero DMI systems [51] via dipolar interactions [52], Néel skyrmions require a threshold value of the DMI. This can be explained by comparing the spin configuration of both skyrmions. The Néel skyrmions have uncompensated surface and volume charges, which add an ‘extra’ magnetostatic term in the Hamiltonian. A larger value of D is therefore required to compensate this term and support stable skyrmions [53].

FIGURE 1.4: Spin rotation in (a) a Néel skyrmion and (b) a Bloch skyrmion obtained using micromagnetic simulations. The arrows illustrate the direction of in-plane (IP) magnetization and the colour scheme follows the variation in out-of-plane (OP) magnetization. Simulations are performed on a film of 50 nm x 50 nm. Magnetic parameters used in simulations are: Exchange constant $A = 11.2$ pJ/m, $D = 2$ mJ/m², $K = 0.05$ MJ/m³ and $H = 300$ mT.

The magnetization profiles of skyrmions can be estimated by a 2π domain wall model [54, 55]. Although this model is not an exact solution of the energy functional, the predicted shape and size are in agreement with experimental observations [54, 55]. The skyrmion profile is controlled by three parameters: the centre of the skyrmion r_c , the radius of the skyrmion ρ , and the width of skyrmion wall Δ . The profile of a 2π domain wall is given by [40, 56]

$$\Theta(\rho, r_c, \Delta) = \begin{cases} \sum_+^- \left[\arcsin \left(\tanh \frac{-\rho \pm r_c}{\Delta/2} \right) \right] + \pi & B_z > 0 \\ \sum_+^- \left[\arcsin \left(\tanh \frac{-\rho \pm r_c}{\Delta/2} \right) \right] & B_z < 0 \end{cases} \quad (1.31)$$

Θ defines the polar angle of the magnetization relative to Θ at $\rho = 0$. The skyrmion radius is defined by the distance of the point at which $m_z = 0$, from the centre of skyrmion. The 2π domain wall approach was further updated in [57] to include the effect of stray fields, which become dominant in thin films with large magnetization.

1.3 Static and Dynamic Properties of Skyrmions

1.3.1 Stability of the Skyrmion Phase: Effect of Field-Temperature Perturbations

The enhanced stability arising from non-zero topological charge is one of the prime attractions of skyrmions. Even still, a skyrmion lattice can be deformed to a helical (ferromagnetic) state at low (high) magnetic fields. Understanding field-induced deformations of skyrmions (leading to their destruction) is of profound interest for both fundamental and technological reasons. Fundamentally, skyrmions provide an excellent platform to understand the evolution of topological states under continuous transformations. Simultaneously, a quantification of skyrmion stability is essential to realise the various proposed data storage solutions.

Increasing the magnetic field changes the magnetization from long helimagnetic stripes at low fields to skyrmions at intermediate fields. At higher fields, the magnetization enters a field polarised state. It has been reported that the decay of the skyrmion lattice into helimagnetic and ferromagnetic phases in bulk crystals is mediated by ‘chiral bobbbers’ [58, 59]. Chiral bobbbers are parabolic spin textures originating from a ‘Bloch point’ buried in the bulk. Two neighbouring skyrmions merge into helical stripes via a Bloch point, which acts as a zipper and stitches skyrmions together to form a stripe. During the skyrmion to ferromagnet transition at high fields, skyrmions evolve into metastable bobbbers, eventually collapsing into the ferromagnetic background. The stability of skyrmions at fields/temperatures beyond their stable phase can be quantified from their decay rates. Experimentally, decay rates can be determined by counting the number of skyrmions (N) over time during the decay process, as demonstrated in [60]. For both transformations (skyrmion to helimagnet at low fields and skyrmion to ferromagnet at high fields), N decays exponentially with time (figure 1.5), provided temperature and field remain constant. The decay in the number of skyrmions can be approximated by

$N = N_0 \exp\left(\frac{-t}{\tau(H,T)}\right)$, where N_0 is the number of skyrmions before application of a perturbation. The decay parameter $\tau(H, T)$ varies with temperature and magnetic field. At constant field, τ follows an Arrhenius dependence on temperature: $\tau(H, T) \approx \tau_0()e^{\Delta E(H)/k_B T}$. The energy barrier ΔE decreases at high fields, with the largest value being at fields close to the stable skyrmion phase. The prefactor τ_0 in the Arrhenius equation is highly sensitive to magnetic field and can vary by a factor up to 30, even for small changes in field [60]. Within the skyrmion

FIGURE 1.5: The exponential decay in the number of skyrmions in $\text{Fe}_{0.5}\text{Co}_{0.5}\text{Si}$ at high fields (a) and low fields (b) leading to formation of ferromagnetic and helimagnetic phases respectively. MFM images are 500 nm x 500 nm and time is measured in seconds. The solid green and blue lines in (a) and (b) indicate the exponential decay model. The images are reproduced from [60].

phase, the effect of temperature variations is two-fold. Firstly, a decrease/increase in temperature causes an inverse effect on magnetic anisotropy, which governs the stability and size of skyrmions. Secondly, thermal fluctuations play a critical role in skyrmion nucleation mechanisms. It has been demonstrated that at low temperatures, skyrmions nucleate in isolated locations, while at higher temperatures a skyrmion lattice is formed [61].

1.3.2 Topological Hall Effect in Skyrmions

Electronic Hall Effect

A unique manifestation of the topological character of skyrmions appears as an additional ‘topological’ contribution to the Hall resistance. In trivial ferromagnets, the Hall resistance comprises two distinct contributions. Firstly, the conventional

Hall resistance which increases proportionally with magnetic field B . This contribution dominates the high field region. Secondly, an anomalous Hall effect (AHE) emerges, which is proportional to the magnetization (M) and hence saturates at high fields.

During the adiabatic motion of a conduction electron over a spatially varying magnetic texture, the magnetic moment of the electron aligns instantaneously with the local magnetization. In a simplistic picture, the physical effect of adiabatic electron flow through a skyrmion can be understood as the electron experiencing a pseudo-magnetic emergent field:

$$\mathbf{H}_i^e = \frac{\hbar}{2} \epsilon_{ijk} \mathbf{m} \cdot (\partial_j \mathbf{m} \times \partial_k \mathbf{m}) \quad (1.32)$$

In the case of thin films, the magnetization variation is only in the xy plane, therefore, the emergent magnetic field equation simplifies to

$$\mathbf{H}_z^e = \frac{\hbar}{2} \mathbf{m} \cdot (\partial_x \mathbf{m} \times \partial_y \mathbf{m}) \quad (1.33)$$

$\mathbf{H}_z^e$ is the pseudo-magnetic field along $\hat{z}$ direction, originating from the spatial variation of the magnetization within a skyrmion. The ‘emergent magnetic flux’ corresponding to an isolated skyrmion is

$$\int_{sk} \mathbf{H}^e \cdot d\sigma = 4\pi\hbar S \quad (1.34)$$

where S is the topological charge of skyrmion and the integration is performed over the area of the skyrmion. The emergent magnetic field exerts an effective force on the moving electrons similar to the Lorentz force, resulting in a so-called ‘topological’ contribution to the Hall resistance.

Magnon Hall Effect

In addition to the topological Hall effect, skyrmions also exhibit a magnon Hall effect [62]. Magnons are quantised spin-waves, often created by a temperature gradient: they flow longitudinally in the direction of such gradients. The emergent magnetic flux of skyrmions causes a deviation from straight line motion of magnons. This deflection in magnon path is referred to as the ‘magnon Hall effect’. The magnon Hall effect has been observed in Lorentz transmission electron microscopy (LTEM) experiments on skyrmion lattices, where an electron beam induces a radial temperature gradient from the beam centre to edge, exciting a radial magnon

current. A tangential Lorentz force due to the emergent field of the skyrmions causes angular deviation of the magnons. A reaction to the Lorentz force rotates the skyrmions, which can be observed in LTEM [62].

1.4 Technological Relevance of Magnetic Skyrmions

Nanometer sizes, efficient manipulation by electrical and thermal currents, and stability against material defects promote skyrmions as excellent candidates for myriad spintronic applications. In this section, we will explore some of the technological possibilities for skyrmions.

Logic Operations and Neuromorphic Applications

FIGURE 1.6: Logic operations and neuromorphic computing by skyrmions. (a) The current-induced motion of a skyrmion resulting in AND operation. The three columns represent the various input conditions of AND gates and the horizontal rows represent the time evolution of skyrmion position. (b) OR gate operation by current-induced motion of skyrmions. Subpanels represent various input conditions for OR gates and rows represent the time evolution of skyrmion position [63]. (c) Proposed model of a skyrmion-based neuromorphic device, where the PMA along the racetrack increases linearly with the relative thickness of the ferromagnet and heavy metal layer. Such configuration provides a control of skyrmion motion dynamics, and can enable neuromorphic functionality [64].

As demonstrated in ref. [63], isolated skyrmions can be utilised to perform logic operations. Figure 1.6 illustrates this principle, where the skyrmion state is considered as 1 and FM background is 0. The OR operation, i.e. high output if any of the inputs is high, is equivalent to merging the skyrmion with either the

background or itself, to form another skyrmion. Motion of skyrmions along the tracks is driven by current pulses. The distinction between OR and AND gates is apparent for 1 and 0 inputs. The output of this input combination is 1 for OR and 0 for AND. In the proposed devices, this distinction is preserved by the width of an intermediate channel. In case of OR gates, a narrow channel supports transmission of a single skyrmion. A wider channel, employed in the AND gate, annihilates a single skyrmion. Two skyrmions merging at the input edge of the channel can form a wide domain wall, which converts into a skyrmion at the output edge. Moreover, the functionalities of skyrmions have been extended to neuromorphic computations [64, 65].

Data Storage and Race Track Applications

Traditional magnetic storage devices (such as magnetic hard disks) utilise the magnetization orientation of grains to encode binary data. The encoding and decoding mechanism involves a mechanical rotary head and a giant magnetoresistance (GMR) sensor. The thermal stability of the magnetic grains is determined by $K_u V / k_B T$, where K_u is the magnetic anisotropy, V is the volume of the magnetic grain, k_B is the Boltzmann constant and T is the temperature. Consequently, a large grain volume is an essential prerequisite for reliable data storage. However, this results in low storage density compared to other forms of storage media such as solid-state drives (SSDs). Another limitation emerges from the use of a mechanical head during reading and writing operations, which increases the vulnerability of devices to vibrations and shocks. Also, since bits are accessed via a mechanical rotation of the head, data accessibility is slow and requires high power consumption. These challenges demand alternative forms of data storage devices, for which skyrmions are potential candidates.

Magnetic skyrmions can potentially overcome both limitations of HDDs. Skyrmions with diameter below 50 nm [46, 47, 55] can be stabilised in thin films at room temperatures. Their small size offers a significant boost to data storage capacity compared to larger magnetic grains. Furthermore, skyrmions can be manipulated and detected by ultra-low electronic and/or magnonic currents, hence eliminating the requirement for mechanical parts, and hence rendering the devices immune to shocks and vibrations. An additional advantage is the low power requirement for skyrmion manipulation which allows the use of skyrmion based data storage devices

in a wide variety of electronics. Moreover, current-based skyrmion manipulation offers a platform to integrate processing and data storage operations into a single chip, reducing the overall footprint of the device.

Recent experimental demonstrations [10, 46, 47, 55] of zero-field, room temperature Néel skyrmions have established the nucleation, annihilation, and transportation of skyrmions by an electric current (figure 1.7). An immediate application is a racetrack-like device based on skyrmions. In the proposed device, low (zero) and high (one) states can be distinguished by the absence and presence of skyrmions. The topological Hall resistance [66, 67] or non-collinear magnetoresistance (NCMR) [68] of skyrmions can subsequently be employed to probe the presence/absence of skyrmions. A stringent requirement for a racetrack device is the straight-line motion of skyrmions along the track. However, as has been demonstrated [69, 70], skyrmions display angular deviations from the straight line paths. Such motion results in undesirable side-effects, such as annihilation or reflection of skyrmions from the edges of race track. To resolve the issue, new methods such as antiferromagnetically coupling skyrmion pairs have been proposed.

FIGURE 1.7: (a) Various schemes for motion of skyrmions in bulk and interfacial DMI materials using spin transfer torque (STT) and the spin Hall effect (SHE) [71]. (b) AFM coupled ferromagnetic layers hosting skyrmions [72]. AFM coupling is proposed to eliminate the skyrmion Hall effect (c) Current driven motion of skyrmions in a Pt/CoFeB/MgO stack [47] (d) Current driven motion and the skyrmion Hall effect in Ta/CoFeB/TaO heterostructures [46].

Skyrmions and Magnonics

An interesting field developing in parallel with skyrmions is ‘magnonics’, which deals with manipulation of magnetization by spin-waves and spin currents. This is achieved by transfer of angular momentum and does not involve electron motion, thus avoiding Joule heating. Proposed applications include magnonic transistors for spin-wave logic, diodes for unidirectional flow of spin-waves and magnonic crystals for spin-wave modulation.

Skyrmions (both in lattice and isolated configurations) can be exploited for novel applications in magnonics. For example, lithographically-created periodic arrays of ferromagnetic dots have been explored for spin-wave modulation [73]. These arrays could be replaced by a skyrmion lattice (figure 1.8)(as observed in the insulator Cu_2SeO_3 [36, 74]) providing several advantages over lithographically fabricated geometry. Firstly, skyrmion size can be reduced to a few nanometers, which is challenging for lithography techniques. Secondly, the periodicity and order of the array would be tunable by magnetic field, thus eliminating the requirement for multiple devices.

Another possible application is a skyrmion-based diode for microwaves. It has been demonstrated that a skyrmion lattice favours asymmetric transmission of microwaves: a functionality which should be developed into a diode. Several numerical and experimental studies have also suggested using skyrmion lattices as nano-oscillators [75] and microwave detectors [76].

FIGURE 1.8: Proposed skyrmion-based spin torque oscillator [75]. The magnetic structure consists of a fixed layer, nonmagnetic spacer, free layer and electrodes at the top and bottom. Current-induced GHz oscillations of skyrmions are used as a microwave generator. (b) Skyrmion based magnonic crystals where band gaps can be introduced by the presence/absence of skyrmions [77].

1.5 Organisation of the Thesis

This thesis explores the resonant dynamics of Néel skyrmions in sputtered Ir/Fe/-Co/Pt multilayers. We observe high and low frequency dispersion modes with distinct magnetic field dependencies corresponding to Néel skyrmions using field and frequency sweep broadband ferromagnetic resonance (FMR) experiments performed in home-built apparatus. Our micromagnetic simulations and analytical calculations identify the origin of low and high frequency resonance modes as counterclockwise gyrations of skyrmion magnetization and scattering of “background” magnons from ferromagnetic regions off skyrmions, respectively. Furthermore, we also investigate the correlation between material architecture and resonance frequencies, which can be utilised to tune the microwave frequency response of skyrmions for specific device applications, such as skyrmion-based nano-oscillators. The thesis is organised as follows:

Chapter 2 explains the fundamentals of resonance in magnetic materials, experimental techniques, and data analysis procedures. In addition, it includes the instrumentation details for the home-built broadband FMR set-up used in this work.

Chapter 3 details the systematic approach of engineering thin film interfaces to optimise the magnetic interactions responsible for non-collinear magnetization. It also includes details of our magnetic characterisation of the skyrmion phase via magnetometry and magnetic force microscopy.

Chapter 4 reports the experimental observation of skyrmion resonance by field and frequency sweep FMR techniques. The experimental results are supplemented by micromagnetic simulations and analytical calculations.

Chapter 5 provides a summary of the work and includes a brief description of ongoing and proposed new experiments on the magnetization dynamics of skyrmions.

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Chapter 2

Broadband Ferromagnetic Resonance (FMR): Technique and Instrumentation

Ferromagnetic resonance (FMR) is a well established technique to characterise the magnetization dynamics and relaxation mechanisms in ferromagnets. Over the years, it has been employed to quantify the static (such as anisotropy and exchange coupling) as well as dynamic (such as damping coefficient and relaxation time) properties of ferromagnets. In the previous chapter, we have detailed the various energy contributions leading to the formation of noncollinear spin textures. This chapter extends the discussion to the magnetization dynamics of ferromagnets. We explain the various relaxation and damping mechanisms and their experimental characterisation by broadband ferromagnetic resonance (FMR) spectroscopy. The later sections of the chapter describe the design and instrumentation of a home-built FMR set-up.

2.1 Magnetization Dynamics in Ferromagnets

At the atomic scale, the magnetic moment of an electron originates from two independent sources. The first contribution is the orbital motion of the electron around the nucleus, characterised by an orbital angular momentum $\mathbf{L}$. This is analogous to the classical phenomenon of electric charge accelerating in a closed-loop circuit, resulting in a magnetic field. The corresponding ‘orbital’ magnetic

moment μ_l is defined as $\mu_l = -\frac{|e|\hbar}{2m}\mathbf{L}$, where e and m are the charge and mass of an isolated electron. The second contribution to the magnetic moment emerges from the intrinsic spin degree of freedom (and its related spin angular momentum $\mathbf{S}$). The ‘spin’ magnetic moment associated with an electron is $\mu_s = -\frac{|e|\hbar}{m}\mathbf{S}$. From this mathematical representation, it is evident that the magnetic momenta are collinear and directed opposite to the angular momenta. The relative amplitude of the two moments scales by an integral multiple of factor $\frac{|e|\hbar}{2m}$, referred to as the gyromagnetic ratio of a free electron. The two atomic momenta interact via the spin-orbit interaction, and the resultant momentum $\mathbf{J}$ is calculated as a vector sum of the orbital and spin magnetic moments: $\mathbf{J} = \mathbf{L} + \mathbf{S}$.

This spin-orbit interaction leads to a precession of the angular momenta $\mathbf{L}$ and $\mathbf{S}$ (and their associated magnetic moments) about $\mathbf{J}$. The net atomic moment is determined by $\mu_j = -\frac{|e|\hbar}{2m}\mathbf{L} - \frac{|e|\hbar}{m}\mathbf{S}$. However, unlike μ_l and μ_s , the total magnetic moment μ_j is not aligned with its corresponding angular momentum $\mathbf{J}$, as indicated in figure 2.1. The higher gyromagnetic ratio of the spin moment tilts μ_j towards μ_s , eliminating the collinearity of μ_j and $\mathbf{J}$. The fast precession of μ_j around $\mathbf{J}$ leads to the cancellation of the perpendicular component of μ_j . The remaining component of μ_j (indicated by μ from here onwards) is related to $\mathbf{J}$ by the relation, $\mu = \frac{g|e|\hbar}{2m}\mathbf{J}$, where $g = 1 + \frac{j(j+1) + s(s+1) - l(l+1)}{2j(j+1)}$ is the spectroscopic splitting factor. s , l and j are the quantum numbers related to the spin, orbital and total angular momentum of an atom.

FIGURE 2.1: Schematic representation of the orbital $\mathbf{L}$ (blue), spin $\mathbf{S}$ (green) and net angular momentum $\mathbf{J}$ (red) of an electron. μ_L , μ_S and μ_{avg} represent the corresponding magnetic momenta. μ_{avg} is tilted towards μ_S due to the large g -factor of $\mathbf{S}$. μ_J is the time average of μ_{avg} , and is collinear with $\mathbf{J}$.

The presence of an effective magnetic field $\mathbf{H}_{eff}$ results in precession of the magnetic moment around $\mathbf{H}_{eff}$. The equation of motion for precession can be written as

$$\frac{1}{\gamma} \frac{d\boldsymbol{\mu}}{dt} = -[\boldsymbol{\mu} \times \mathbf{H}_{eff}] \quad (2.1)$$

where $\gamma > 0$ is the gyromagnetic ratio $\frac{g|e|}{2m}$. $\mathbf{H}_{eff}$ is related to the total energy by $\mathbf{H}_{eff} = \partial_M(E_{tot})$: it is a vector sum of applied, anisotropy and demagnetization fields. The motion defined by equation 2.1 is known as Larmor precession and the precession frequency is calculated as $\omega_l = \gamma H_{eff}$. The torque vector $-\boldsymbol{\mu} \times \mathbf{H}_{eff}$ is directed perpendicular to $\boldsymbol{\mu}$, and hence $\boldsymbol{\mu}$ remains conserved during the precession.

In ferromagnetic materials, exchange interactions induce parallel alignment of neighbouring magnetic moments. The net volume magnetization is expressed as $\mathbf{M} = \sum_i \boldsymbol{\mu}_i$. Under a large effective magnetic field (greater than the saturation field of the material), all atomic moments are aligned along $\mathbf{M}$ (with low amplitude thermal fluctuations causing random orientation of atomic moments) and hence $\mathbf{M}$ can be approximated by an algebraic sum of parallel atomic moments. The algebraic sum approximation is not valid for materials with spatially varying anisotropy. In such cases, the orientation of the local magnetization will vary and the net magnetization will be a vector sum. In the algebraic sum approximation, the macroscopic magnetization dynamics arise from the collective response of all the atomic moments, expressed as

$$\frac{1}{\gamma} \frac{d\mathbf{M}}{dt} = -[\mathbf{M} \times \mathbf{H}_{eff}] \quad (2.2)$$

The motion represented by equation 2.2 is known as ‘uniform mode precession’. In case of low dimensional magnets, $\mathbf{H}_{eff}$ includes a large contribution from the shape anisotropy of the material. The role of shape anisotropy in the resonance spectra can be understood by a simple example of an ellipsoid with the major and minor axes along the x , y and z directions. The ellipsoid is positioned in a static magnetic field $\mathbf{H}_{ext} = \mathbf{H}_z$. $\mathbf{H}_{ext}$ is assumed to be equal to $\mathbf{H}_{eff}$, since large external fields will dominate the dynamics, applied along the $\hat{z}$ direction. A microwave field $\mathbf{h}$ of frequency ω_0 is applied in the plane of the material. The demagnetization factors are N_x , N_y and N_z along the three major axes. The magnetic field in each direction

will be

$$H_{xi} = h - N_x M_x \quad (2.3)$$

$$H_{yi} = -N_y M_y \quad (2.4)$$

$$H_{zi} = H_z - N_z M_z \quad (2.5)$$

where index i refers to the instantaneous magnitude of the field. Substituting the scalar values of $\mathbf{H}$ into equation 2.2 and assuming a sinusoidal time evolution of the magnetization, the equation of motion can be written in a simplified scalar form:

$$\frac{dM_x}{dt} = \gamma[H_z + (N_y - N_z)M_z]M_y \quad (2.6)$$

$$\frac{dM_y}{dt} = \gamma[M_z h + (N_x - N_z)M_z M_z - M_x H_z] \quad (2.7)$$

$$\frac{dM_z}{dt} = 0 \quad (2.8)$$

The assumption of a sinusoidally evolving magnetization is valid only for low amplitudes of the microwave field $\mathbf{h}$, causing a small deflection in the magnetization from the equilibrium direction. Also, the sinusoidal evolution of the magnetization allows us to replace $\frac{d}{dt}$ by $i\omega$. Solving the above three equations for susceptibility $\chi_x = \frac{M_x}{h}$, we obtain

$$\chi_x = \frac{\chi_0}{1 - \frac{\omega^2}{\omega_0^2}} \quad (2.9)$$

where $\chi_0 = \frac{M_z}{H_z + (N_x - N_z)M_z}$ and the resonant angular frequency is

$$\omega^2 = \gamma \sqrt{[H_z + (N_y - N_z)M_z] \times [H_z + (N_x - N_z)M_z]} \quad (2.10)$$

The demagnetization factors are functions of the material geometry. In the case of a perpendicularly magnetized film, ($N_x = N_y = 0, N_z = 1$ (or, 4π in SI units)), the resonant angular frequency is

$$\omega = \gamma(H_z + M_z) \quad (2.11)$$

In typical experiments, frequency $f = \omega/2\pi = \frac{\gamma}{2\pi}(H_z + M_z)$ is the measured quantity. In practice, $M_{eff} = M_z - \frac{2K}{M_s}$ is used in calculations, since this includes the effects of the PMA and demagnetization field on M_z . The above model is valid only for coplanar magnetic moments and cannot be applied to resonance in canted spin textures. Experimentally, parallel moment orientation can be obtained only

above the saturation limit and hence resonance experiments are performed in the field range $\mathbf{H}_z > \mathbf{H}_{sat}$.

2.2 Magnetic Relaxation and Damping

Equation 2.2 predicts a dissipationless precession of $\mathbf{M}$ around $\mathbf{H}_{eff}$. However, this prediction contradicts experimental observations in which $\mathbf{M}$ relaxes to its equilibrium position, i.e. parallel to $\mathbf{H}_{eff}$, in a finite time. Several models have been developed to accommodate a magnetization relaxation term within the equation of motion for precession (2.2).

In one of the earliest models, a radially-directed damping torque is added to the precession equation. The radial direction provides the shortest path to the equilibrium position and hence is in agreement with experimental observations (figure 2.2). Mathematically, the modified equation of motion is written as [1]

$$\frac{d\mathbf{M}}{dt} = -\gamma \mathbf{M} \times \mathbf{H}_{eff} - \gamma_L \frac{\lambda_L}{M_s^2} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{eff}) \quad (2.12)$$

where γ_L is a gyromagnetic constant and $\lambda_L > 0$ is a material dependant phenomenological constant. The second term on the RHS accounts for the magnetization relaxation. A cross product in both RHS terms ensures the conservation of magnetization.

Landau-Lifshitz-Gilbert Equation

A more accepted formulation for magnetic relaxation was proposed by Gilbert and is referred to as Landau-Lifshitz-Gilbert (LLG) equation. It describes the time evolution of the transverse part of the magnetization [2]

$$\frac{d\mathbf{M}}{dt} = -\gamma \mathbf{M} \times \mathbf{H}_{eff} + \frac{\alpha_G}{M_s} \left(\mathbf{M} \times \frac{d\mathbf{M}}{dt} \right) \quad (2.13)$$

where α_G is a material dependent phenomenological damping constant which determines the decay rate of magnetization towards its equilibrium position. Rearranging the terms leads to

$$\frac{d\mathbf{M}}{dt} = -\gamma_G \mathbf{M} \times \left(\mathbf{H}_{eff} - \frac{\alpha_G}{\gamma_G M_s} \frac{d\mathbf{M}}{dt} \right) \quad (2.14)$$

FIGURE 2.2: Magnetic precession described by the Landau-Lifshitz equation. The blue arrow points in the direction of the tangential torque responsible for precession, while the green arrow indicates the direction of damping torque.

where γ_G is the modified gyromagnetic ratio. It is evident from equation 2.14 that the relaxation is modelled by subtracting a viscous drag field (proportional to the time derivative of the magnetization) from the net effective magnetic field. For the $\alpha_G = 0$ case, the magnetization will revolve around the $\mathbf{H}_{eff}$ axis indefinitely. For finite damping ($\alpha_G \neq 0$), the magnetization decays along a spiral path on a timescale of $(\alpha_G \omega)^{-1}$ towards the effective field [3], where ω is the precession frequency.

We will now solve the LLG equation for an IP magnetized thin film and derive an expression for the dynamic susceptibility using the LLG formalism. For thin films with IP anisotropy $\mathbf{H}_{uni} = \frac{2K_u}{M_s} \hat{x}$, an external field $\mathbf{H}_{ext}$ along the x direction, and a small microwave perturbation $\mathbf{h}$ in the y direction, the effective magnetic field is

$$\mathbf{H}_{eff} = (H_{ext} + H_{uni})\hat{x} + h\hat{y} - \frac{M_{eff}}{M_s}m_z\hat{z} \quad (2.15)$$

The last term combines the demagnetization field and PMA along the z direction through $M_{eff} = M_s - \frac{2K_\perp}{dM_s}$, where d is the film thickness. The net magnetization $\mathbf{M}$ of the film in an applied field $\mathbf{H}_{ext}$ is

$$\mathbf{M} = M_s\hat{x} + m_y\hat{y} + m_z\hat{z} \quad (2.16)$$

Inserting $\mathbf{M}$ into the LLG equation and separating the three magnetization components, we obtain

$$0 = -\gamma(-m_y \frac{M_{eff}}{M_s} m_z - m_z h) + \frac{\alpha}{M_s} (m_y \frac{dm_z}{dt} - m_z \frac{dm_y}{dt}) \quad (2.17)$$

$$\frac{dm_y}{dt} = -\gamma(m_z(H_{Ext} + H_{uni} + M_{eff}m_z) - \alpha \frac{dm_z}{dt}) \quad (2.18)$$

$$\frac{dm_z}{dt} = -\gamma(M_s h - m_y(H_{Ext} + H_{uni})) + \alpha \frac{dm_y}{dt} \quad (2.19)$$

The above equations can be linearised by ignoring the terms containing products of hm_i . Also, the time dependence can be introduced via $m_i = m_i \exp(i\omega t)$ due to the sinusoidal excitation field $\mathbf{h}$. The resultant equations are

$$0 = \tilde{m}_y(i\omega) + \tilde{m}_z(\omega_H + \omega_{eff} + i\omega\alpha) \quad (2.20)$$

$$\omega_M \tilde{h} = \tilde{m}_y(i\omega\alpha + \omega_H) + \tilde{m}_z(-i\omega) \quad (2.21)$$

in which

$$\omega_H = \gamma(H_{Ext} + H_{Uni}) \quad (2.22)$$

$$\omega_M = \gamma M_s \quad (2.23)$$

$$\omega_{eff} = \gamma M_{eff} \quad (2.24)$$

Rewriting the above equations in matrix form yields

$$\begin{pmatrix} \tilde{h} \\ 0 \end{pmatrix} = \begin{bmatrix} \omega_H + i\omega\alpha & -i\omega \\ i\omega & \omega_H + \omega_{eff} + i\omega\alpha \end{bmatrix} \begin{pmatrix} \tilde{m}_y \\ \tilde{m}_z \end{pmatrix} \quad (2.25)$$

The susceptibility tensor can be extracted by inverting the above equation:

$$\begin{pmatrix} \tilde{m}_y \\ \tilde{m}_z \end{pmatrix} = \begin{bmatrix} \chi_{yy} & \chi_{yz} \\ \chi_{zy} & \chi_{zz} \end{bmatrix} \begin{pmatrix} \tilde{h} \\ 0 \end{pmatrix} \quad (2.26)$$

The above equation is of the form $\mathbf{m} = \chi \mathbf{h}$. Expanding the susceptibilities, we obtain

$$\begin{pmatrix} \tilde{m}_y \\ \tilde{m}_z \end{pmatrix} = \frac{\omega_M}{\omega_H(\omega_{eff} + \omega_H) - \omega^2 + i\omega\alpha(2\omega_H + \omega_{eff})} \begin{bmatrix} \omega_H + \omega_{eff} + i\omega\alpha & i\omega \\ -i\omega & \omega_H + i\omega\alpha \end{bmatrix} \begin{pmatrix} \tilde{h} \\ 0 \end{pmatrix} \quad (2.27)$$

As is evident from equations 2.26 and 2.27, the susceptibility term consists of real and imaginary parts. The real part of the susceptibility originates from magnetization precession in phase with the excitation field $\mathbf{h}$, while the imaginary parts relates to the magnetization component delayed by a phase of $\pi/2$ relative to $\mathbf{h}$. The imaginary susceptibility represents the absorption of electromagnetic energy and follows a Lorentzian line shape with a resonant maximum at frequency ω_{res} . The real component of the susceptibility represents the dispersion and vanishes at ω_{res} . Figure 2.8b and 2.8c display the real and imaginary components of the microwave absorption. An important consequence of equation 2.27 concerns the dependence of the resonance spectra on the damping constant α , which appears in the denominator of the RHS, suggesting a decrease in the absorption amplitude at resonance for high damping materials. This is consistent with experimental observations, where large damping materials such as metallic and/or multilayer films display weak absorption of microwaves.

The observed intensity of the FMR signal is proportional to the net magnetization of the film [4]:

$$I \propto M_s \frac{H_{Res} + M_{eff}}{2H_{Res} + M_{eff}} \quad (2.28)$$

Consequently, thick magnetic films exhibit a stronger FMR signal.

A large damping implies that long time scales are required to switch the magnetization, constituting a fundamental bottleneck towards the high-speed operation of logic devices and magnetic memories. A significant proportion of research in magnetism is dedicated to engineering new materials and novel mechanisms to reduce damping values. Magnetic heterostructures, similar to those explored in this thesis, allow new mechanisms such as spin-orbit torques to manipulate magnetization states.

The LLG equation (2.13) assumes a radially symmetric rotation of the magnetization around $\mathbf{H}_{eff}$. This is not always the case. A local in-plane anisotropy lifts the rotational symmetry of the precession by modifying its trajectory to an ellipse [5]. For an elliptical precession orbit, α is a tensor quantity.

The magnetization decay towards the effective field is accompanied by a transfer of energy from the uniform mode to the crystal lattice, typically referred to as magnetic relaxation. The uniform mode can relax via direct and/or indirect mechanisms (figure 2.3) [6]. For direct relaxation, energy is dissipated directly into the crystal lattice. A crystal lattice can couple to the magnetization via magnetoelastic or spin-orbit interactions, hence facilitating the relaxation of the uniform mode to the lattice sites. External contributions such as the presence of rare earth impurities with large orbital momenta also favour direct magnon to phonon relaxation, where magnons are quantised modes of magnetic precession. This effect is primarily dominant at low temperatures. The presence of such contributions is verified by a distinct peak in the linewidth-temperature graph for a given frequency [7].

FIGURE 2.3: Dissipation of magnetic energy of the uniform mode into the crystal lattice. The magnetization can relax to the crystal lattice ‘directly’ via spin-orbit interactions, or ‘indirectly’ via relaxing into lower energy spin wave modes first. These spin wave modes eventually dissipate the energy to the crystal lattice.

During indirect relaxation, the uniform precession mode (characterised by a zero spin-wave vector $k = 0$) scatters into $k \neq 0$ modes. This can occur by halving the frequency and splitting the uniform mode into two degenerate magnons with opposite wave vectors: $\omega_0 = \omega_{k+} + \omega_{k-} = 2\omega_k$. Similarly, another possibility is for two uniform mode quanta to scatter into two degenerate magnons: $2\omega_0 = \omega_{k+} + \omega_{k-} = 2\omega_k$. A uniform precession mode can dissipate energy via spin-wave

formation. Mode splitting is primarily caused by the surface non-uniformity (bumps and pits) or the impurities and defects present in bulk materials. This relaxation mechanism is broadly referred to as ‘two magnon scattering’ and is characterised by a non-linear proportionality of the FMR linewidth to the frequency of the microwave field [8, 9]. Furthermore, since the magnon formation is a thermal process, signatures of this contribution to the damping can be observed in the temperature dependence of the resonance linewidth. During indirect relaxation, energy remains in the spin configuration but is eventually dissipated to the lattice over longer time scales. Dissipation via magnon modes is only feasible in large samples where spatial variation of spin orientation over long distances does not cost additional exchange energy.

Bloch Equations

The LLG equation 2.13 is valid under the assumption of a conserved magnetization $\mathbf{M}$ spiralling towards $\mathbf{H}_{eff}$. This formalism does not permit the formation of nonuniform magnon modes $k \neq 0$ during the relaxation process. Therefore, indirect relaxation processes cannot be explained by the LLG model. In contrast, the Bloch equations allow variation in the transverse magnetization component, thus enabling the formation of $k \neq 0$ magnon modes, which facilitate the indirect relaxation of the uniform precession mode. In this approach, the magnetization components parallel and perpendicular to the effective field relax via two independent mechanisms and hence exhibit distinct relaxation rates. The Bloch equations can be written as [10]:

$$\frac{dm_{x,y}}{dt} = -\gamma(\mathbf{M} \times \mathbf{B})_{x,y} - \frac{m_{x,y}}{\tau_2} \quad (2.29)$$

where the damping torque acts along the radial direction, and

$$\frac{dM_z}{dt} = -\gamma(\mathbf{M} \times \mathbf{B})_z - \frac{M_z - M_0}{\tau_1} \quad (2.30)$$

where the damping torque is directed in the $\hat{z}$ direction. M_z and M_0 are the instantaneous and equilibrium magnetization in the $\hat{z}$ direction, and $B = B_0$ is the external magnetic field applied in the $\hat{z}$ direction. The first equation describes the time evolution of the IP magnetization component (either $\hat{x}$ or $\hat{y}$) and the second describes the evolution of the OP magnetization. τ_1 and τ_2 are time constants characterising the dynamics of the magnetization components. τ_1 measures the time in which the magnetization will return to its equilibrium position after a

perturbation. This is referred to as the ‘longitudinal relaxation time’ or ‘spin-lattice relaxation time’ and represents the direct transfer of energy from spins into the crystal lattice. The energy lost during magnetization reorientation ($M_z \cdot B$) is transferred to the lattice. τ_1 is typically of the order of 10^{-6} s. Equation 2.29 describes the relaxation in the transverse magnetization component, which gradually decays to zero in time τ_2 . This is known as the transverse relaxation time or ‘spin-spin relaxation time’ which measures the decay of the uniform mode into other magnon modes. The typical duration of τ_2 is $\approx 10^{-10}$ s: its value is independent of temperature.

A more general classification separates the damping mechanisms into intrinsic and extrinsic contributions. In a broad sense, intrinsic contributions originate from the physical state of the material and include interactions between electrons, phonons and magnons. One such example is the scattering of spin-waves by electrons and phonons, as observed in metals. The extrinsic contributions arise from the presence of material defects and impurities. Surface pits on spherical samples of Yttrium Iron Garnet (YIG) acting as scattering sites for magnons [11] are an example of an extrinsic damping contributor.

The damping term appearing in the LLG equation originates from the spin-orbit interactions present in the material and can hence be classified as an intrinsic relaxation. The coupling between spin and angular momentum connects the precession of the uniform mode with the lattice, causing dampening of the motion towards the effective magnetic field [12]. This form of damping is characterised by a linear dependence of the linewidth on frequency, and is a general hallmark of the intrinsic relaxation processes [13]. Experimentally, the damping constant is obtained by a linear fitting of the linewidth versus frequency using equation 2.31, and contains all the intrinsic contributions including the ‘Gilbert damping’. We will hence refer to it as the ‘effective Gilbert damping’.

$$\Delta H = \Delta H_o + \frac{4\pi\alpha_{eff}}{\gamma}f \quad (2.31)$$

The first term is the zero frequency linewidth, denoting the frequency-independent extrinsic contribution to the linewidth. The most common sources of extrinsic damping are inhomogeneous magnetic parameters and impurities or defects in the film, which are symptomatic of poor film quality.

2.2.1 Damping in Metals

The intrinsic damping in ferromagnetic metals originate from a combination of spin-orbit interactions and electron-lattice scattering. Low temperature damping measurements have revealed two contributions to the damping, which exhibit opposite trends with temperature. While one increases proportionally with temperature, the other shows an inverse temperature dependence. A combined effect of these two contributions results in a diverse range of minimum damping temperatures (at which the damping reaches its lowest value) for various metals (for example, 100 K in Co and 310 K in Ni). It was identified that the temperature dependences of the above contributions are similar to those of electrical conductivity (which decreases with temperature) and resistivity (which increases with temperature), hence sometimes these are referred to as ‘conductivity like’ and ‘resistivity like’ dampings [14].

A qualitative understanding of the damping contributions can be developed by using Kambersky’s torque-correlation model. This model describes the damping of the uniform mode due to variations $\partial\epsilon_{n,k}/\partial\theta$ in the energies ϵ_{nk} of single particle states with respect to the spin direction θ . Due to spin-orbit coupling, the energy of the electronic states changes during magnetic precession. Consequently, some unoccupied energy levels are pushed below the Fermi level and simultaneously, some occupied states are pushed above the Fermi level. This results in formation of electron-hole pairs. The newly created electrons and holes exist for a finite lifetime, and ultimately relax via lattice scattering. Therefore, this model describes a relaxation of energy into the crystal lattice, mediated by spin-orbit coupling and charge carriers. The damping rate (d) in Kambersky’s model [15] is

$$d = \pi\hbar \frac{\gamma^2}{\mu_0} \sum_{nm} \int |\Gamma_{mn}^-(k)|^2 \frac{d^3k}{(2\pi)^3} \times \int d\epsilon_1 A_{nk}(\epsilon_1) A_{mk}(\epsilon_1) \eta(\epsilon_1) \quad (2.32)$$

n and m are band indices and k is the electron wavevector. The matrix element Γ_{mn}^- describes transitions between the n and m states induced by spin-orbit torques. The second integral accounts for the scattering of charge carriers by the crystal lattice. $\eta(\epsilon)$ is the derivative of the Fermi function which peaks around the Fermi level. A_{mk} and A_{nk} are Lorentzians in energy space centred at band energies ϵ_{nk} , whose width is determined by the scattering rate of charge carriers. For $m = n$, i.e., for scattering within the same band, the relaxation is referred to as ‘intra-band’, otherwise the process is referred to as ‘inter-band’ relaxation. The spectral integral

$\int d\epsilon_1 A_{nk}(\epsilon_1) A_{mk}(\epsilon_1) \eta(\epsilon_1)$ for intra-band transitions is proportional to the scattering time (similar to the conductivity), leading to an inverse dependence on temperature. For the inter-band transitions, the spectral integral decreases inversely with the scattering time (similar to resistivity) and a proportional increase with temperature is observed.

As described in the previous section, the physical origin of the inter and intra-band damping terms lies in the deviations of the equilibrium charge carrier population around the Fermi surface, caused by the magnetization precession. The total electronic energy of a system is given by $E = \sum_{nk} \rho_{nk} \epsilon_{nk}$, where ϵ_{nk} is the energy of a single electron and ρ_{nk} is the state occupancy. The effect of variation in electronic energy can be treated as an effective magnetic field, given by the partial derivative of the energy with respect to magnetization [3, 15, 16]:

$$H_{eff} = -\frac{1}{\mu_0 M} \sum_{nk} \left[\rho_{nk} \frac{\partial \epsilon_{nk}}{\partial M} + \frac{\partial \rho_{nk}}{\partial M} \epsilon_{nk} \right] \quad (2.33)$$

The first term describes the variation in the spin-orbit energy of the state caused by magnetization precession, which causes the Fermi surface to ‘breathe’. Physically, this refers to an oscillation in the Fermi surface size which opens an intra-band damping channel in ferromagnetic metals.

The second term indicates the creation of electron and hole pairs. The change in the spin-orbit interaction strength, caused by the magnetic precession, leads to creation of electron-hole pairs near the Fermi surface. Creation of charge carriers in this manner is similar to bubble formation in a liquid, and hence this process is described as Fermi surface ‘bubbling’. The electron hole pairs generated from spin precession relax via lattice scattering, causing the damping to rise.

Another approach to explain damping in ferromagnetic metals is the $s-d$ exchange model [17, 18], where the localised $3d$ orbital spins $\mathbf{S}$ interact with $4s$ electrons ($\mathbf{s}$) via the exchange interaction, $E = -2J_{sd} \mathbf{S} \cdot \mathbf{s}$, where J_{sd} is the exchange constant. Coherent scattering of electrons and magnons by the $s-d$ exchange interaction leads to creation of electron-hole pairs. The total angular momentum during $s-d$ exchange is conserved, and as a result, the electron spin flips to accommodate the magnon magnetic moment. The newly created spin-flipped electrons scatter incoherently off thermal excitations (phonons) in the material, causing broadening of the FMR linewidth. The damping contribution due to the exchange interaction

is given by

$$\alpha = \frac{s}{(S + s)(\omega_s \tau_{sf}^s)}$$

where S and s are the magnitudes of $\mathbf{S}$ and $\mathbf{s}$. $\omega = \frac{2J_{sd}S}{\hbar}$ is the precession frequency of s electrons in the $s - d$ exchange field and τ_{sf}^s represents the spin relaxation time [18].

In addition to the factors mentioned above, several other mechanisms contribute to resonance linewidth widening. In sputtered films, local inhomogeneity in the magnetic parameters of the material leads neighbouring grains to resonate at slightly different fields, hence creating broadened resonance signals. This behaviour does not scale with frequency and is an extrinsic contribution [19].

2.2.2 Damping in Thin Film Multilayers

It has been reported that ferromagnetic multilayer thin films exhibit relatively higher damping values, due to the presence of additional relaxation channels in comparison with bulk magnets. The effects of these extra channels are observed in the form of increased resonance linewidths in multilayers [20]. This increase is particularly notable in multilayers with ferromagnetic-heavy metal (FM-HM) interfaces, which have been explored for various spintronic applications.

Various theoretical models have been proposed to explain the enhancement of damping in FM-HM heterostructures. Berger's model [21] predicts an increased linewidth in FM-HM-FM compositions due to the presence of additional exchange torques along the direction of the Gilbert damping torque. The origin of this torque at the interface is believed to be electron-magnon interactions between conduction s electron and magnetic d orbitals in the magnetic layer, leading to spin-flip transitions via annihilation of magnons.

In FM-HM multilayers, an additional contribution to the Gilbert damping emerges from the 'spin-pumping' [22, 23] effect, in which the magnetization precession acts as a pump transferring the angular momentum from the ferromagnet to the heavy metal. The angular momentum transfer does not involve charge motion and can be written as a spin current:

$$\mathbf{I}_s^{pump} = \frac{\hbar}{4\pi} \left(C_r \mathbf{m} \times \frac{d\mathbf{m}}{dt} - C_i \frac{d\mathbf{m}}{dt} \right) \quad (2.34)$$

where $C \equiv A_r + iA_i$ is the ‘spin pumping conductance’ [22] which is determined by the spin reflection and transmission coefficients of the ferromagnet. Following the conservation of angular momentum, the adiabatic spin emission can be written as a torque acting on the ferromagnet. Including this additional term in the LLG equation leads to a modification of the gyromagnetic ratio (γ) and the damping coefficient (α):

$$\frac{1}{\gamma} = \frac{1}{\gamma_0} \left[1 + g_L \frac{C_i}{4\pi m} \right] \quad (2.35)$$

$$\alpha = \frac{\gamma}{\gamma_0} \left[\alpha_0 + g_L \frac{C_r}{4\pi m} \right] \quad (2.36)$$

where g_L is the Landé g factor, m is the total magnetization and subscript 0 represents the bulk values. Equations 2.35 and 2.36 are only valid for ideal spin sinks, which can dissipate the incoming spin current. In the absence of such mechanisms, the incoming spins accumulate in the HM and eventually start diffusing into the ferromagnet, generating a reverse spin current $\mathbf{I}_s^{back}$. In this case, the net spin current is $\mathbf{I}_s = \mathbf{I}_s^{pump} - \mathbf{I}_s^{back}$ (figure 2.4). With the inclusion of the ‘additional’ torque term arising from the spin pumping, the LLG equation can be written as [22],

$$\frac{d\mathbf{m}}{dt} = -\gamma \mathbf{m} \times \mathbf{H}_{eff} + \alpha_0 \mathbf{m} \times \frac{d\mathbf{m}}{dt} + \frac{\gamma}{M_s V} \mathbf{I}_s \quad (2.37)$$

where α_0 is the bulk Gilbert damping. The last term on the RHS represents the effect of spin pumping and is directed along the damping torque, resulting in an increase of the effective damping in the film.

FIGURE 2.4: Schematic illustrating spin pumping at a ferromagnet-nonmagnetic metal (FM-NM) interface. Magnetic precession in the FM injects a spin current $\mathbf{I}_s^{pump}$ into the NM. In the absence of a proper diffusion mechanism for incoming spins, a reverse spin current $\mathbf{I}_s^{back}$ is generated in the NM. Typically a heavy metal is used as a nonmagnetic metal.

Eddy current effects in FM-NM layered materials

A variation in the microwave magnetic field can induce eddy fields in metals. Typically, the magnetic fields generated by eddy currents in the nonmagnetic layer are not in phase with the applied RF field. This introduces a phase lag between the precession magnetization and the external RF field, causing an increase in the anti-symmetric component of the FMR linewidth. More recently, it was observed that in FM-NM-FM type structures, the amplitude and phase of magnetization precession in the two FM layers differ significantly [24], which was attributed to the eddy currents in the nonmagnetic layer [25]. Moreover, it has been demonstrated that eddy currents in thick nonmagnetic layers can shield the applied microwave field, thus causing a significant decrease in the amplitude of the resonance [26].

2.3 Instrumentation for Broadband Ferromagnetic Resonance

The fundamental working principle of a FMR spectrometer is to excite the magnetization (from the equilibrium state) using a microwave and subsequently measure the microwave absorption in the ferromagnet. A typical experimental set-up includes an electromagnetic source (≈ 10 MHz - 50 GHz), a detector to measure the absorption/transmission of the microwave, a waveguide to couple the microwave with the magnetic film and coaxial transmission lines to link the source, waveguide and detector. The basic components of an FMR spectrometer are displayed in figure 2.5. Typically, the waveguide is mounted on a sample holder placed in a magnetic field. A Hall sensor is used to record the magnetic field. In low temperature apparatus, an additional heater-temperature sensor assembly is included to control and monitor the temperature.

A FMR experiment can be performed via two protocols. In field sweep FMR, a fixed frequency microwave is applied to the material, in synchronisation with a varying magnetic field. Absorption (or transmission) spectra as a function of magnetic field are used to extract the resonance field and linewidth, which can be analysed to calculate the effective magnetization, gyromagnetic ratio and effective Gilbert damping. Field sweep measurements are useful to characterise the resonance spectra above the saturation field, where varying the field does not alter the magnetic state of the material. For field sweeps below the saturation limit, a

FIGURE 2.5: Schematic of a VNA-based broadband FMR set-up. A coplanar waveguide (CPW) sample holder is positioned between the poles of an electromagnet and an in-plane (IP) magnetic field $\mathbf{H}$ is applied to the film. The VNA acts as both source and detector for the microwaves.

continuous evolution of the magnetic phase diagram can generate artefacts such as mode softening [27].

The limitations of the field sweep FMR method can be eliminated by ‘frequency sweep’ measurements, where the frequency is swept at a constant magnetic field. This technique is more suitable to distinguish the resonant responses of distinct magnetic phases, as the desired magnetic order can be stabilised by fine-tuning the magnetic field. This technique is particularly useful in investigating the dynamic response of magnetic features which are stable only within a limited magnetic field range, or magnetic phases with a strong dependence on the magnetic history of material, such as skyrmions.

In traditional methods of measuring FMR (known as ‘Cavity FMR’), a klystron is used to generate microwaves. A metallic cavity couples the sample to the microwaves and a Schottky barrier diode acts as a detector. A circulator is connected between the sample and the EM source to prevent mixing incident and reflected waves. The signal sensitivity is enhanced by using a lock-in technique. The primary limitation of this approach originates from the cavity, which can support only a limited range

of frequencies (restricted by its geometry and dimensions). This form of apparatus is therefore unsuitable for ‘frequency sweep measurements’ and only allows ‘field sweep measurements’.

Another more common technique utilises a vector network analyser (VNA) and a co-planar waveguide (CPW). This is referred to as ‘broadband’ VNA-FMR, due to its wide range of operating frequencies. A VNA serves as both source and detector of the microwaves, and can support a wide range of frequencies at variable power levels. For VNA-based FMR spectrometers, the coupling between microwave and the material is achieved by a CPW. A CPW supports a wide range of frequencies and can hence utilise the full capabilities of the VNA. Additional benefits of CPWs include smaller footprint and lithographic design, which allows its integration into complex electronic circuitry.

2.3.1 Transmission and Coupling of RF Signals

The general characteristics of microwave propagation in transmission lines (TL), which include microchips, wave-guides and coaxial cables, are governed by the Telegrapher’s equations. In all true TLs, the electric and magnetic fields lie perpendicular to the direction of wave propagation. Such propagation modes are referred to as TEM modes, due to the transverse Electric and Magnetic fields.

A short coaxial cable of length δx can be utilised to derive the transmission line characteristics (figure 2.6a). Unlike constant DC signals, RF currents depend on the length of the cable as well as time. The advantage of considering a small size element is that the electrical effects are instantaneous, thus allowing the use of circuit theory. A transmission line consists of both conduction and displacement currents (due to the electric field generated by the time-varying magnetic field of the microwave) and the associated impedances.

Conduction current impedance effects include a resistance R (due to losses in the conductor) and an inductance L (due to magnetic flux linked with the current path). Displacement current effects include a conductance G (due to dielectric loss between the conductors) and a capacitance C due to charge accumulation on each conductor. The voltage and current differences at the two ends of conductor are

$$dV = -(R\delta x I + L\delta x \frac{\partial I}{\partial t}) \quad (2.38)$$

FIGURE 2.6: (a) Circuitry for deriving the Telegrapher's equations. δx is a small section of a transmission line. The resistance R , inductance L , capacitance C and conductance G depend on the material and geometry of the TL. (b) Two port model for determination of scattering parameters using a VNA. a_1 (b_1) and a_2 (b_2) are the incident (reflected) intensities at ports 1 and 2.

$$dI = -(G\delta x V + C\delta x \frac{\partial V}{\partial t}) \quad (2.39)$$

$$\frac{\partial V}{\partial x} = -(RI + L \frac{\partial I}{\partial t}) \quad (2.40)$$

$$\frac{\partial I}{\partial x} = -(GV + C \frac{\partial V}{\partial t}) \quad (2.41)$$

Assuming a sinusoidal time dependence, the voltage and current can be expanded as $V(x, t) = \text{Re}(\tilde{V}(x)e^{i\omega t})$ and $I(x, t) = \text{Re}(\tilde{I}(x)e^{i\omega t})$. The new forms of the equations are as follows:

$$\frac{\partial \tilde{V}}{\partial x} = -(R + i\omega L)\tilde{I} \quad (2.42)$$

$$\frac{\partial \tilde{I}}{\partial x} = -(G + i\omega C)\tilde{V} \quad (2.43)$$

$$\frac{\partial^2 \tilde{V}}{\partial^2 x} = \gamma^2 \tilde{V} \quad (2.44)$$

$$\frac{\partial^2 \tilde{I}}{\partial^2 x} = \gamma^2 \tilde{I} \quad (2.45)$$

where $\gamma = \sqrt{(R + i\omega L)(G + i\omega C)}$.

The solution of the Telegrapher's equations yields

$$\tilde{V}(x) = \tilde{V}^+ e^{-\gamma x} + \tilde{V}^- e^{-\gamma x}$$

$$\tilde{I}(x) = \tilde{I}^+ e^{-\gamma x} + \tilde{I}^- e^{-\gamma x}$$

The characteristic impedance is defined as,

$$Z_c = \frac{\tilde{V}^+}{\tilde{I}^+} = \frac{\tilde{V}^-}{\tilde{I}^-} = \sqrt{\frac{(R + i\omega L)}{(G + i\omega C)}} \quad (2.46)$$

The impedance of a transmission line is a function of its inductance and capacitance, which depend on the geometry and material parameters of the transmission line. Next, we will extend this approach to the CPW in order to understand the origin of the signal transmission/reflection induced by coupling magnetic materials with a CPW.

Coplanar Waveguide

Structurally, a CPW consists of a thin metallic stripe surrounded by two grounded electrodes, deposited over a dielectric. The electrodes and the central conductor are isolated via an air gap (figure 2.7a). The width of the central conductor w and the gap b between the central and ground conductors govern the transmission characteristics of a CPW. Typically, the dielectric thickness is a few times the gap width. A microwave signal is applied to the central conductor of a CPW, and the resulting oscillating magnetic field, the quantity of interest for FMR experiments, forms a closed loop around the central conductor. For practical purposes, the orientation of the magnetic field is considered to be in-plane.

The effective dielectric constant of a CPW is given by

$$\epsilon_{eff} = \frac{\eta_r + 1}{2} \quad (2.47)$$

where η_r and 1 are the relative dielectric constants of the substrate and air, respectively. The impedance of a CPW can be determined from its geometric parameters by the following [28]:

$$\begin{aligned} Z_c &= \frac{\eta_0}{\pi\sqrt{\epsilon_{eff}}} \log \left(2\sqrt{\frac{b}{w}} \right) & 0 < \frac{w}{b} < 0.173 \\ &= \frac{\pi\eta_0}{4\sqrt{\epsilon_{eff}}} \log \left[\frac{1 + \sqrt{w/b}}{1 - \sqrt{w/b}} \right]^{-1} & 0.173 < \frac{w}{b} < 1 \end{aligned} \quad (2.48)$$

To ensure maximal signal transfer, our CPWs are designed to operate at 50 Ω .

In our experiments, a U-shaped CPW is used to couple a microwave field to the magnetic material under test. The CPW design is shown in figure 2.7b. The

CPW was fabricated using Rogers™ RT6010 substrate, which offers stable low-loss performance at low temperatures. The temperature stability is particularly critical in this case, as the CPW is intended for use at 4 K. Silver-plated copper was used as the conductor for the CPW.

FIGURE 2.7: (a) Electromagnetic field distribution in a CPW, due to a microwave signal propagating in the central conductor of width w . The microwave magnetic field is directed tangentially on the surface, and perpendicular to the surface in the gap between the central conductor and grounded conducting stripes at the edges. (b) Schematic of the CPW used in our experiments with dimensions in mils. (c) CPW mounted on the sample stage. The two ends of the CPW are connected to K-type end launch connectors.

The magnitude of the microwave magnetic field across the waveguide is equal to $I/2w$ (in units of Am^{-1}), where I is the current and w is the width of the central conductor [29]. In our case, the source power was set at 5 dBm, which is equivalent to 3.162 mW. The voltage and current over a $50 \, \Omega$ impedance are 0.40 V and 7.95 mA (both RMS quantities). The CPW central conductor width is $116.84 \, \mu\text{m}$, which generates a RF magnetic field of 0.04 mT. The actual amplitude of the microwave field can be further reduced due to signal attenuation between the VNA and the CPW.

Origin of a Resonance Signal

A magnetic sample coupled to a CPW will exhibit oscillations in its magnetization as described by the LLG equation. The in-plane magnetization m_y oscillations lead to a flux variation $\delta\phi$ across the CPW, calculated as

$$\delta\phi = \mu_0 m_y(t) l \delta x \quad (2.49)$$

where l is the thickness of the material and δx is the length over the CPW. The temporal flux variation induces an emf in the CPW. Using Maxwell's equations,

the induced voltage is calculated to be

$$\delta V = -\frac{\delta\phi}{dt} = -\mu_0 \frac{\delta m_y(t)}{dt} l \delta x \quad (2.50)$$

Considering a sinusoidally varying magnetization (which can be ensured by using a low RF field amplitude h_y), the derivative can be replaced by $i\omega$ or $i2\pi f$. We then obtain

$$\delta V = -\mu_0 i 2\pi f m_y(t) l \delta x \quad (2.51)$$

Since the in-plane deflection of the magnetization m_y can be related to h_y by the susceptibility relation, $m_y = \chi_{yy} h_y$ and $h_y = \frac{I}{2w}$, we may write

$$\frac{\delta V}{\delta x} = -i 2\pi f \chi_{yy} \frac{\mu_0 l}{2w} I \quad (2.52)$$

Following the Telegrapher's equations, the induced magnetization oscillation can be considered as an additional impedance, given by

$$\Delta L = \chi_{yy} \frac{\mu_0 l}{2w} \quad (2.53)$$

Due to the complex nature of χ_{yy} , the change in inductance ΔL has both real and imaginary parts. This additional inductance causes a variation in the impedance Z of the CPW, deviating from 50Ω . This mismatch leads to an additional reflection of the signal, which is measured by the VNA as an increased S_{11} parameter (with a corresponding decrease in S_{21}).

RF Coaxial Cables

RF coaxial cables were used to transmit the signal between the VNA and CPW. The general structure of a coaxial cable includes a central conductive wire surrounded by an insulating dielectric. A conductive tube shield forms the outermost layer of cable. In some cases, an additional nonconductive layer is used as the outermost protective layer. The RF signal is applied to the innermost conductor while grounding the conductive shield. The associated electromagnetic field is confined in the dielectric between two conductors. The frequency range for signal transmission is controlled by the diameter of the inner conductor and dielectric. Typically copper or silver-coated copper is used for the central conductor and tin-plated copper or tin-plated aluminium is used for the outer layer.

In our experiments, we used non-magnetic semi-rigid cables in the section of our probe between the poles of electromagnet. This limits the effect of the additional

torque on the cables due to the finite magnetic field and hence improves the signal to noise ratio. For the rest of the connections, we used hand-formable cables with 2.4 mm connectors.

2.3.2 The Vector Network Analyser

The VNA is an essential part of a broadband FMR probe, operating as both source and detector for microwaves. A major advantage of a VNA is its ability to simultaneously measure the magnitude and phase of a microwave signal, which is necessary for complete characterisation of a device under test (DUT). The RF signal from our VNA is sourced at an impedance of $50\ \Omega$. All our transfer lines and loads are designed to be of $50\ \Omega$ impedance, to minimise the signal reflection between different connectors, cables and devices.

The high frequency response of a DUT is characterised in terms of the reflection (R) and transmission (T) coefficients. The reflection coefficient is the ratio of reflected signal voltage to the incident signal voltage:

$$R = \frac{V_{reflected}}{V_{incident}} = \frac{Z_l - Z_0}{Z_l + Z_0} \quad (2.54)$$

where Z_0 is the characteristic line impedance $= 50\ \Omega$ and Z_l is the load impedance. For $Z_0 = Z_l$, there will be no reflection. The transmission coefficient is the ratio of transmitted signal to the incident signal:

$$T = \frac{2Z_l}{Z_l + Z_0} \quad (2.55)$$

For a two-port electrical network (figure 2.6b), we can write

$$b_1 = S_{11}a_1 + S_{12}a_2 \quad (2.56)$$

$$b_2 = S_{21}a_1 + S_{22}a_2 \quad (2.57)$$

where the indices 1 and 2 refer to the two ports on either side of the device and a , b are the incident and reflected voltage amplitudes. The S parameters can be

extracted from a and b as follows:

$$S_{11} = \frac{b_1}{a_1}|_{a_2=0}, \quad S_{12} = \frac{b_1}{a_2}|_{a_1=0} \quad (2.58)$$

$$S_{21} = \frac{b_2}{a_1}|_{a_2=0}, \quad S_{22} = \frac{b_2}{a_2}|_{a_2=0} \quad (2.59)$$

All S parameters are complex quantities: their real and imaginary components can be resolved separately by a VNA and plotted as a function of frequency. The frequency dependence of the reflection and transmission parameters can be written as [30]

$$S_{11} = \frac{i\omega L + R + \frac{Z_0}{1+Z_0(G+i\omega C)} - Z_0}{i\omega L + R + \frac{Z_0}{1+Z_0(G+i\omega C)} + Z_0} \quad (2.60)$$

$$S_{21} = \frac{2\frac{Z_0}{1+Z_0(G+i\omega C)}}{i\omega L + R + \frac{Z_0}{1+Z_0(G+i\omega C)} + Z_0} \quad (2.61)$$

where ω is the microwave frequency, L , R , G and C are the inductance, resistance, conductance and capacitance of the transmission line and Z_0 is the characteristic impedance of the CPW (50Ω). From equations 2.60 and 2.61, we obtain

$$i\omega L + R = \frac{1 + S_{11} - S_{21}}{1 - S_{11}} Z_0 \quad (2.62)$$

The change in the inductance L due to magnetization precession (equation 2.53) can be related to the S parameters. The parameters recorded after the onset of precession can be written as $S_{11}^1 = S_{11}^0 + \Delta S_{11}$ and $S_{21}^1 = S_{21}^0 + \Delta S_{21}$. The superscript 0 refers to the S parameter in the absence of any magnetic load. The magnetic susceptibility is calculated using

$$\chi(H) = \chi_0 \left(\frac{1 + S_{11}^1 - S_{21}^1}{1 - S_{11}^1} - \frac{1 + S_{11}^0 - S_{21}^0}{1 - S_{11}^0} \right) \quad (2.63)$$

where χ_0 is a constant which depends on the material parameters [31]. In the weak reflection limit ($|S_{11}| \ll 1$), the susceptibility reduces to

$$\chi(H) \simeq \chi_0[(2 + 2S_{11}^0 - S_{21}^0)\Delta S_{11} - (1 + S_{11}^0)\Delta S_{21}] \quad (2.64)$$

In the quasi-classical limit $\Delta S_{11} = -\Delta S_{21}$ and the susceptibility further simplifies to

$$\chi(H) \simeq -\chi_0[3(1 + S_{11}^0) - S_{21}^0]\Delta S_{21} \quad (2.65)$$

If the resonance field and linewidth are the only quantities of interest, the S_{21} data can be fitted with a simplified model [31]:

$$S_{21}(H, t) = S_{21}^0 + Dt + \frac{\chi}{\tilde{\chi}} \quad (2.66)$$

D is a fitting parameter which accounts for the linear drift in the VNA stability over time and $\tilde{\chi} = \chi_0[3(1 + S_{11}^0) - S_{21}^0]$.

$\chi(H)$ can be approximated by a superposition of symmetric and anti-symmetric Lorentzian functions (figure 2.8) [32–34]:

$$\chi(H) = A_{sym} \frac{(\Delta H)^2}{(H - H_{Res})^2 + (\Delta H)^2} + A_{asym} \frac{(\Delta H)(H - H_{Res})}{(H - H_{Res})^2 + (\Delta H)^2} \quad (2.67)$$

Here H_{Res} is the resonance field for a given frequency, ΔH is the full-width half-maximum (FWHM) and A_{sym} , A_{asym} are the amplitudes of the symmetric and anti-symmetric functions respectively. For a frequency sweep spectrum, H , ΔH and H_{Res} are replaced by the corresponding frequency sweep quantities f , Δf and f_{Res} [35].

FIGURE 2.8: Field sweep FMR spectra (a) fitted with a superposition of symmetric (b) and anti-symmetric (c) Lorentzian functions. Symmetric and anti-symmetric spectra are proportional to the variation in imaginary and real components of the susceptibility [29, 32]. Details of the fitting procedure are included in the appendix.

2.3.3 Sample Holder and the Magnet Configurations

In this section, we present the details of our home-built FMR set-up, which allows low-temperature measurements down to 80K. The sample stage is housed in a

vacuum chamber which fits between the poles of an electromagnet (figure 2.9a). The vacuum chamber is connected to a turbomolecular pump backed by a rotary pump. The electromagnet is driven by a Lakeshore 625 bipolar power supply and can generate fields up to ± 0.55 T. Both the electromagnet and the vacuum chamber are mounted on a vibration-free platform. The vacuum chamber comprises four sub-stages, built in a concentric manner (figure 2.10). The probe is located inside the inner-most chamber. This multichamber design allows a gradual decrease between the temperatures of the sample stage and the outer environment, thus improving the thermal stability of the probe.

FIGURE 2.9: Nitrogen cooled thin film broadband FMR probe for operation with an electromagnet. (a) The vacuum chamber (2) lies between the poles (1) of the electromagnet. (b) Detailed view of the sample chamber showing the thermally anchored semi-rigid RF cables (3) and the rotating sample stage (4). (c) Close-up image of the sample holder which includes bobbin heaters (5), end launch connectors (6) and the spring loaded sample holder (7). (d) illustrates an alternative in-line design of the sample holder.

The sample stage is made of CuBe alloy. The large thermal and electrical conductivity, nonmagnetic nature and ease of machining CuBe make it an ideal choice for precision experiments on magnetic materials at cryogenic temperatures. A coplanar waveguide (CPW) is bonded to the sample stage using a thermally conductive epoxy to ensure uniform thermal contact between the sample stage and CPW. The

reverse side of the sample stage contains a thermal control assembly, in which a pair of bobbin heaters and a temperature sensor are used to control the temperature. A model 336 temperature controller from LakeshoreTM is connected to the heater and temperature sensor assembly (figure 2.9c). The 336 temperature controller allows multiple ranges for heater current and can accommodate a variety of temperature sensors, thereby increasing the versatility of the probe. Electrical current for these heaters is supplied via thick copper wires to minimise resistive heating, which can destabilise the temperature of the sample stage. Commercially available 2.4 mm end launch connectors (ELC) from Southwest MicrowaveTM (enabling microwave transmission up to 50 GHz) are screwed onto the edges of the sample stage. ELCs are standard connectors used for coupling coaxial cables with CPWs. The central pin of the ELC is soldered onto the central conductor of the CPW to avoid any parasitic inductance from an air gap between the CPW and ELC pin, hence maximising the signal transmission between coaxial cable and CPW. The other ends of the ELCs are connected to vacuum-sealed, thermally anchored semi-rigid coaxial cables (figure 2.9b) which transmit microwaves from the CPW to female-female (FF) RF adaptors located outside the vacuum chamber. These FF adaptors are connected to the VNA by hand formable coaxial cables. The set-up was controlled using a LabView program.

FIGURE 2.10: Schematic of the cooling stages in the nitrogen cooled FMR probe. Successive shielding stages, arranged in a Russian doll fashion, provide efficient thermal isolation of the sample stage. Heat arriving through the RF cables is eliminated by thermal anchoring to the sample stage.

During our experiments, we measure diced thin films of size $8 \times 4 \text{ mm}^2$. The films were first coated with a thin layer of insulating resist to avoid an electrical short between the signal and ground strips of the CPW. The films were mounted on the CPW using a spring-loaded clamp. For low temperature experiments, the vacuum

chamber was pumped down to 10^{-5} mbar. A pressurised liquid nitrogen dewar was connected to the probe for continuous cooling of the sample stage. Temperature set points were stabilised by balancing the cooling power of liquid nitrogen (by manually controlling its flow rate) with the heater output, controlled by the Model 336. Our experimental data were analysed using MATLAB scripts: details of the fitting methods are included in chapter 4 and the appendix.

4K FMR Probe

In addition to the electromagnet-based apparatus described in the previous section, FMR experiments were also performed in another home-built FMR probe which operates in a superconducting vector magnet [36, 37]. A 3-stage dipper probe (figure 2.11) can stabilise temperatures down to 4.2 K. A large magnetic field can be applied either parallel or perpendicular to the film surface. Our superconducting vector magnet is manufactured by JanisTM Research Company Inc. and CryomagneticsTM Inc.: it incorporates solenoid and split pair coils to generate magnetic fields of 9 T (vertical direction) and 4 T (horizontal direction) respectively. The dipper probe is equipped with a Hall sensor, mounted close to the film for accurately measuring the magnetic field. The probe is mounted inside a cylindrical vacuum chamber, which is immersed in a liquid helium bath to provide the necessary cooling power. The sample stage is fitted with $50\ \Omega$ thin film resistors, for use as heaters, which are connected to the heater output of a Lakeshore 340 temperature controller. A Cernox temperature sensor is mounted on the reverse side of the sample stage. The temperature sensor and resistor assembly control the temperature of the sample stage. An additional temperature sensor is mounted on the upper stages of the probe for monitoring purposes. The probe utilises a spring-loaded sample holder design to place the film directly above the centre strip of a CPW in flip-chip geometry. The advantages of this probe include a lower minimum temperature, higher magnetic field and better thermal stability.

FIGURE 2.11: Home-built broadband FMR probe with an operational range of 4.2 -300 K and 0-9 T.

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Chapter 3

Multilayer Heterostructures for Room Temperature Skyrmions

Non-collinear spin textures stabilised by an interplay of spin canting and spin aligning interactions are promising candidates for fundamental physics as well as technological applications. In the last decade, metallic multilayers hosting chiral Néel domain walls and skyrmions have been intensively studied in the hope of achieving fast, energy efficient current-induced dynamics in spintronic devices. This chapter details the architecture and composition of ferromagnet - heavy metal (FM-HM) multilayers hosting nanoscale Néel skyrmions at room temperature.

3.1 Skyrmions in Non-centrosymmetric Magnets

Spin-orbit coupling and a lack of spatial inversion symmetry generate a finite DMI in non-centrosymmetric magnets. An important class of such materials crystallise with space group $P2_13$, including metals (MnSi , $\text{Fe}_{1-x}\text{Co}_x\text{Si}$, FeGe) as well as insulators (Cu_2OSeO_3). MnSi and FeGe are two widely explored non-centrosymmetric magnets which are known to exhibit helical magnetism [3, 4]. Magnetic interactions follow a hierarchical order in these materials: Heisenberg exchange being the strongest, followed by the Dzyaloshinskii-Moriya (DM) interaction and magnetic anisotropy. This sequence of energy scales leads to a similar phase diagram among the magnetic materials in the $P2_13$ space group (figure 3.1). The magnetic order evolves from a helical state (where neighbouring spins rotate in the plane perpendicular to the direction of propagation of the helix) to conical states with an increase in external

FIGURE 3.1: Structure and phase diagram of B-20 non-centrosymmetric magnets. (a) Crystal structure and (b) phase diagram of MnSi. Blue and yellow spheres represent Mn and Si atoms. (c) Phase diagram of Cu_2OSeO_3 . Data points for (b) and (c) are from [1] and [2].

magnetic field. A field polarised phase is eventually reached at large fields. The direction of spin rotation (clockwise or anticlockwise) is restricted by the DM vector, which itself depends on the crystal structure [5]. The skyrmion lattice, historically referred to as the *A* phase in MnSi, exists in a narrow region near the ferromagnetic-paramagnetic transition temperature $T_c = 29.5$ K.

Following theoretical predictions of skyrmions in non-centrosymmetric magnets [6], small angle neutron scattering (SANS) experiments revealed the spin texture of the *A* phase to be a superposition of three coplanar helices aligned at 120 degrees [7], commonly known as a triple-Q structure. Theoretical calculations based on SANS data indicated that the triple-Q structure originated from a Bloch skyrmion lattice. The presence of skyrmions in MnSi was further confirmed in magnetotransport measurements, where skyrmions added a ‘topological’ contribution [1] to the Hall resistance, beyond the normal Hall effect (proportional to applied field) and the anomalous Hall resistance (which scales with magnetization). It was demonstrated that the critical currents required to move skyrmions were of the order of 10^6 Am^{-2} , five orders of magnitude less than for domain walls (DWs) [8]. Such energy efficient dynamics triggered intense research into potential spintronic applications of skyrmions.

FeGe is another well-studied magnet belonging to the $P2_13$ space group. Lorentz transmission electron microscopy (LTEM) experiments (in which electrons passing through the ferromagnet are deflected due to the Lorentz force from the local magnetization, creating bright and dark contrast in images) have revealed a stable

Material	Space group	Skyrmions	T_c (K)
MnSi [1, 7]	P2 ₁ 3	Bloch SkL	29
Fe _{1-x} Co _x Si [9, 10]	P2 ₁ 3	Bloch SkL	x= 0.10, T_c = 43
			x=0.5, T_c = 90
			x=0.6, T_c = 174
			x=0.7, T_c = 230
FeGe [11]	P2 ₁ 3	Bloch SkL	278
Cu ₂ OSeO ₃ [12]	P2 ₁ 3	Bloch SkL	Bulk, T_c = 59
Co _x Zn _y Mn _z [13]	P4 ₁ 31	Bloch SkL	x= 8, y =8, z= 4, T_c = 320
			x= 8, y =9,z = 3 , T_c = 330
			x= 8, y =10,z = 2 , T_c = 360
			x= 10, y =10,z = 0 , T_c = 480
MnGe [14, 15]	P2 ₁ 3	3D Bloch SkL	170
GaV ₄ S ₈ [16]	R3m	Néel SkL	13

TABLE 3.1: Magnetic skyrmions in non-centrosymmetric magnets.

skyrmion phase at temperatures up to 278 K [11] in thin films of FeGe. The stability of the skyrmion phase varies with the ratio of sample thickness to the skyrmion lattice constant. A related observation is the shrinking of the skyrmion phase temperature window in thicker films, which extends down to 60 K in 15 nm thin films, compared with only 250 K in 75 nm thick films. Due to its high Curie temperature (278.2 K) and multiple skyrmion configurations, FeGe has been studied to understand the dynamics of individual skyrmions, inter-skyrmion interactions and skyrmion-material edge interactions [17]. A key fundamental discovery is the quantised nature of skyrmion nucleation in confined geometries, where transitions between neighbouring stable states are achieved by formation/deletion of one skyrmion [18].

The dynamics of skyrmion crystals in nanodisks were investigated via LTEM experiments. The observation that a radial thermal gradient for the electron beam induced a ratchet motion of the skyrmion crystal in MnSi, Cu₂OSeO₃ [19] and FeGe [18] suggested a low energy requirement for skyrmion motion. The direction of rotation (clockwise or anticlockwise) depends on the sign of the temperature gradient between the centre and edge of the disk. Moreover, the three materials differed in spin order length scales and transition temperatures, suggesting that such motion is an intrinsic property of the skyrmion lattice and independent of material parameters.

Bloch skyrmion lattices provide an excellent platform to understand the fundamentals of skyrmion physics. However, low Curie temperatures, difficult fabrication methods and limited tunability of magnetic parameters restrict their useability for technological purposes (table 3.1). Moreover, their stable ground state consists of a skyrmion lattice, which has limited applications. One such example is in magnonics, where a skyrmion crystal can function as a grating for microwaves. For data storage and logic applications, it is critical to achieve control of isolated skyrmions, which can be utilised as logic bits [20, 21]. Recently discovered [22, 23] Néel skyrmions in ferromagnet-heavy metal (FM-HM) multilayers can exist in an isolated state at room temperature. Furthermore, magnetic interactions at the interface can be modulated to drive transitions from lattice to disordered (and vice-versa) skyrmion configurations. The next sections are dedicated to the optimisation of magnetic interactions at such interfaces by varying the composition and architecture of the interface.

3.2 Magnetic Interactions at Ferromagnet-Heavy Metal Interfaces

A surface can be described as a two-dimensional imperfection in a crystal structure, which breaks the translational and spatial inversion symmetry of the material. The abrupt change of symmetry at the surface is accompanied by sudden changes in the electronic structure and crystal potential. While atoms in the bulk are strongly bonded, weakly bonded surface atoms result in enhanced chemical reactivity and surface reconstructions. Consequently, surface atoms exhibit distinct properties from their bulk counterparts. Typical examples include catalysis in chemistry,

friction in mechanics, surface tension in fluids etc.. In ferromagnets, surfaces are associated with a magnetic charge, generating an additional ‘magnetostatic contribution’ to the energy landscape of the material.

Surface-induced effects play a dominant role at nanometer length scales, due to an enhanced surface/volume ratio. Novel effects at ferromagnet-heavy metal (FM-HM) interface include [24, 25]:

- a) Proximity induced magnetization, where a ferromagnet induces a magnetic moment in an adjacent heavy metal which decays with the distance from the interface. The magnitude of induced magnetization is correlated with the electronic configuration of the heavy metal, and in general scales with the number of unpaired electrons in d orbital [26]. Technologically, proximity-induced magnetization is known to enhance domain wall velocities due to chiral spin torques [27].
- b) Perpendicular magnetic anisotropy (PMA), which stabilises out-of-plane (OP) magnetization in thin films and has been harnessed for myriad spintronic applications. A typical example is the use of OP magnetized bits in hard disk drives, offering superior thermodynamic stability and space efficiency in comparison to in-plane (IP) bits. More recent investigations have been directed towards understanding and implementing PMA to facilitate non-collinear magnetic order in low dimensional magnets.
- c) Spin-charge conversions and related torque effects, allowing efficient coupling between charge currents and spin textures and thus enabling novel methods for manipulation of magnetic order.

In this thesis, we are primarily interested in understanding the role of a FM-HM interface in modulating the magnetic interactions and characteristics of a ferromagnet. We start with the interfacial Dzyaloshinskii-Moriya (DM) interaction at the FM-HM interface, which arises due to a robust spin-orbit coupling (SOC) from the HM layer and the lack of spatial inversion symmetry.

At an FM-HM interface, the DMI vector $\mathbf{D}_{AB}$ lies in the plane of the interface and is oriented perpendicular to $\mathbf{R}_{AB}$, connecting spins at sites A and B (figure 3.2a). Depending on the orientation of $\mathbf{D}_{AB}$, spins rotate around $\mathbf{D}_{AB}$ in a right-handed ($\uparrow \rightarrow \downarrow \leftarrow \uparrow$) (for $\mathbf{D}_{AB}$ into the page) (figure 3.2b) or left-handed ($\uparrow \leftarrow \downarrow \rightarrow \uparrow$) ($\mathbf{D}_{AB}$ out of the page) fashion. The direction of spin rotation, controlled by the DMI, is referred to as the ‘chirality’ of the magnetic order and plays a critical role in non-collinear magnetism. For example, the chirality of DWs dictates their propagation direction and velocity during spin-orbit torque (SOT) induced motion [28]. The

action of spin-orbit torques is characterised in terms of the spin Hall angle, which varies with FM-HM composition and hence can be engineered at the interface.

FIGURE 3.2: (a) Interfacial DMI at a FM-HM interface. The interaction between spins S_A and S_B (red spheres) is mediated by a nonmagnetic HM atom (blue sphere). $\mathbf{R}_A$ and $\mathbf{R}_B$ are the position vectors of the spins with respect to the HM atom. $\mathbf{R}_{AB}$ connects spins S_A and S_B . The DMI vector $\mathbf{D}_{AB}$ lies in the plane of the interface and induces a Néel rotation of the spins, displayed in (b).

Experimental Determination of the DMI

Various experimental techniques have been developed for accurately determining the DMI in thin films. Typically, experimental methods harness either the asymmetric domain wall expansion of magnetic bubbles, attributed to an additional in-plane (IP) field due to the DMI, or asymmetric spin-wave propagation induced by the DMI. Details of various techniques based on these two methods are presented below.

1. Spin-Wave Propagation Based Techniques

(a) Brillouin Light Spectroscopy (BLS)

BLS measurements are performed in a Damon-Eshbach (DE) geometry, where the film is magnetized IP by a tangential magnetic field and spin waves propagate perpendicular to the magnetization direction [29, 30]. Oppositely propagating spin waves have clockwise and anticlockwise spin chirality. In the absence of a DMI, the dispersion relation is symmetric for both chiralities. However, a finite DMI favours a certain chirality over the other, inducing an asymmetry in the dispersion relation (figure 3.3a). The volume averaged value of the DMI can be calculated from the measured frequency asymmetry in oppositely-propagating spin waves.

2. Domain Wall Based Techniques

In thin films, the interfacial DMI can be considered as an effective IP magnetic field, which modifies the DW magnetization orientation from Bloch to Néel type. The DMI field is calculated as $H_{DMI} = D_s/\mu_0 M_s t \Delta$, where t is the film thickness and $\Delta = \sqrt{A/K_0}$ is the DW width. For Néel DWs the internal magnetization of the wall is parallel to the wall width. For circular (bubble) domains, this leads to a radially evolving magnetization which is axisymmetric in direction and magnitude. This symmetry can be eliminated by an in-plane magnetic field which favours an anisotropic expansion of the domain walls (figure 3.3b and c), driven by an external current or OP field pulse.

FIGURE 3.3: (a) DMI induced asymmetry in spin-wave spectra, as observed in BLS experiments [29]. (b), (c) Asymmetric domain wall expansion of bubble domains in the presence of a DMI, imaged under opposite orientations of a longitudinal field [31].

(a) Current Induced Motion of Domain Walls

In magnetic nanowires hosting Néel DWs, the DMI can be modelled as an effective IP magnetic field along the wire axis. The DMI coefficient can be extracted from the longitudinal field value at which the DW velocity approaches zero [32]. The major limitations of this technique emerge from the assumptions regarding domain wall dynamics. This model assumes a negligible effect of spin transfer torques and other adiabatic torques, which is invalid for multilayers.

(b) Field Induced Motion of Domain Walls

As the name suggests, DW motion in this case is driven by a small OP field, in the presence of an IP field. The DMI field can be extracted from DW velocity vs. IP field graphs. A creep model fit to domain wall velocity data can be employed to extract the DMI field as a fitting parameter. The IP DMI field compensates the applied longitudinal field; consequently, the longitudinal field resulting in the minimum velocity of the domain wall is determined to be the DMI field [33]. In a more rigorous method, DW velocity data can be fitted with a creep law, and the DMI field can be extracted as a fitting parameter [31]. Typically such measurements are performed in tandem with imaging techniques such as MFM and Kerr microscopy to measure the domain wall width. The superiority of the field-driven DW motion technique over current-driven DW motion approaches can be attributed to its simple experimental set-up and less ambiguous data interpretation. Unlike current-driven DW motion, field-induced DW motion experiments do not require lithographically patterned nanodevices. Furthermore, current induced motion involves additional forces (such as spin transfer, Rashba and spin Hall torques) with field-like contributions, which cannot be clearly distinguished and therefore add uncertainty to the extracted DMI values. Another source of error is related to the precise internal structure of the DWs. Bloch and Néel walls follow distinct forms of creep law, and hence any uncertainty regarding the internal structure of the DW will lead to an erroneous fitting parameter. In the case of sputtered thin films, local inhomogeneity of the magnetic parameters can result in a variation of DW width for domains located in different parts of the film. Moreover, the presence of pinning sites can cause a deviation from standard creep

laws. The combined effect of all these factors can result in a significant error in extracting the DMI field and DMI coefficient.

(c) NV Magnetometer Scanning

The previous two methods utilise DW motion to determine the DMI coefficient. A more recent technique known as NV magnetometer scanning [34] utilises a nitrogen vacancy centre in a diamond tip to probe the stray magnetic field originating from a DW. This stray magnetic field is measured as a deviation in the resonance frequency of the NV centre. The frequency shift profile can be fitted with stray field models from Bloch or Néel walls, and the value of the DMI coefficient can be determined from the analysis of the extracted fitting parameters. The primary advantage of this method over DW motion methods is that it excludes any assumptions related to the DW structure and is relatively insensitive to the presence of defects and pinning sites.

The origin of the DMI at a thin film interface can be understood by the ‘three particle interaction model’ of Fert and Levy [35]. The two necessary components of the DMI, i.e. the absence of crystal inversion symmetry and a spin-orbit interaction, are provided by a nonmagnetic impurity located at the intersection of two $3d$ atoms (figure 3.2a). This model calculates the effect of localised d electron spins on the energy levels of a neighbouring conduction electron of a nonmagnetic impurity, in the presence of SOC. A perturbation from the localised spins introduces additional contributions to the free electron wave function and corresponding energy eigenfunction. The shift in energy levels of the conduction electron is determined by the magnitude of the SOC and the relative lattice structure of the interfacial atoms, and can be used to estimate the magnitude of the DMI.

Although the three-particle model works well at FM-HM interfaces, it is inadequate to describe multilayers comprising multiple FM-HM interfaces. Such multilayers pose extra complexity over the Fert-Levy model due to additional effects such as orbital hybridisation, proximity-induced magnetization and several other inter- and intra-layer interactions.

Theoretical work [26] on $3d - 5d$ interfaces has investigated the role of $3d$ orbital occupancy on the magnitude of the DMI and the correlation between the DMI and proximity-induced magnetization in the $5d$ metal. At a $3d - 5d(4d)$ interface, the net magnetization consists of two independent contributions. The major contribution

is from the intrinsic magnetization of the $3d$ ferromagnet, complemented by a relatively small proximity-induced magnetization in the adjacent heavy metal. Induced magnetization in heavy metals can couple ferromagnetically (for example in Pt and Ir) or antiferromagnetically (for W) with the ferromagnet. The net magnetization is a vector sum of both contributions. It has been calculated that the magnetization of the $3d - 5d$ stack peaks at Mn (for a given $5d$ metal) and decreases monotonically for elements on both sides, following the trend in the number of unpaired d spins in the ferromagnet (figure 3.4a). Furthermore, a reduced magnetization of an FM-HM composite relative to a free-standing FM layer suggests a quenching of magnetization at the interface. However, such observations should be explained with caution: a similar reduction in magnetization can also be attributed to an AFM coupling between the intrinsic and induced magnetizations. At a $3d - 5d$ interface, the DMI increases up to Mn and decreases beyond that, mimicking trends in the induced magnetization (figure 3.4b). These trends in the DMI are valid for most of the $5d$ heavy metals. Such similarity in the variations of proximity-induced magnetization and DMI across the $3d$ row suggests a correlation between the two quantities.

FIGURE 3.4: (a) Proximity-induced magnetic moment in a HM layer adjacent to a ferromagnet [26] (b) Calculated DMI at FM-HM interfaces for various $3d - 5d$ combinations. Both the proximity-induced magnetic moment and the DMI display a similar dependence on the number of unpaired d electrons in the ferromagnet, suggesting a correlation between the two quantities.

The strength of the DMI at a $3d - 5d$ interface can be deduced by analysing the spin-flip transitions between the e_g and t_{2g} orbitals of the ferromagnet, mediated by the SOC from the $5d$ underlayer. The probability of such spin-flip transitions is estimated by the relative positions of the spin up and down bands in the ferromagnet

with respect to the Fermi energy of the heavy metal, and the number of up or down spins available for the transition (figure 3.5). A smaller energy difference and a higher number of participating spins both lead to a larger DMI. In the case of V/Ni, elements at the extremities of the $3d$ orbital, the Fermi level resides deep within the occupied/unoccupied $3d$ bands, limiting the number of opposite spin orbitals available for transition. Consequently, these materials exhibit a weak DMI.

The dependency of the DMI on the $3d$ orbital filling can be understood by a simplified model. In the case of Mn (d^5), each of the 3 t_{2g} and 2 e_g orbitals hold single electrons with parallel spins. A low energy separation between the t_{2g} and e_g states facilitates spin flip transitions from e_g to t_{2g} . The relative strength of the DMI in $3d$ elements can be estimated by counting possible spin flip transitions. For Mn, two electrons from the e_g orbitals can populate three t_{2g} orbitals, resulting in six possibilities. For Fe (d^6), the t_{2g} orbitals contains two unoccupied states for spin flip transitions, thus reducing the possibilities to four. This number further decreases with Co (d^7), which allows only one empty state in the t_{2g} orbitals, reducing the possibilities to two. For Ni (d^8), a completely filled t_{2g} orbital does not allow any spin flip transitions, resulting in the lowest DMI. Similarly for all the $3d$ elements before Mn ($d < 5$), the absence of electrons in the e_g orbitals reduces the probability for spin flip transitions.

The $3d - 5d$ hybridisation alters the crystal field splitting of the $3d$ orbitals, thus changing the relative energy difference between the t_{2g} and e_g orbitals: a detrimental factor to spin-flip transitions. The d symmetry of the wave functions affects the hopping probabilities for electrons across the various orbitals. The sign of the DMI is correlated with the $5d$ orbital filling in the HM and the DMI coefficient is proportional to the expectation value of the SOC $\langle l.s \rangle$ [36]. The sign of this coupling reverses around the d^5 electron configuration of the heavy metal. In the case of Co and CoFeB, the DMI remains positive for HM with fewer than five outer electrons, while it flips to negative for d orbitals with more than five electrons ($d > 5$) [36].

The DMI strength is also sensitive to the relative alignment of the d bands in the ferromagnet and heavy metal. All d orbitals (d_{xy} , d_{yz} , d_{xz} , $d_{x^2-y^2}$, d_{z^2}) do not undergo the same form of hybridisation, and therefore have non-identical contributions to the DMI. It has been predicted that $5d_{xy}$ and $5d_{yz}$ bands couple strongly to the $3d$ orbitals, while others show weak coupling.

FIGURE 3.5: Spin-flip transitions between the e_g and t_{2g} orbitals of a ferromagnet, mediated by the relative position of the Fermi level (E_f) in the 3d ferromagnet and 5d heavy metal. The probability of spin-flip transitions increases with the number of unpaired d electrons and exhibits a maximum at Mn. The strength of the DMI at a 3d – 5d interface is proportional to the number of spin-flip transitions, and hence peaks at Mn. Diagram reproduced from [26].

Although the DMI is an interfacial phenomenon, a sharp dependence of the DMI strength on the thickness of FM and HM layers has been reported. Theoretical calculations suggest that 80 % of the DMI contribution arises from the top two 5d atomic layers, with the contribution from the top layer being significantly higher than the second one. This has been observed in Co/Pt interfaces [37], where the DMI increases sharply with the thickness of the Pt layer and saturates at 3 nm. An exception to the saturation of the DMI with HM thickness appears in FM/AFM materials, where the DMI strength increases with the addition of several AFM layers [38]. The DMI depends weakly on the thickness of the FM layer [39] and is known to exhibit an inverse dependence on the FM thickness [40].

3.3 Interface Optimisation in Multilayers for Tunable Skyrmions

As established in the previous sections, magnetic interactions in FM-HM thin films can be modulated with the composition and architecture of the interface. This approach has been utilised to tune the size and configuration of Néel skyrmions in thin films [41, 42]. The FM-HM interfaces can be extended to a HM(1)-FM-HM(2) design to exploit both surfaces of the ferromagnet. HM(1) and HM(2) can be identical or different metals, with varying thicknesses. Symmetric compositions are known for cancellation of interface effects [31]: they are not a suitable choice for this design. A reported exception is Pd/Co/Pd [43], where asymmetry in the proximity-induced magnetization of the top and bottom Pd layers is believed to generate a non-zero DMI. Based on the choice of heavy metal, additional top layers can be used to tune the exchange, DMI, or both interactions in the ferromagnet. For example, as observed in Fe/Ir films, an additional top layer of Pd modifies the size, lattice order and phase diagram of the emergent skyrmions. While Fe/Ir films host a skyrmion lattice with square symmetry, skyrmions in Pd/Fe/Ir exist in both isolated and ordered configurations [44]. The additional Pd layer does not alter the magnitude of the DMI significantly. The SOC responsible for the DMI is proportional to the magnitude of the nuclear charge and is dominated by the $5d$ configuration of Ir in Pd/Fe/Ir films. Addition of a Pd layer modifies the exchange interaction [41] of the Fe layer. Similar observations have been reported previously, where the ground state of Fe can be toggled from an anti-ferromagnetic to ferromagnetic state via successive filling of d orbitals in an adjacent heavy metal [45]. In addition to the DMI and exchange, FM-HM interfaces in ultrathin films are known to exhibit perpendicular magnetic anisotropy (PMA), which controls the thermodynamic stability and size of skyrmions. Since PMA is a consequence of SOC, it can also be manipulated by varying FM-HM combinations.

Though the addition of a Pd overlayer on the Fe/Ir interface does not improve the DMI strength of the stack, the same approach can be utilised to create HM($5d$)-FM($3d$)-HM($5d$) interfaces, in which both $5d$ heavy metals contribute to the DMI. This was experimentally demonstrated in Pt/Co/Ir(x)/Pt (x is the variable thickness) heterostructures [31] via domain wall expansion experiments, where two interfaces with opposite DMI additively generate a large DMI. For example, in Pt/Co/Ir/Pt heterostructures, $\mathbf{D}$ vectors at the Co/Pt and Co/Ir interfaces are

oriented in opposite directions and hence inversion of any of the interfaces will align $\mathbf{D}$ at both interfaces in the same direction. This approach has been successfully applied in Pt/Co/Ir [46], Pt/Co/W [47] and Pt/Co/Ta [48] to stabilise nanoscale skyrmions at room temperature (figure 3.6). An improvement on this approach is the inclusion of an additional FM layer, resulting in a HM(1)-FM(1)-FM(2)-HM(2) architecture. This was demonstrated in Pt/Co/Fe/Ir [42] multilayers, where opposite DMIs at the two interfaces (positive DMI in Pt/Co and negative DMI in Fe/Ir) superpose to enhance the net DMI of the material.

FIGURE 3.6: Room temperature Néel skyrmions in FM-HM multilayer films. (a) 60 nm skyrmions in Ir/Co/Pt multilayers at 22 mT [46]. (b) 130 nm skyrmions in Pt/Co/MgO at zero magnetic field [49]. (c) Skyrmion clusters of average size 60 nm in Pt/Co/Ta at 78 mT [48]. (d) 250 nm skyrmions in a symmetric Pd/Co/Pd stack at zero field [43]. (e) Current pulse induced sub-50 nm skyrmions in Pt/Co/Ta nanodisks at 6 mT [22]. (f) Skyrmions in Pt/Co/W with an average size of 145 nm (at 100 mT) [47], respectively.

An important caveat arises from the vector nature of the DMI. For a successful implementation of additive DMI, it is imperative that each isolated interface, i.e. FM(1)-HM(1) and FM(2)-HM(2), has an opposite orientation of $\mathbf{D}$. In this way, an HM(1)-FM(1)-FM(2)-HM(2) composition will result in both $\mathbf{D}$ vectors pointing in the same direction, leading to a larger net DMI. For interfaces with identical DMI orientation, mutual cancellation of the DMI at the two interfaces will reduce the net DMI.

It is crucial to note that the interfacial DMI is highly sensitive to the quality of the interface. For example, in Pt/Co/Pt structures, identical DMIs at each interface should result in a perfect cancellation of the DMI. This has been observed only for epitaxial films with atomically sharp interfaces. In sputtered Pt/Co/Pt films, non-identical top and bottom interfaces (due to intrinsic limitations of the sputtering growth process) result in a finite DMI.

Though the FM-HM-FM architecture provides a wide range of parameters to control various magnetic phases, a finite magnetic field is often required to achieve stable skyrmion states. Moreover, skyrmions are stable only in a narrow region of phase space below the room temperature. One example is Pd/Fe/Ir, which hosts skyrmions at 11 K in a large OP field (2T). Such constraints limit the applicability of skyrmions for all practical purposes. The requirement of a large external magnetic field can be circumvented by using magnetic multilayers in which magnetic layers are separated by a nonmagnetic spacer. In such compositions, the ‘extra’ magnetic field required for stable skyrmions is provided by interlayer exchange coupling [50].

Ferromagnetic films separated by a nonmagnetic spacer can interact via exchange (ferromagnetic or antiferromagnetic) and dipolar interactions. Such interactions are broadly classified into two categories resulting from their distinct origins: direct and indirect coupling. The direct coupling arises due to pinholes in the spacer layer and can be avoided in smooth or thick films. Indirect coupling is further classified into several subcategories based on its origin and lengthscale.

One such example is the indirect exchange coupling, where the sign (ferromagnetic/antiferromagnetic) and strength of coupling oscillates with the thickness of the nonmagnetic spacer. The strength of coupling can be modulated with the thickness of the ferromagnet [51] and spacer layer [52]. Indirect exchange coupling has been investigated in detail for FM-HM-FM compositions [52]. The coupling strength: (a) increases with the number of d electrons across a row in the periodic table, and (b) increases exponentially from $5d$ to $4d$ to $3d$, but the oscillation period of the coupling remains similar for all transition metals. Magnetic Tunnel Junction (MTJ) devices display an excellent example of exchange coupling, where the magnetization of the hard layer is locked by antiferromagnetically coupling to a thick magnetic underlayer.

Another example of indirect coupling is provided by magnetostatic interactions (sometimes referred to as dipolar interactions) mediated via stray fields of spin textures. These are long-range interactions as stray fields can extend up to 70-80 nm. The lengthscale associated with stray fields can be estimated by MFM imaging, which converts stray field amplitudes into magnetization maps. Since magnetostatic fields are determined by the net magnetization of the layer, this coupling varies with the evolution of any magnetic features. The magnetostatic coupling has been utilised to replicate magnetic textures in multilayers [53]. In ferromagnetic layers hosting skyrmions, a finite dipolar coupling results in an evolution of the skyrmion structure across the thickness of the film [54].

In addition to interfacial engineering and layer stacking, geometric confinements can also be utilised to stabilise and fine tune skyrmions. It has been reported that the sizes and periodicity of skyrmions can be tuned with the relative size of the nano-disks in which they are nucleated [23]. Also, skyrmion velocities of 100 m/s have been achieved in racetrack-like nanostructures. This velocity is comparable to those of domain walls and promises a similar functionality for skyrmions.

3.4 Skyrmions in Ir/Fe/Co/Pt Multilayers

The thin film multilayers studied in this thesis are of general composition $[\text{Ir}(1)/\text{Fe}(x)/\text{Co}(y)/\text{Pt}(1)]_n$ (where the layer thickness is displayed in nm). x and y indicate variable thicknesses, and n denotes the number of repetitions of the Ir(1)/Fe(x)/Co(y)/Pt(1) stack. Films were deposited on thermally oxidised 500 μm Si wafers at room temperature using a RF magnetron sputtering system (Chiron UHV from Bestec GmbH). Two seed layers, Ta(3 nm)/Pt(10 nm), were deposited to optimise the crystalline structure of the upper layers. A Pt (2 nm) cap layer was finally deposited to prevent oxidation of stack. The deposition conditions for each layer were adjusted to control the thickness and roughness of the interfaces. The active layers in the stack create two FM-HM interfaces with oppositely oriented DM vectors, Co/Pt ($d_{\text{Co-Pt}} \approx +3$ meV) and Fe/Ir ($d_{\text{Fe-Ir}} \approx -2$ meV). Reversing the stacking order of Co/Pt in Ir/Fe/Co/Pt aligns both DM vectors in the same direction, resulting in a large net DMI. The selection of this material composition was motivated by density functional theory (DFT) calculations, which indicated a significant variation in the magnetic interactions with the thickness of the Fe/Co

layers. Details of the calculation procedure and results are mentioned elsewhere [42].

The predictions of these DFT calculations have been verified in experiments. Our observations are summarised in figure 3.7, which displays a variation in M_s , K_{eff} , A and D with the relative thickness of the Fe/Co layers. The estimated values of A and D were determined by comparing domain wall widths in experimental MFM images with those of simulated images. In general, M_s and D vary inversely, while A and K_{eff} increase proportionally to the relative thickness of Fe/Co. We notice that the magnetic parameters also depend on the absolute thickness of the Fe and Co layers. For 4/4, 5/5 and 6/6 films, although the relative thickness is identical in all cases, the magnetic properties vary significantly. While A remains constant for all three compositions, K_{eff} and D decrease monotonically in thicker films.

FIGURE 3.7: Tuning magnetic parameters via the thickness of Fe/Co layers in [Ir/Fe/Co/Pt]₂₀ multilayer films. (a) Experimentally measured M_s and K_{eff} and (b) estimated values of A_{est} and D_{est} for various Fe/Co compositions.

Skyrmion stability is determined by the relative magnitudes of the exchange, uniaxial anisotropy, and DM interactions. While the exchange and uniaxial anisotropy support a collinear spin arrangement, DM interactions stabilise canted spin textures [55]. Mathematically, the thermodynamic stability of the skyrmion phase can be expressed via a parameter κ

$$\kappa = \frac{\pi|D|}{4\sqrt{AK}} \quad (3.1)$$

where D , A and K are the DMI coefficient, exchange coefficient and effective magnetic anisotropy respectively. Néel skyrmions in thin films also generate a stray field. This extra energy term is incorporated in κ by adding an effective demagnetization factor to the uniaxial anisotropy K . A thermodynamically stable

skyrmion configuration exists only for $\kappa > 1$. For $0 < \kappa < 1$, only metastable, isolated skyrmions can form. The skyrmion configuration hence evolves from an isolated, disordered phase to a densely packed lattice with increasing κ in Ir/Fe/Co/Pt films.

In this thesis, we investigated thin films with 20 repeats of Ir(1)/Fe(0.5)/Co(0.5)/Pt(1) and Ir(1)/Fe(0.4)/Co(0.6)/Pt(1) stacks. The field evolution of the magnetization was tracked by magnetometry and finite field magnetic force microscopy (MFM). A Quantum DesignTM Magnetic Property Measurement System (MPMS) was used to determine the total film magnetization from 300 K to 100 K. Figure 3.8 and 3.9 display the field dependent magnetization of our films at 300 K. Both Fe(0.5)Co(0.5) and Fe(0.4)Co(0.6) follow qualitatively similar trends. The zero-field ground state exhibits an equal spatial density of oppositely-oriented labyrinth stripes, separated by Néel domain walls. An increase in the magnetic field results in expansion of stripes with positive OP magnetization (along the field direction), while the oppositely-magnetized stripes split into smaller worm-like features. A further increase in the field leads to skyrmion nucleation by splitting of short stripe segments, with stripes and skyrmions coexisting in this intermediate field range. As the field rises, more and more skyrmions are formed, eventually converting the mixed skyrmion/stripe phase into a densely packed skyrmion array. Skyrmions persist over a wide field range in both samples. A further increase in the applied field causes a reduction in the skyrmion size, accompanied by slight rearrangements in the skyrmion positions. The decrease in size continues at higher fields, resulting in the eventual annihilation of the majority of skyrmions. Only a few small skyrmions survive close to the ferromagnetic saturation limit of the film (possibly due to pinning by defects), distributed randomly over a field-polarised background. The skyrmion phase ceases to exist beyond the saturation field and the magnetization enters a polarised state. The magnetization measurements were performed down to 100K: both the saturation magnetization and magnetic anisotropy increase at lower temperatures, which is consistent with reported trends.

Magnetic Force Microscopy (MFM)

The preliminary confirmation of the skyrmion phase was obtained in room temperature MFM imaging. MFM experiments were performed with 25 nm radius, low magnetic moment tips (model-MESP-LM from BrukerTM) in a BrukerTM 3100D MFM. The tip-sample lift height (in addition to other parameters) was optimised for individual scans and was generally restricted to 40-60 nm. Diced

FIGURE 3.8: Field evolution of the magnetization in Fe(0.5)Co(0.5) (a) and Fe(0.4)Co(0.6) (b) measured at 100K, 200K and 300K. (c) Temperature dependence of the effective anisotropy K_{eff} for both compositions. The saturated magnetization and effective anisotropy both increase at lower temperatures.

FIGURE 3.9: Room temperature MFM images recorded at finite out-of-plane (OP) fields, displaying the field evolution of the magnetization in Fe(0.5)Co(0.5) at 45mT, 100mT and 200 mT. The magnetization evolves from long labyrinthine stripes to worm-like domains at 100 mT. Nucleation of skyrmions from worm-like domains begins around 200 mT. Scale bar = 500 nm.

samples of size $5 \times 5 \text{ mm}^2$ were used to acquire all images. The MFM images were recorded at zero and finite DC fields. Combinations of neodymium disk magnets with variable thicknesses and diameters were used to generate out-of-plane DC magnetic fields. The magnetic field of each neodymium disk was measured using a handheld gaussmeter. An MFM image is formed due to interactions between the MFM tip magnetization and the stray field above the sample surface created by magnetic textures. In multilayers, a finite dipolar interaction between the various magnetic layers induces an evolution of the magnetic textures across the thickness of the film. This layer-resolved magnetization cannot be detected by MFM and requires depth-sensitive techniques.

MFM imaging alone cannot provide conclusive confirmation of the skyrmion

phase, as it cannot distinguish between similarly-sized magnetic bubbles and large skyrmions due to its limited spatial resolution ($\approx 10 - 15$ nm). This low resolution prevents us from accurately determining the internal magnetization of the observed circular features. The magnetization of a skyrmion is marked by a continuous, gradual reorientation of spins from the skyrmion centre to its perimeter. This is distinct from a bubble domain, where a uniformly polarised core of finite size is surrounded by a thin domain wall. A finite DMI can stabilise both structures and as a result the terms are often used interchangeably [56]. Despite having a similar axisymmetric magnetization distribution, skyrmions and bubbles exhibit distinct static and dynamic properties. For example, the velocities (both longitudinal and transverse to the applied current direction) of skyrmions increase steeply with their size. In contrast, the longitudinal velocity decreases with size for a bubble, while its transverse velocity shows a weaker size dependence [57]. In our films, the circular objects were conclusively identified as skyrmions by magnetic transmission X-ray microscopy (MTXM), which shows distinct contrasts for skyrmions and bubbles. Once confirmed as skyrmions, the size of the circular features was extracted by fitting the magnetization profile of a radial linecut to a Gaussian function.

Néel skyrmions in sputtered films are considerably different from the Bloch skyrmions observed in non-centrosymmetric magnets and bulk alloys. For example, while Bloch skyrmion lattices in non-centrosymmetric magnets consist of identically sized skyrmions, the size of Néel skyrmions may vary by up to 100% in different regions of a film. In Fe(0.5)/Co(0.5) films the mean radius of the skyrmions is 56 nm, with maximum and minimum radii of 75 and 35 nm (figure 3.11a). Such a large variation in skyrmion sizes originates from inhomogeneity of the magnetic parameters due to the granular nature of the films. Local variations in the DMI and K_{eff} introduce size and shape distortions in skyrmions, and often act as pinning centres. This is noticeable in figure 3.10, where two skyrmions not only differ in size, but also have different core magnetizations. The typical skyrmion density in Fe(0.5)Co(0.5) is $10 - 20 \mu\text{m}^{-2}$, which limits the mean skyrmion separation to 200 nm; the average skyrmion radius is 56 nm in comparison. Despite the relatively high number densities, the skyrmions are not arranged in an orderly manner. This suggests a strong influence of material disorder on the skyrmion lattice, which overcomes skyrmion-skyrmion interactions and prevents ordering. The presence of skyrmions in the Ir/Fe/Co/Pt multilayers was also confirmed by topological Hall experiments [58], which quantify the emergent electromagnetic field of skyrmions from the amplitude of the topological Hall signal.

FIGURE 3.10: Spatially resolved magnetization of skyrmions in a Fe(0.5)/Co(0.5) multilayer at 5 K and 300 mT [59]. (a) MFM image containing two different sized skyrmions, a typical observation in sputtered films where inhomogeneity in the magnetic parameters leads to variations in the shape and size of skyrmions. (b) Radial linecuts through the skyrmion centre for both skyrmions. The gradual variation of the magnetization confirms the skyrmionic nature of the circular features.

3.4.1 Effect of Dipolar Interactions on Skyrmion Structure in Multilayers

In multilayer heterostructures, the magnetic textures in various layers can interact via exchange and magnetostatic fields. In the case of exchange interactions, the magnetic structure is expected to be identical across all the layers [53], enhancing the thermal stability. This form of coupling has been utilised to stabilise skyrmions at room temperature [46].

The interfacial DMI at a FM-HM interface can generate Néel skyrmions within the magnetic layers of a multilayer heterostructure. Néel rotation of spins induces a surface magnetic charge, resulting in a finite magnetostatic field. For relatively thin spacer layers (between successive magnetic layers), the magnetic textures within the various layers can interact via the magnetostatic fields. In this case, the chirality and relative orientation of neighbouring magnetic moments exhibit a thickness dependence, an effect unique to multilayers hosting Néel domain walls or Néel skyrmions. As previously reported [60], magnetostatic interactions in multilayer structures can stabilise ordered and isolated Bloch skyrmions over a wide temperature and field range, even in the absence of any DMI. Unlike DMI-induced

FIGURE 3.11: (a) MFM image recorded at 200 mT and 300K, showing a variation in the size and shape of skyrmions in a Fe(0.5)Co(0.5) film. The size variation is quantified in (b), revealing a Gaussian distribution (blue line) of skyrmion sizes centred around 50 nm. (c) Magnetization map of Fe(0.4)Co(0.6) at 160 mT and 300K (d) Analysis similar to (b) shows a Gaussian distribution in skyrmion sizes with mean size ≈ 56 nm, slightly larger than that of the Fe(0.5)Co(0.5) film. The scale bar in the MFM images is 500 nm.

skyrmions, whose chirality is restricted by the DMI vector, Bloch skyrmions stabilised by magnetostatic interactions exhibit equilibrium populations of skyrmions with both clockwise and counterclockwise rotations of in-plane moments.

In multilayers, the interplay between the DMI and dipolar coupling can create non-identical complex structures in different layers [54], where the skyrmion chirality oscillates between Néel and Bloch type across the layer thickness. Furthermore, the relative size of the skyrmions evolves across the thickness. A finite magnetostatic coupling between the various magnetic layers facilitates flux closure across the thickness, resulting in opposing polarities of the in-plane dipolar field along the top and bottom layers of material. The precise magnetic texture of a layer is determined by the relative strengths of the dipolar and DMI fields. Typically, the middle layers host Bloch skyrmions while the outer layers host Néel skyrmions

FIGURE 3.12: Micromagnetic simulations illustrating the layer-dependent magnetization profile of a Néel skyrmion in $[\text{Ir}(1)/\text{Fe}(0.5)/\text{Co}(0.5)/\text{Pt}(1)]_{20}$. (a), (b) and (c) display the spin texture in the top, middle and bottom layers of the film. (d) Magnetization cross-sections of the skyrmions in all 20 layers. The skyrmion size evolves in a barrel-like shape across the film thickness, due to magnetostatic interactions between the various magnetic layers. Scale bar = 20 nm.

of opposite chirality [54]. The chirality reversal of the Néel skyrmions is a direct consequence of flux closure.

For Ir/Fe/Co/Pt multilayers, while a large DMI ensures identical Néel rotation across all layers, the domain wall width decreases monotonically with thickness (figure 3.12). These results are in agreement with the theoretical model proposed in [61], which predicts a sharp decrease in the wall width from bottom to top for ultra thin films. The proposed origin of this trend is the variation of anisotropy with thickness which manifests itself as a change in the domain wall width $\Delta = \sqrt{A/K}$.

Contrary to the general understanding of skyrmions forming in a tube-like structure spanning all the layers (an expected outcome of exchange coupling between magnetic layers), the skyrmion sizes evolve with the thickness. Skyrmions are smallest at the top and bottom layers of the stack, but the skyrmion core increases towards the middle, almost appearing similar to a bubble with a finite size core of uniform magnetization. The strength of the magnetostatic interactions can be tuned with

the relative thickness of the non-magnetic spacers as well as by the number of magnetic layers in the material, providing a novel method to engineer the chirality and size of skyrmions.

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Chapter 4

Microwave Dynamics of Néel Skyrmions in Ir/Fe/Co/Pt Multilayers

This chapter describes the magnetization dynamics of the Néel skyrmion- hosting Ir/Fe/Co/Pt multilayers. Broadband FMR experiments reveal four distinct dispersion curves corresponding to the stripes, skyrmions and saturation magnetization of the material. While the stripe and field polarised phases exhibit resonance spectra consistent with expectations from the literature, two unique dispersion modes are observed for the skyrmion phase. Micromagnetic simulations and analytical calculations are employed to understand the physical mechanisms responsible for these resonance modes.

4.1 Magnetization Dynamics of the Ferromagnetic Phase: Uniform Precession and Damping

We begin by discussing the familiar uniform precession mode in the ferromagnetic (FM) phase. Field sweep FMR experiments were performed to determine the gyromagnetic ratio (γ), effective magnetic anisotropy ($\mu_0 H_{eff}$) and effective Gilbert damping (α_{eff}) of [Ir/Fe/Co/Pt]₂₀ films. Figure 4.1 displays the resonance spectra

of Ir(1)/Fe(0.5)/Co(0.5)/Pt(1)₂₀ (referred to as Fe(0.5)Co(0.5) hereafter) at 300K, measured over a frequency range of 5-20 GHz. Figure 4.1a shows the field sweep linecuts recorded at constant frequency. The resonance spectrum for each frequency was fitted with a combination of symmetric and anti-symmetric Lorentzian functions superimposed on a linear background. Details of the fitting method are provided in the appendix. The resonance field H_r and linewidth ΔH were extracted from the fitting parameters. The frequency vs. resonance field data from our fits are presented in figure 4.1b. An agreement between the experimental data and the Kittel model ($2\pi f = g \frac{\mu_B}{h} (H - M_{eff})$) establishes uniform magnetization precession, as the physical mechanism responsible for our high field resonance spectra. The effective magnetic field $\mu_0 H_{eff}$ is determined from the intercept of the Kittel fit with the H axis. A positive $\mu_0 H_{eff}$ reconfirms the perpendicular magnetic anisotropy of the films. The correlation between f and ΔH is presented in figure 4.1c: a linear increase in the linewidth with frequency can be interpreted as a Gilbert-like damping contribution [1–3]. The damping parameter is extracted from a linear fit [4]

$$\Delta H = \Delta H_0 + \frac{4\pi\alpha_{eff}}{\mu_0\gamma} f \quad (4.1)$$

where ΔH_0 is the zero-frequency linewidth. α_{eff} is a combination of the various contributions to the total damping, including the expected Gilbert and intrinsic dampings in magnetic metals.

FIGURE 4.1: Field sweep resonance spectra in the ferromagnetic state beyond 350 mT in a Fe(0.5)Co(0.5) film at 300 K. (a) Representative linecuts at 1 GHz intervals, fitted with a combination of symmetric and anti-symmetric Lorentzian functions. (b) Microwave frequencies and field values corresponding to absorption maxima fitted to the Kittel model. g and $\mu_0 H_{eff}$ are extracted from the fit. (c) Linear increment in linewidth with microwave frequency, fitted with equation 4.1. α_{eff} and ΔH_0 are extracted from the fit.

The observed effective damping ≈ 0.09 for Ir/Fe/Co/Pt multilayers is significantly higher than that reported for bulk Fe and Co films [5]. Such a large α_{eff} is expected from multiple FM-HM interfaces, which can open new channels for magnetization relaxation such as spin pumping into the HM layer. Details of spin relaxation mechanisms in multilayers are provided in chapter 2.

The large zero frequency linewidth can be related to the homogeneity of our thin films. The films were grown by magnetron sputtering, a technique known for its lack of uniform thickness and sharp atomic interfaces, especially at sub-nanometer thickness. Rough interfaces and defects in the film can scatter the uniform mode into lower energy magnon modes, thus increasing the linewidth. Another consequence of non-uniform thickness is a non-homogeneous energy landscape in the film. Non-uniform thickness is expected to produce local variations in the anisotropy, DMI, and exchange interaction, which are all highly sensitive to film thickness [6–8]. Such non-uniformity in the magnetic parameters is also evident in the MFM images (chapter 3), in which the shape and size of the skyrmions vary significantly in different regions of the film surface. The non-uniformity of the parameters - particularly the anisotropy, which appears in the Kittel model - will result in small resonance shifts for neighbouring magnetic grains. The experimentally detected spectrum is a superposition of shifted spectra from different areas of the film, leading to a ‘mosaic broadening’ [9, 10] of the linewidth.

Similar field sweep experiments were performed on a $[\text{Ir}(1)/\text{Fe}(0.4)/\text{Co}(0.6)/\text{Pt}(1)]_{20}$ (referred to as Fe(0.4)Co(0.6)) thin film (figure 4.2). The slight variation in composition leads to obvious differences in the resonance spectra of the film. This observation is consistent with our magnetometry experiments, where a slight variation in composition induces a change in magnetic properties. At low temperatures, we observe a shift in resonance spectra to higher frequencies. The dispersion spectra for both films at low temperatures are shown in figure 4.3. Such frequency shifts are attributed to larger anisotropy fields at low temperatures, and are consistent with the literature. Moreover, the g -factor also increases at lower temperatures.

4.2 Dynamics of Topological Solitons

The magnetization dynamics of topological solitons differ from those of non-topological magnetic structures. For example, in the presence of a magnetic field

FIGURE 4.2: The high field resonance spectra of a Fe(0.4)Co(0.6) film at 300 K. (a) Field sweep line-cuts fitted with a combination of symmetric and anti-symmetric Lorentzian functions. (b) Frequency - resonance field dispersion data fitted with the Kittel model. (c) Variation of linewidth with microwave frequency, fitted with equation 4.1.

FIGURE 4.3: Low temperature resonance spectra of Fe(0.5)Co(0.5) in (a) and Fe(0.4)Co(0.6) in (b). The $f(H)$ curve shifts to higher frequencies at lower temperatures. (c) Increase in the g -factor at lower temperatures.

gradient, magnetic vortices (or bubbles) move perpendicular to the field direction, while non-topological spin structures move along the direction of the gradient. This is similar to the Hall effect experienced by electrons in a magnetic field. In the case of topological objects, the dynamics are linked to the finite topological charge density $q_T = 1/2\epsilon_{\mu\nu} \mathbf{m} \cdot (\partial_\nu \mathbf{m} \times \partial_\mu \mathbf{m})$, and the associated topological number $Q = \frac{1}{4\pi} \int q \, dx dy$.

For an axially symmetric magnetic skyrmion, the parametric form of the magnetization in spherical coordinates (r, θ, ϕ) is

$$\mathbf{m} = (\sin \Theta \cos \Phi, \sin \Theta \sin \Phi, \cos \Phi) \quad (4.2)$$

where $\Theta = \theta(r)$ and $\Phi = \phi + \pi/2$.

It can be demonstrated that topological and non-topological textures follow distinct conservation laws, even though their magnetizations exhibit similar symmetry. For example, one can define an axial symmetric, non-topological magnetic structure called skyrmionium [11]. Unlike skyrmions, whose magnetization is characterised by boundary conditions, $m(r)_0 = 0, m(r)_\infty = \pi$, the core and perimeter spins are aligned ($m(r)_0 = 0, m(r)_\infty = 0$) in skyrmionium (figure 4.4). To characterise

FIGURE 4.4: Magnetization profiles of skyrmions with opposite topological charge Q , compared with zero- Q skyrmionium [12].

the difference in the dynamics of topological and non-topological structures, we formulate a tensor σ for the theoretical treatment [13, 14] of the magnetization, which is defined as

$$\partial_\nu \sigma_{\mu\nu} = -\mathbf{f} \cdot \partial_\mu \mathbf{m} \quad (4.3)$$

where $\mathbf{f}$ appears in the Landau-Lifshitz equation

$$\frac{\partial \mathbf{m}}{\partial t} = -\mathbf{m} \times \mathbf{f} \quad (4.4)$$

as an effective field. In the framework of the Landau-Lifshitz equation, the time evolution of the magnetization can be written in terms of the time derivative of the topological charge density [13–15]:

$$\dot{q} = -\epsilon_{\mu\nu} \partial_\mu (\mathbf{f} \cdot \partial_\nu \mathbf{m}) = \epsilon_{\mu\nu} \partial_\mu \partial_\lambda \sigma_{\nu\lambda} \quad (4.5)$$

Equation 4.5 has two main consequences [13, 14]. Firstly, the equation conserves the skyrmion number Q , which can be interpreted as the stability of the skyrmion

structure. Secondly, the equation implies the conservation of low order moments. One such moment is

$$\mathbf{I}_\mu = \int x_\mu q \, dx \, dy \quad \mu = 1, 2, \quad (4.6)$$

which is a conserved quantity.

The physical meaning of the moment can be understood by performing a coordinate transformation: $x_\mu \rightarrow x_\mu + c_\mu$. The new moment is

$$\mathbf{I}_\mu \rightarrow \mathbf{I}_\mu + 4\pi Q c_\mu \quad (4.7)$$

An interesting consequence of the above equation is the distinct response of topological and non-topological structures. For non-topological particles with $Q = 0$, the moments remain unchanged under a coordinate transformation, consistent with Newtonian physics. In this case $\mathbf{I}_\mu$ represents the linear momentum of a classical particle. For $Q \neq 0$, the shift in moment is proportional to the topological number of the texture. This suggests even though non-topological particles such as skyrmionium exhibit an axially symmetric magnetization similar to skyrmions (or bubbles), the unique properties of skyrmions are exclusively due to their topological nature. Even for the $Q \neq 0$ case, the moment $\mathbf{I}_\mu$ can be treated as a ‘topological’ linear momentum, which is a conserved quantity in the absence of field gradients.

A more physically understandable form of $\mathbf{I}_\mu$ is

$$\mathbf{R}_\mu = \frac{\mathbf{I}_\mu}{4\pi Q} = \frac{1}{4\pi Q} \int x_\mu q \, dx \, dy \quad (4.8)$$

which represents the mean position of the centre of the soliton, under a $\mathbf{R} \rightarrow \mathbf{R} + \mathbf{C}$ coordinate transformation. An important consequence of the conservation law for $\mathbf{I}_\mu$ is that the guiding centre of the soliton remains stationary in the absence of time or space dependent fields. However, this does not restrict internal deviations of the skyrmion core around the guiding centre. In the presence of a magnetic field gradient, the soliton drifts in a direction perpendicular to the gradient.

4.2.1 Translational Motion of Skyrmions

In general, the motion of a topological soliton is described by the Thiele equation [16]:

$$\mathbf{G} \times \dot{\mathbf{R}} + D\dot{\mathbf{R}} - \partial_{\mathbf{R}}U = 0 \quad (4.9)$$

The first term in equation 4.9 is the gyroscopic force, where $\mathbf{G}$ is the gyroscopic vector. It is referred to as a ‘topological Magnus force’ and induces transverse motion of a skyrmion. It is known that the field-induced motion of magnetic bubbles is perpendicular to the applied field. Similar dynamic effects have been reported for skyrmions. During current-induced motion, isolated skyrmions move at an angle to the current direction [17, 18]. Such angular deviation from a straight path is referred to as the ‘skyrmion Hall effect’ and is a consequence of the topological character of skyrmions. An intuitive explanation of the ‘topological Magnus force’ is to consider it as a reaction to the Lorentz force experienced by electrons passing over the emergent magnetic field of a skyrmion.

The second term in the Thiele equation is the velocity-dependent viscous force, where D is the dissipation tensor. The final term $\partial_{\mathbf{R}}U$ is the net conservative force due to a potential U , which drives the steady state motion. The Thiele equation serves as a generalised model for the motion of circular features with gradual spin variation, and has been applied to superconducting vortices, magnetic bubbles, and skyrmions. The equation is valid only for steady state motion $R(t) = Vt$ and predicts vortex motion similar to that of a massless charged particle moving in a magnetic field and a viscous medium. The gyroscopic vector $\mathbf{G} = (0, 0, G)$ is proportional to the topological charge Q , and can be calculated from the topological charge via $G = -4\pi Q$ [17]. The finite skyrmion topological charge ($Q = \pm 1$) results in a non-zero G , which explains the skyrmion Hall effect. The precise form of G depends on the material characteristics and is written as $G = -4\pi Q t M / \gamma$, where t is the film thickness and M / γ is the density of angular momentum.

Although the Thiele equation successfully provides a qualitative explanation of the skyrmion Hall effect, it does not reproduce the correct trajectory of a skyrmion [19, 20]. The primary reason for this discrepancy is embedded in the general structure of the Thiele equation, which does not include a mass term, and hence is only applicable to massless particles. An updated formulation of the equation was proposed by Makhfudz et al. [21], which includes an inertial mass M combined with a second derivative of the position:

$$-M\ddot{\mathbf{R}} + \mathbf{G} \times \dot{\mathbf{R}} + D\dot{\mathbf{R}} - \partial_{\mathbf{R}}U = 0 \quad (4.10)$$

For $G \neq 0$, the dissipative force $D\dot{\mathbf{R}}$ is significantly less than the gyroscopic force and hence can be neglected. The inertia of skyrmions is attributed to deformations of their shape and size under external perturbations. Such distortions

are characterised as resonance modes of skyrmions: they consist of gyrations of the skyrmion core around a fixed centre and radial oscillations in the skyrmion size. The massless form of the Thiele equation is valid for so-called ‘hard’ skyrmions, where the structure of the skyrmion is a preserved quantity. Writing the conservative force as $\kappa \mathbf{R}$ (for the simplest case of a harmonic potential), the equation converts to

$$-M\ddot{\mathbf{R}} + \mathbf{G} \times \dot{\mathbf{R}} - \kappa \mathbf{R} = 0 \quad (4.11)$$

Unlike the first order form of the Thiele equation, the presence of a double-derivative term due to the finite mass of the skyrmion leads to two non-degenerate solutions to the equation of motion. The solution includes two circular modes moving with different frequencies in the GHz regime [21]:

$$\omega_{\pm} = -\frac{G}{2M} \pm \sqrt{\left(\frac{G}{2M}\right)^2 + \frac{\kappa}{M}} \quad (4.12)$$

The two frequencies are associated with the clockwise and counter-clockwise gyrations of a skyrmion. The precise trajectory of a skyrmion is determined by a superposition of the two gyration modes. The inertial term is associated with the deformations of the skyrmion [22] caused by synchronous motion of all the constituent spins, referred to as collective resonance modes of skyrmions. Furthermore, as established recently, the inertia is related to the size of the skyrmions [23]. This is particularly valuable for skyrmion dynamics, where the trajectories of a skyrmion can be controlled by optimising its size. Also, it was noted that the two gyration modes of skyrmions are non-degenerate and the skyrmion trajectory varies with the relative amplitudes of each mode [21]. Therefore, a deeper understanding of the collective resonances is an essential prerequisite for the utilisation of mobile skyrmions.

4.2.2 Oscillation Modes of Skyrmions

As noted in the previous section, the inclusion of a mass term is necessary for the accurate description of skyrmion trajectories under external perturbations. An important consequence of this skyrmion mass is the existence of non-degenerate, GHz frequency internal modes of skyrmions. An external disruption (magnetic field or current) can cause a deformation in the shape and/or size of skyrmions. The

response to such perturbations is governed by the internal modes of the skyrmions, which ultimately decide the trajectory of skyrmion motion [22].

Internal Modes of a Lattice of Bloch Skyrmions

A pioneering work [24] on Bloch skyrmion lattices postulated three internal modes corresponding to the collective gyration of skyrmion cores in counter-clockwise (CCW) and clockwise (CW) directions around the centre of mass of the topological charge density (q_T , referred to as the ‘guiding centre’ in subsequent sections), and synchronous radial oscillations of the skyrmion core, referred to as the breathing (BR) mode. Physically, the CW and CCW modes relate to the periodic motion of the ‘first moment of magnetization’ and the maxima in the topological charge density around the guiding centre of the skyrmion, which remains stationary in the absence of field gradients. For gyration modes, the sense of rotation is restricted by an external magnetic field (via the orientation of the skyrmion magnetization), which is also responsible for lifting the degeneracy of the CCW and CW oscillations. Two gyration modes are excited by the in-plane (IP) components of an microwave field, while the breathing mode is induced from an OP oscillating field. A remarkable feature of such modes in skyrmion lattices is the coherent oscillation of all skyrmions in an identical fashion, resulting in a collective resonant absorption.

Experimental Observation of Resonance Modes of Bloch Skyrmions

Experimentally, the internal modes of Bloch skyrmion lattices have been observed in a diverse set of materials, primarily using the broadband ferromagnetic resonance (FMR) technique. Recent work [25] has established the universal nature of skyrmion oscillations among three distinct materials (an insulator, metal, and semiconductor) belonging to the same space group P_213 . As described in chapter three, magnetic interactions follow a definite energy hierarchy for crystals in the same space group, leading to similar magnetic phase diagrams. Such hierarchy was also observed in the dispersion spectra of the various skyrmion resonances. The frequency (f) versus field (H) dispersion curves display distinct trends for various magnetization configurations. The ferromagnetic phase above the saturation field exhibits a positive slope $df/dH > 0$ (figure 4.5a), due to uniform precession of the magnetization. For the regime hosting skyrmions at low fields, three different modes are observed. These include two in-plane oscillations (CCW and CW) and one breathing oscillation (BR). The energy scales for the different modes follow the order $CCW < BR < CW$. These three skyrmion modes also show different frequency-field dependencies. The high energy CW mode shows a weak positive

slope, appearing almost flat. The CCW mode follows a positive slope, though less steep than the FM mode. The BR mode follows a negative slope. At even lower fields, a single dispersion mode which is weakly dependent on magnetic field was observed.

The helimagnet Cu_2OSeO_3 is a chiral insulator in the P_213 space group, which hosts a triangular skyrmion lattice at low temperatures. It is also a multiferroic and is known for exhibiting a spin texture-mediated ferroelectric polarization. Such characteristics establish Cu_2OSeO_3 as a perfect material to explore the interdependence of charge and spin effects, their coupling mechanism and potential applications. Among several interesting features of the magnetization dynamics in Cu_2OSeO_3 , a particularly remarkable one is the nonreciprocal directional dichroism (NDD) induced by resonant oscillations of skyrmions [26]. This implies that microwaves moving in opposite directions through the skyrmion crystal will experience different absorptions. The cause of such transmission properties lies in the relation between oscillations of the magnetization $\mathbf{M}$ and polarization $\mathbf{P}$ of material. An in-phase oscillation leads to higher absorption, while an out-of-phase oscillation leads to weaker absorption. This property could be utilised to develop skyrmion-based microwave diodes.

Internal Modes of Isolated Skyrmions

As observed in recent experiments on FeGe, the skyrmion configuration can be tuned from hexagonal lattice to disorder, by the action of temperature and magnetic field [27]. Isolated skyrmions in a uniformly polarised ferromagnetic background provide a less complex environment to study the internal modes of individual skyrmions, since the resonant properties of polarised ferromagnets are well understood.

Ferromagnetic order exhibits a continuum for magnon modes in the absence of skyrmions. Skyrmions introduce local potentials for magnons, which support localised internal modes of skyrmions with energies smaller than the magnon gap. The spatial extension of the potentials is limited by the skyrmion size (usually 10-100 nm) and hence the skyrmions do not affect the long wavelength magnon modes. For a single skyrmion, the lowest stable energy mode is the translational mode: uniform translation of skyrmions without any shape distortion. The translational mode is unique to isolated skyrmions and is not permitted in skyrmion lattices due to symmetry constraints. A slightly higher energy mode is due to CCW gyrations of skyrmion core: the breathing mode only emerges at the highest

energies. Translational motion and the associated gyration and breathing modes have been observed by X-ray transmission experiments [22]. The dispersion relation for these modes and the stability of the skyrmion phase are sensitive to uniaxial anisotropies, which are responsible for variations in the experimentally reported resonance spectra in various magnets [28].

4.3 Distinguishing Resonance Spectra in Néel and Bloch Skyrmions

The resonant oscillations of Néel skyrmion lattices have been observed in single crystals of the polar magnet GaV_4S_8 at low temperatures. Néel skyrmions exhibit CW, CCW and breathing modes at different frequency ranges, similar to Bloch skyrmions. However, a major difference in the resonance characteristics of Bloch and Néel skyrmions appears in the energy hierarchy of the various modes. While the Bloch skyrmion lattice exhibits $\text{CCW} < \text{BR} < \text{CW}$ energy scales (figure 4.5a), Néel skyrmion lattices follow a $\text{BR} < \text{CCW} < \text{CW}$ energy sequence (figure 4.5b). The renormalisation of the energy scales associated with resonance in Néel skyrmions is attributed to the uniaxial anisotropy present in such materials. Another distinction in the resonance of the two types of skyrmions is manifested in the field dependency of their modes. While the dispersion modes for Bloch skyrmions exhibit a strong field dependence, the dispersion modes of Néel skyrmions scale weakly with field. Moreover, the transitions between various modes are smoother in the case of Néel skyrmions. For Néel skyrmions in thin films, dispersion modes are predicted for isolated and ordered skyrmions phases. While the lattice displays all three modes, only breathing and CCW modes have been suggested for isolated skyrmions [29].

4.4 Resonance Modes of Néel Skyrmions in Ir/Fe/-Co/Pt Multilayers

In this thesis, we have investigated the resonance modes of various magnetic phases in $[\text{Ir}(1)/\text{Fe}(0.5)/\text{Co}(0.5)/\text{Pt}(1)]_{20}$ thin films (referred to as Fe(0.5)/Co(0.5) hereafter). The frequency and field sweep experiments are performed using the apparatus described in chapter two. The physical mechanisms responsible for

FIGURE 4.5: Comparison of microwave resonance spectra in Bloch (a) and Néel (b) skyrmions. Both skyrmions exhibit CW, CCW and BR modes, but the energy hierarchy and field dependencies are different in the two cases. (a) and (b) are reconstructed from the data points in [25] and [30] respectively. The dispersion spectra in (a) are plotted on a normalised scale.

the experimentally observed resonance spectra are explained with the help of micromagnetic simulations and analytical calculations.

The frequency sweep FMR technique is used to characterise the magnetization dynamics of magnetic phases below the saturation field of the film. A frequency sweep is performed at a constant field and hence does not alter the film magnetization during the measurement. This is a critical requirement in our case, as the nucleation of skyrmions depends strongly on the magnetic history of the film. The field evolution of the magnetization and the corresponding frequency sweep FMR spectra for Fe(0.5)Co(0.5) at room temperature are displayed in figure 4.6. The spectra were fitted with a Dysonian model superposed on a linear background [30]:

$$S_{12}(\nu) = \frac{I[\Delta\nu + \beta(\nu - \nu_0)]}{(\nu - \nu_0)^2 + \Delta\nu^2} + D\nu + B \quad (4.13)$$

I is a fitting parameter proportional to the amplitude of the resonance spectrum, ν_0 is the resonance frequency, $\Delta\nu$ is the frequency linewidth, and β is the dispersion to absorption ratio. Parameters D and B compensate the linear drift and baseline offset of the spectrum. The field evolution of the resonance spectra reveals four distinct modes corresponding to the various magnetic phases in the film (figure 4.7b). The frequency sweep spectra are obtained in progressively increasing OP magnetic fields (figure 4.7c). The resonance spectrum at each field is fitted with a Dysonian function. Figure 4.7a displays horizontal linecuts of the transmission intensity

FIGURE 4.6: Field evolution of magnetization and resonance spectra in a $[\text{Ir}(1)/\text{Fe}(0.5)/\text{Co}(0.5)/\text{Pt}(1)]_{20}$ film at 300 K. (a), (b) and (c) display finite field MFM images of magnetic stripes (100 mT), skyrmions (230 mT) and the ferromagnetic phase (300 mT). The scale bar in the MFM images measures 500 nm. Corresponding frequency sweep FMR spectra for stripes, skyrmions and the ferromagnetic phase are shown in (d), (e) and (f). Resonance spectra are fitted with the Dysonian model described in equation 2.13. The dotted lines in (d), (e) and (f) mark the resonance frequencies.

graph (figure 4.7b, equivalent to a field sweep FMR experiment). The corresponding resonances, obtained from figure 4.7a and b, are overlaid on the measured transmission intensity (S_{12}) of the microwave signal for all magnetic phases (figure 4.7b). The FM phase (above ~ 300 mT) is marked by a proportional increase in the resonance frequencies with the external field. This linear dependence is the expected response of a uniformly precessing saturated magnetization and can be fitted with the Kittel dispersion equation. In the intermediate field regime where a stable skyrmion state exists, two dispersion modes with distinct field dependencies are observed. The high frequency (HF) mode exhibits a weak correlation with the magnetic field and remains close to 9 GHz for all fields. The low frequency (LF) mode rises steeply with the magnetic field and extends from approximately 7 to 9 GHz in a narrow field range just below the saturation limit. At even lower fields,

skyrmions merge to form stripes and the corresponding dispersion spectra are characterised by an inverse proportionality of the resonance frequency to magnetic field.

FIGURE 4.7: Frequency sweep resonance spectra for a Fe(0.5)Co(0.5) film, measured at 300 K. Overlaid filled circles mark the resonance frequencies corresponding to various magnetic phases. Pink circles denote resonance frequencies for magnetic stripes, blue for the ferromagnetic phase, and cyan and green for the skyrmion phase. (a) Interpolated horizontal linecuts of the transmission intensity in (b). (b) Transmission intensity map acquired via high resolution frequency sweep measurements. (c) Experimentally recorded frequency sweep linecuts at various OP fields. Resonance frequencies for various modes, represented by filled circles, were extracted by fitting raw data in (a) and (c). Green and blue lines in (b) correspond to linecuts at 9.5 GHz (a) and 250 mT in (c).

The nucleation and number density of skyrmions are correlated with the magnetic history of the film [31]. To understand the role of magnetic field history on resonance spectra we also performed field sweep FMR experiments on Fe(0.5)/Co(0.5) film. Surprisingly, we again observe distinct dispersion curves in the stripe, skyrmion (HF and LF) and FM phases, identical to our results from frequency sweep spectra. Figure 4.8a displays experimental field sweep curves obtained for discrete microwave frequencies. The field sweep spectra at high frequencies are a superposition of two resonance signals corresponding to the stripe and ferromagnetic (FM) phases at

low and high field respectively. For lower frequencies the spectra merge into a single peak corresponding to LF resonance. The HF resonance was extracted by analysing the vertical linecuts, which are equivalent to frequency sweep spectra. High frequency spectra were fitted with a dual peak Lorentzian function, with symmetric and asymmetric contributions superposed on a linear background. Single peak spectra at low fields were fitted with a single peak Lorentzian function. Details of the fitting procedure are included in the appendix.

FIGURE 4.8: Field sweep resonance spectra for Fe(0.5)Co(0.5) at 300 K. (a) Experimentally measured field sweep spectra at various frequencies. An extended set of field sweeps (a) is plotted as a transmission intensity in (b). (c) Interpolated frequency sweep spectra obtained from vertical linecuts of (b). The overlaid points in (b) indicate the field at which absorption is maximal for each frequency, determined from the fits to the resonance spectra in (a) and (c). Green and blue lines in (b) correspond to resonance spectra at 9.6 GHz in (a) and 250 mT in (c) respectively.

In our field sweep measurements, the film is saturated above 500 mT in an out-of-plane field, followed by a continuous decrease to 0 mT for all microwave frequencies. The transmission parameter S_{12} was recorded at successive intervals of 2 mT during the field sweep. A fast field sweep leads to the formation of metastable magnetic phases, particularly near the stripe-skyrmion phase boundary. Similarly, near the

saturation limit, mode softening can lead to deviations from the Kittel model which can be wrongly interpreted as resonant absorption from skyrmions.

We notice an excellent agreement between field and frequency sweep methods, for all magnetic phases. This is particularly interesting for resonance spectra in the skyrmion phase, as the field and frequency sweep techniques are expected to create distinct magnetic histories, which may lead to different number densities and/or size distributions of skyrmions. Such an agreement shows the insensitivity of resonance spectra to the precise skyrmion configuration, which is a function of the magnetic history of the material. Since the skyrmion number density can be correlated with the inter-skyrmion interactions (a higher number density ensures a shorter inter-skyrmion separation, and hence stronger skyrmion-skyrmion interactions), it can be concluded that these resonances are negligibly dependent on the interactions between neighbouring skyrmions.

4.4.1 Origin of Resonance Spectra: Full-stack Micromagnetic Simulations

The physical origin of the resonance spectra was deduced via micromagnetic simulations performed using the MuMax3 package [32]. The film structure was modelled as 20 repetitions of a FM layer of thickness 1 nm (corresponding to Co/Fe), separated by 2 nm of a nonmagnetic spacer layer (Pt/Ir). The following parameters were used for simulations: A - 11 pJ/m², D -1.95 mJ/m², M_s -1.02 MA/m, K_{eff} -0.69 MJ/m³. These values are consistent with our experiments and have been reported previously [33]. The effective damping constant α_{eff} was determined from field sweep resonance spectra in the ferromagnetic state. All simulations were performed at 0 K, but finite temperature effects were incorporated by using the 300 K values of M_s and K_{eff} . Interaction between the magnetic layers was allowed in the form of long range magnetostatic interactions.

To simulate the magnetization dynamics, an isolated Néel skyrmion was relaxed at the centre of a 400x400 nm² grid, with a mesh size of 4x4x1 nm³. An IP microwave field was applied as $h(t) = h_0 \sin(2\pi f_c t)/2\pi f_c t$. The amplitude h_0 was set to 10 mT, which is consistent with our experiments. However, unlike our experiments where the microwave field decays moving away from the CPW, $h(t)$ is assigned to be uniform across the thickness of the material. The transient dynamics were

FIGURE 4.9: Simulated resonance spectra calculated using a full-stack model [FM(1)/Spacer(2)]₂₀ of a Fe(0.5)Co(0.5) film. The dashed green line highlights the weak LF mode and the overlaid data points are from frequency sweep experiments. Simulations are performed using $\alpha_{eff} = 0.086$.

simulated for 10 ns and the power spectral density was calculated from the Fourier transform of the variation in OP magnetization. This procedure was repeated for successively increasing magnetic fields up to 350 mT.

We compare the simulated total microwave absorption of the film with our experimental data. Figure 4.9 displays FMR spectra for a single skyrmion in a full-stack model, overlaid with experimental resonances from our frequency sweep spectra. We notice a significant difference in the HF and LF mode intensities, the HF mode being more prominent. The resonant frequencies in our experiments and simulation are in good agreement for the stripe, HF and FM modes. However, the simulated LF spectra show an offset of ≈ 50 mT from our experimental results.

The reasons for this disagreement are discussed in section 4.4.3. Furthermore, we will demonstrate that the skyrmions gyrate in counter-clockwise (CCW) fashion at both HF and LF resonances. Typically the emergence of two modes at the same field with different frequencies and highly asymmetric amplitudes is associated with excitation of both clockwise and counter-clockwise modes [30]. The occurrence of two modes with an identical sense of rotation is unique to multilayers.

It is noteworthy that despite the agreement in the resonance spectra, there is a significant difference between the skyrmion configurations in our experiments and simulations. The simulations were performed on an isolated skyrmion relaxed

at the centre of the film. In our experiments, the skyrmions are densely packed as shown in our MFM images and can therefore interact with their neighbours. Furthermore, unlike the single ideal skyrmion in our simulations, the skyrmion sizes and shapes vary significantly over our actual films, presumably due to inhomogeneity. Consequently, the experimental spectrum is a collective response of a large number of skyrmions with varying shapes and sizes. Details about the shape and size variation obtained from MFM imaging are included in chapter three.

The large effective damping ($\alpha_{eff} = 0.086$) in our films widens the linewidth at resonance, resulting in a broadening of the LF and HF modes. To explore the precise nature of both resonances, we performed full-stack simulations at lower α_{eff} values (0.04). A lower α deconvolves the resonance spectra corresponding to the LF and HF modes (figure 4.10). Notably, the modification in α does not alter the position of maximal absorption, and hence the agreement between experiments and simulations remains valid. Although the layer-averaged simulated spectra exhibit both LF and HF modes, not all layers oscillate at both frequencies. As we discovered in layer-resolved simulations (figure 4.10b and c), only the internal layers in our film contribute to the LF mode, while the HF mode is extended over the entire thickness of the film. We notice that the middle layers of the stack in which the LF mode appears host larger bubble-like skyrmions. It has been reported that the skyrmion oscillation frequency decreases inversely with its size: $f_r \approx D_{sk}^{-1}$ [23], suggesting that a stack-induced size variation could be responsible for the LF mode. However, as we explain in section 4.4.3, skyrmion size variation alone cannot explain LF resonance. Furthermore, we emphasize the fact that all layers experience an identical microwave field in our simulations, which eliminates nonuniformity in the RF field as a cause for the two modes.

The asymmetry in the resonance spectra of external and internal layers can be resolved by consideration of the dipolar field appearing due to the large thickness of the stack [34]. Due to the flux closure condition, the dipolar field is aligned parallel to the film surface at the outer layers, but points perpendicular to the film in its interior. The influence of the dipolar field is responsible for splitting the resonance spectra into HF and LF modes within the skyrmion phase.

FIGURE 4.10: Simulated FMR spectra of Fe(0.5)Co(0.5) at a lower $\alpha_{eff} = 0.04$. (a) Absorption intensity colour map showing the HF and LF modes. Black and white dashed lines indicate the low intensity LF mode and high intensity HF mode respectively. (b) Layer-resolved magnetization of all ferromagnetic layers in the film, showing the variation in skyrmion size and skyrmion wall width across the thickness. (c) Layer-resolved resonance spectra from the 20th, 10th and 1st layers at 290 mT. Dotted cyan and green lines indicate the positions of resonance frequencies. While the HF mode exists in all layers, the LF mode is observable only in the middle layers of the film.

4.4.2 Distinction Between LF and HF Modes: Analysis of the Time-Resolved Magnetization

Although our micromagnetic simulations reveal the CCW gyrations of the skyrmion cores for both HF and LF resonances, several important questions remain unanswered. If the skyrmion core is oscillating in a CCW direction for both modes, why the frequency range so different? Also, why is the intensity of the HF mode significantly higher than the LF mode? To resolve these questions, we analysed the time-resolved OP magnetization from our micromagnetic simulations and extracted the spatial evolution of both modes around the skyrmion centre. Before proceeding

into the details of the time-resolved analysis, we present a brief introduction of the theoretical model employed in this study.

As defined in equation 4.2 of section 4.1, the axially symmetric solutions of a Néel skyrmion in cylindrical coordinates can be written as $\Theta = \theta(r)$ (representing the OP magnetization) and $\Phi = \phi + \pi/2$ (controlling the IP magnetization). The time-dependent variation (from the equilibrium skyrmion magnetization) of the polar and azimuthal projections of the magnetization during skyrmion oscillation is characterised by a pair of quantum numbers, referred to as the radial and azimuthal quantum numbers, which account for the number of modes along the radial and azimuthal directions. Furthermore, the sign of the azimuthal quantum number dictates the direction of azimuthal rotation. Conventionally, positive (negative) corresponds to clockwise (counter-clockwise) gyration [23, 35–37].

Analytically, the excitation of a skyrmion with small deviations from its equilibrium configuration can be modelled by adding small perturbations to the stationary state solutions [23]: $\Theta = \theta(r) + \nu$, $\Phi = \phi + \frac{\psi}{\sin\theta}$, where ν and ψ are the variations in OP and IP magnetization. The precise form of ν and ψ determines the spatial and temporal evolution of the excitation. The evolution of the excited state can be calculated by solving the Landau-Lifshitz equation for ν and ψ . We obtain

$$\nu(r, \theta, \tau) = f(r) \cos(\omega\tau + \mu\theta + \eta), \quad \phi(r, \theta, \tau) = g(r) \sin(\omega\tau + \mu\theta + \eta) \quad (4.14)$$

where $f(r)$ and $g(r)$ are proportional to the variations in the OP and IP magnetizations during the excitation, ω is the excitation frequency and μ is the azimuthal quantum number. $\mu \in \mathbb{Z}$ denotes the number of nodes (2μ) around the skyrmion centre, and η is an arbitrary phase. Typically, three localised modes are defined for skyrmions: $\mu = 0$ (corresponding to breathing mode oscillations) and $\mu = \pm 1$ (gyrotropic modes). The mode $\mu = -1$ is referred to as the counter-clockwise mode. The exact form of f and g depends on the gyration modes of the skyrmion (see figure 3 in [23], which portrays f and g for various types of skyrmion oscillation).

As we discovered in our full-stack micromagnetic simulations, the middle layers of the film exhibit both LF and HF resonances. Therefore, we have selected layer 10 for our time-resolved analysis. The simulation data for this analysis were recorded for a lattice geometry of $400 \times 400 \text{ nm}^2$, relaxed at a finite field of 280 mT. The ground state configuration includes a skyrmion of diameter 70 nm, located at the centre of the film and surrounded by a ferromagnetic background. In our case, f

and g were extracted from the Fourier transform of the time-resolved magnetization recorded over a 10 ns period. The spatial profiles of f and g were extracted from radial linecuts outward from the skyrmion centre.

In the absence of a skyrmion, we expect the f and g functions to overlap, characterising the uniform precession of the FM background. Moreover, since the uniform mode is delocalised over the entire film surface, the amplitudes of f and g will remain constant throughout the film. In contrast, for skyrmions the variation in magnetization is localised within the skyrmion area and hence f and g will be confined within the skyrmion. For a skyrmion set in a FM background, the spatial evolution of the f and g functions indicates a response superposing three distinct contributions. First, the oscillation of the skyrmion, which is obviously localised within the skyrmion. Second, the uniform precession of the ferromagnetic background, delocalized over the film plane. The third contribution originates from the scattering of the uniform magnon mode by the emergent magnetic flux of the skyrmions which has been theoretically investigated previously [38, 39].

In our analysis, we noticed that during LF resonance, the maximum of the f and g functions resides within the skyrmion and decays rapidly beyond the skyrmion boundary (figure 4.11). This indicates a relatively larger contribution of the internal oscillations of skyrmions relative to the precession of the FM background. In contrast, the f and g functions for the HF mode display a finite amplitude even at distances several times the skyrmion radius, indicating spatial delocalisation in the resonance. This suggests a relatively larger contribution (in comparison to the LF mode) from uniform precession of the ferromagnetic background. The area of the ferromagnetic background is equal to the total film area ($400 \text{ nm} \times 400 \text{ nm} = 160000 \text{ nm}^2$) minus the area of skyrmion ($\pi \times 35 \text{ nm} \times 35 \text{ nm} = 3846.50 \text{ nm}^2$), covering 97 % of the film. Since most of the simulated film exhibits uniform precession, this explains the larger intensity of the HF mode in comparison to the LF mode. The frequency of the HF mode is higher than the Kittel dispersion extrapolated to low fields. This is a non-trivial result as one would expect the uniform background precession to follow the Kittel dispersion law. Such a shift in the HF mode frequency is attributed to the scattering of uniform precession magnons from oscillating skyrmions. This has been further confirmed in multi-skyrmion simulations, where the frequency shifts to higher values with an increase in the number of skyrmions.

FIGURE 4.11: Spatial variation of $f(r)$ and $g(r)$ for the LF and HF modes. (a) and (b) display the $f(r)$ and $g(r)$ functions extracted from our simulations at LF resonance. The overlaid dashed lines represent $f(r)$ and $g(r)$ for CCW gyration of an ideal Néel skyrmion. (c) and (d) show $f(r)$ and $g(r)$ derived from our simulations at HF resonance.

We have explained the physical mechanisms responsible for the origin of two distinct resonance modes for skyrmions in individual FM layers. However, one question remains unanswered. Is there a correlation between the skyrmions oscillating in different magnetic layers of the film? This could be resolved by determining the evolution of phase factor η across various layers.

However, such phase difference cannot be resolved experimentally by our FMR technique, which measures the microwave absorption simultaneously from all magnetic layers. Layer-resolved magnetization dynamics have been probed in X-ray experiments on spin-valve structures [40]. It was observed that even in a simple FM/NM/FM stack, the amplitude and phase of the spin precession differ significantly between two FM layers. Following this, we expect similar incoherent oscillations of the skyrmions within our films. Furthermore, as shown in figure 4.10b the shape and size of the skyrmions varies significantly across the film thickness and over the film surface (see chapter three MFM images). This can result in skyrmions oscillating at slightly different frequencies, as their resonant frequency scales inversely with their size. In a FMR experiment, this appears as a wide

resonant minimum in the microwave transmission formed due to closely-spaced resonances of similarly sized skyrmions.

4.4.3 Periodic Boundary Condition (PBC) Simulations for Skyrmion Resonance

The existence of LF and HF modes at the interior and outer edges of the stack can be misinterpreted as a byproduct of the variation in skyrmion size. To eliminate this possibility, we performed micromagnetic simulations on a FM/NM bilayer with periodic boundary conditions (PBCs) along the z direction (figure 4.12a). Note that all simulations use PBCs in the x, y plane to capture the field-dependent skyrmion density. The magnetic parameters used are identical to those in our full-stack simulations. Additionally, dipolar interactions between various magnetic layers were allowed. The primary distinction between full-stack and PBC simulations is the evolution of magnetization across the thickness of the multilayer stack. While a full-stack model allows a barrel-like variation in skyrmion size, only one skyrmion of constant size (at a given OP field) exists in PBC simulations.

To understand the influence of dipolar interactions on the resonance spectra, we first investigate the correlation between skyrmion size and dipolar fields. Figure 4.12b displays the variation in the skyrmion diameter D_{sk} for successively thicker spacer layers. Typically, for a given spacer, the diameter is inversely proportional to the out-of-plane (OP) field. This is an expected outcome, as larger skyrmions cost more Zeeman energy. The spacer layer modulates the dipolar interaction between the magnetic layers in the stack. Consequently, changing the thickness of the spacer layer will modulate the strength of the dipolar interaction. We observe an inverse proportionality between the spacer thickness and skyrmion size. Furthermore, the field evolution of the skyrmion size exhibits distinct trends for different spacer layers. At the extreme ends of spacer thicknesses, i.e., 0 nm (no spacer) or 4 nm spacers, the trend of size reduction varies from strictly linear (0 nm) to approximately exponential (4 nm).

Next, we simulate the effect of varying the skyrmion size on the resonance spectra. Figure 4.13a shows the evolution of the LF mode resonant frequency f_{LF} with D_{sk} for various spacer thicknesses. In general, f_{LF} scales inversely with D_{sk} . Importantly, f_{LF} for different spacers display distinct dependences on the skyrmion

FIGURE 4.12: (a) Material geometry for PBC simulations in which the FM layer thickness is restricted to 1 nm (consistent with experiments) and the spacer thickness t is the tuning parameter. (b) Magnetic field dependence of the skyrmion size for various spacer thicknesses.

size. If the skyrmion size were the only controlling parameter for resonance, one would expect data for all spacers to collapse onto a single curve. On the contrary, the various curves in figure 4.13a are well spaced. This suggests that size variation (caused by dipolar interactions) alone is not sufficient to explain the variation in the LF resonant frequency, implying that dipolar interactions play a more direct role in skyrmion dynamics.

We extend our simulations to understand the dependence of both resonances on the dipolar interaction, tuned by the thickness of the spacer layer. Figure 4.13b displays the field evolution of the LF and HF resonances for various spacer thicknesses. We observe a steep increase in the resonant frequency of the LF mode, while the HF mode evolves weakly with OP field, consistent with our experimental results. The trend in the field evolution of both modes remains valid for all spacer thicknesses. We notice that the fields at which the low frequency mode appears vary widely for different spacers. This can be understood from figure 4.12, where the field range required to stabilise large skyrmions varies by up to 200 mT for different spacers. The spacer thickness-controlled modulation of resonance spectra is therefore technologically relevant for developing applications in specific frequency ranges.

Incidentally, we notice that the field evolution of the HF and LF mode for a 3 nm spacer matches perfectly with our experimental results. This is a surprising discovery, since the 3 nm spacer thickness is higher than the experimental value (2 nm of Ir/Pt spacer). We suspect that this may be caused by a magnetically

‘dead’ region of our films which increases the effective spacer thickness. However, the precise reason for this discrepancy remains uncertain.

FIGURE 4.13: (a) Variation in the LF mode resonance frequencies with skyrmion size. (b) Frequency shifts in the LF and HF modes with the nonmagnetic spacer layer thicknesses. (c) PBC simulated resonance spectra with a spacer thickness of 3 nm. The overlaid data points are from frequency sweep experiments.

Our PBC simulations show that the ‘centre of mass’ of OP magnetization $R_{CoM} = \int r(1 - r)dr / \int (1 - m_z)dr$ gyrates in counter-clockwise fashion. This is shown in figure 4.14. However, the guiding centre of the skyrmion remains pinned during the oscillations. This is expected from the conservation laws mentioned in section 4.2, where a magnetic field gradient is necessary for motion of the guiding centre.

4.4.4 Temperature and Field History Dependence of Resonance Spectra

The static characteristics and spatial configuration of skyrmions evolve with temperature. As recently reported [41], the number density of skyrmions decreases with temperature in Fe(0.5)Co(0.5) thin film. To understand the effect of skyrmion density on resonance, we measure FMR spectra at temperatures down to 100K (figure 4.15). At low temperatures, the dispersion curves shift to higher frequencies, consistent with the reported increase in uniaxial anisotropy at lower temperatures. We expect a reduction in the skyrmion size due to the increased anisotropy field. This size decrease can further shift the dispersion curves to higher frequencies. Furthermore, the observation of skyrmion resonance even at 100K proves the robustness and stability of the skyrmion gyrations.

FIGURE 4.14: CCW gyration of the skyrmion magnetization during HF resonance. White dots represent the topological charge centre of the skyrmion, which remains stationary during oscillations. Magnetization snapshots 1-8 are sequentially separated by equal intervals of ~ 12.5 ps, representing the evolution of the magnetization over time. Scale bar = 20 nm.

It has been established that skyrmion nucleation is strongly depends on the magnetic history of the material. We have observed similar dependencies in our field sweep FMR experiments. Skyrmions are known to exhibit hysteretic behaviour, which has been utilised to develop skyrmion nucleation protocols [31]. To test the magnetic history dependence of our resonance spectra, we perform resonance experiments from two distinct initial magnetization states.

In the first case, the magnetization was initially polarised along the OP direction by an external field larger than the saturation limit (~ 300 mT) of the film. During the field sweep, the magnetization evolves from ferromagnetic order to the skyrmion phase, followed by the emergence of labyrinth stripes close to zero field. In the second case, the magnetic field was swept in the opposite direction from zero to 500 mT. The magnetization transforms from stripes to skyrmions and eventually saturates beyond 300 mT. Depending on the initial magnetization of the film (field polarised or random), the resonance fields in the stripe and FM phases differ

FIGURE 4.15: Resonance modes of Fe(0.5)Co(0.5) measured at low temperatures. (a), (b) and (c) show transmission intensity spectra at 300K, 200K and 100K respectively, acquired via the field sweep FMR technique. The overlaid points are the resonance frequencies extracted by fitting and interpolating the spectra. The bottom row compares resonance spectra at 12 GHz from 100-300K. Dotted white lines in (a), (b) and (c) mark 12 GHz.

significantly (figure 4.16). This is particularly noticeable for the stripe phase, where the resonance shifts to lower fields for a saturated initial magnetization. This is an expected result as the magnetization of the stripe phase, at a given magnetic field, will depend on the initial magnetization state. Essentially we are performing field sweeps across the two separate branches of a hysteresis loop, which follow different magnetization paths for an identical magnetic field.

The field history dependence can also be observed in the relative magnitude and linewidth of the resonance spectrum. An initially saturated state generates a weaker absorption signal with a large field linewidth, while the field sweep up from zero magnetization displays a strong and relatively sharp spectrum. The effect of changing the magnetic history is also visible in the high and low frequency resonances within the skyrmion phase. The zero-field initial state exhibits an

FIGURE 4.16: Magnetic history dependence of resonance spectra in Fe(0.5)Co(0.5) at 300K. For (a) and (b), the field sweep starts from zero field and saturated magnetizations, respectively. (c) and (d) are resonance spectra acquired at 10 GHz (marked by white dotted lines in (a) and (b)).

HF mode spread over a narrow field range, while the HF resonance for a field-polarised initial magnetization is extended to comparatively high fields. For the low frequency mode, the field range displays an offset of ~ 50 mT. Such differences in the resonance spectra can be attributed to the distinct number densities of the skyrmions caused by different initial magnetization states. Furthermore, the size of the skyrmions varies inversely during opposite field sweep directions, which can also cause differences in the resonance dispersions.

Broadband FMR experiments were also performed on a Fe(0.4)/Co(0.6) film, which displays similar resonance spectra corresponding to the various magnetization states of the magnetic phase diagram (figure 4.17). A major difference from the Fe(0.5)Co(0.5) film can be seen in the frequency ranges corresponding to the different modes. We attribute this to the distinct magnetic parameters exhibited by the two films. The difference in the frequency ranges for each mode suggests a tunability of resonance modes with the relative thickness of the FM layers, which could be utilised for future practical applications of skyrmion resonance.

FIGURE 4.17: Normalised resonance spectra in a Fe(0.4)Co(0.6) film from 300K to 100K. The spectra exhibit four modes corresponding to FM, skyrmion and stripe magnetizations at all temperatures, similar to Fe(0.5)Co(0.5) films.

4.5 Conclusion and Outlook

In this chapter, we described the magnetization dynamics of various magnetic phases in Ir/Fe/Co/Pt films. Notably, we demonstrate that, unlike Bloch skyrmions in bulk magnets, skyrmions in multilayer films exhibit two distinct dispersion modes in the field region occupied by skyrmions. The high and low frequency modes (HF and LF) show different dependences on the magnetic field. While the LF mode rises steeply with the magnetic field, the frequencies of the HF mode are almost independent of the field. From our micromagnetic simulations, we identify counter-clockwise (CCW) gyrations of the skyrmion magnetization during both modes. However, as we deduce from Fourier analysis of our time-resolved magnetization data, while the CCW oscillations are the primary contributor to the LF mode resonance, the HF mode mainly originates from scattering of magnons (generated by uniform precession of the ferromagnetic background) off the skyrmions. Furthermore, we demonstrate that the frequency range of the resonant excitations can be tuned for specific applications by varying the nonmagnetic spacer layer thickness.

As already established [31, 33, 41], skyrmions in Ir/Fe/Co/Pt multilayers exist in a densely packed, disordered configuration. Moreover, as MFM images indicate, there is a significant variation in the shape and size of skyrmions, with the latter following a Gaussian distribution centred at ≈ 50 nm. Remarkably, even a disordered cluster of non-identical skyrmions can generate robust resonance spectra, which can be detected by two independent techniques with varying levels of sensitivity. Our time-resolved simulations indicate that the HF mode is delocalised. In the field range 150-250 mT, the skyrmions are tightly packed with a mean separation of less

than 50 nm (see the MFM images in chapter three). Therefore, it is possible that within certain field ranges, an array of skyrmions (in the same layer) could oscillate coherently. A suitable geometry to understand the effect of skyrmion-skyrmion interactions on resonance spectra would be an array of nanodots, where the ratio of skyrmion size and dot radius governs the number of stable skyrmions in the dot [27, 42]. Within this geometry, the dot size and separation would be varied to tune the inter-skyrmion interactions. For weak inter-skyrmion interactions, the skyrmions will gyrate individually, while for strongly interacting skyrmions (similar to a lattice) the resonance will involve coherent oscillations of all the skyrmions.

Irrespective of the precise origin of the resonance (collective or isolated), skyrmions in these films offer an unprecedented opportunity to explore microwave applications for skyrmions. Unlike previous studies, which have observed skyrmion resonance phenomena at low temperatures and in single crystals, Ir/Fe/Co/Pt films exhibit skyrmion resonance over a wide range of temperature and material parameters. Also, sputtered thin films offer excellent compatibility with existing semiconductor electronics, thus facilitating application development. Moreover, as our experiments and simulations suggest, the frequency range of the mode dispersions can be tuned with the thickness of the constituent atomic layers.

As the field of skyrmionics has progressed rapidly in the past few years, proposals already exist to exploit skyrmion oscillations for novel applications [43–45]. These include RF diodes, which harness the asymmetric wave propagation in skyrmions, and nano-oscillators utilising radial oscillations of the skyrmion core. Another proposal explores the functionalities of skyrmions as spin-transfer torque diodes (STTDs) [45], which convert a microwave current into a DC voltage. The output voltage is proportional to the input microwave current I_{rf} , the oscillating resistance ΔR of the material and the phase difference between I_{rf} and ΔR . The performance of a STTD is measured in terms of its sensitivity (i.e. the ratio of its output DC voltage to input microwave power). Traditionally, due to their low sensitivity (1.4V/W), STTD effects are employed only in magnetic tunnel junctions (MTJ) for specific purposes. It has been proposed that using zero-field stable skyrmions as the free layer of a MTJ could enhance the sensitivity of the STTD.

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Chapter 5

Conclusions and Future Directions

5.1 Summary of Work Performed

In this thesis, we studied the magnetization dynamics of Néel skyrmions, stabilised by an interfacial DMI and perpendicular magnetic anisotropy (PMA) in Ir/Fe/-Co/Pt multilayer thin films. We described the fundamentals of magnetization dynamics and details of our experimental set-up. We provided a detailed magnetic characterisation of our films and demonstrated the ability to tune skyrmion properties by varying the thickness of the Fe/Co layer. Next, we used a home-built broadband ferromagnetic resonance (FMR) probe to perform field and frequency sweep FMR experiments in which we observed four distinct modes originating from the labyrinthine stripe phase at low fields, the skyrmion phase at intermediate fields and the field polarised ferromagnetic phase at high fields in our films. While the stripe and ferromagnetic phase displayed the expected resonance spectra, we observed two resonance modes within the skyrmion phase. We identified the physical mechanisms responsible for both resonances by using micromagnetic simulations and analytical calculations.

We began the thesis with a description of the microscopic magnetic interactions responsible for skyrmion formation in noncentrosymmetric magnets and thin films comprising ferromagnet-heavy metal interfaces. In particular, we focused on the Dzyaloshinskii-Moriya interaction generated by strong spin-orbit coupling and

the absence of spatial inversion symmetry in the material. We noted that the unique properties exhibited by skyrmions - such as an emergent electromagnetic field and a skyrmion Hall effect - are due to their unique ‘topological’ character. Subsequently, we described the potential advantages of skyrmions over silicon-based CMOS (complementary metal-oxide-semiconductor) technology. Due to their small size and energy-efficient dynamics (when driven by low amplitude electric and/or spin currents), skyrmions are suitable candidates for building the next generation of spintronic devices. Finally, we discussed the postulated applications of skyrmions in logic operations, data storage and radio frequency (RF) technology.

To realise the potential applications of skyrmions, it is of profound importance to understand the eigenmodes of excited skyrmions, which govern their field and current-induced dynamics. We employed a broadband ferromagnetic resonance (FMR) technique to study the internal excitations of skyrmions in thin films. Chapter two detailed the fundamentals of magnetic relaxation, which is often quantified by an effective damping parameter (α_{eff}). We briefly described the various intrinsic and extrinsic mechanisms of magnetic relaxation, including spin-pumping effects in multilayers comprising ferromagnet-heavy metal interfaces, where spin currents from the ferromagnet are pumped into the adjacent heavy metal, resulting in a decay of the magnetization. In the later sections, we described the operating principles and instrumentation details of a home-built broadband FMR probe capable of measurements at temperatures down to 4.2 K.

We synthesised Ir/Fe/Co/Pt multilayers hosting magnetic skyrmions by magnetron sputtering. In chapter three, we detailed the design and optimisation of FM-HM interfaces with tunable magnetic interactions, leading to a stable skyrmion phase. We demonstrated that magnetic interactions (exchange, DMI and uniaxial anisotropy) at the FM-HM interface can be modulated via the composition and architecture of the interface (figure 5.1). We employed novel material design techniques, involving an additive DMI at two interfaces resulting in a large net DMI, to stabilise sub-50 nm skyrmions in Ir/Fe/Co/Pt multilayers. We characterised the magnetic properties of our films using magnetic force microscopy (MFM) and magnetization measurements. The magnetization was observed to evolve from labyrinthine stripes at low fields, to magnetic skyrmions at intermediate fields and eventually a saturated magnetization above 300 mT.

Chapter four described the FMR experiments and micromagnetic simulations on Ir/Fe/Co/Pt films. Our FMR experiments were performed via field as well as

FIGURE 5.1: (a) Additive DMI from Fe/Ir and Pt/Co interfaces in Ir/Fe/Co/Pt multilayers. (b),(c) Tuning of A , D , M_s and K_{eff} with the thickness of the Fe/Co layers.

frequency sweep methods (figure 5.3). Although skyrmion nucleation is sensitive to the magnetic history of a material, we observed near-identical resonance spectra in both field and frequency sweep experiments. Four distinct dispersion modes were detected, corresponding to the various magnetic phases in our Ir/Fe/Co/Pt films. For applied fields larger than the saturation limit, we noticed a linear proportionality between the resonance field and the excitation frequency, originating from the precession of the magnetization around the effective magnetic field. The field sweep resonance spectra were fitted with a combination of symmetric and anti-symmetric Lorentzian functions to extract the linewidth and (eventually) the effective damping. We attribute the large observed $\alpha_{eff} \simeq 0.09$ to the metallic nature and multiple FM-HM interfaces ($\simeq 80$) in our films (figure 5.2).

FIGURE 5.2: (a) Home-built FMR probe capable of measurements down to 4.2 K. (b) Field sweep resonance spectra in the ferromagnetic phase of Fe(0.5)Co(0.5), measured at 300 K and fitted with a combination of symmetric and antisymmetric Lorentzian functions. (c) Linear variation in the resonance linewidth with frequency, demonstrating a large ‘effective’ damping $\alpha_{eff} = 0.08$.

Slightly below the saturation field, in the field ranges corresponding to the skyrmion phase, the Kittel mode splits into high and low frequency (HF and LF) branches. The HF resonance exhibits a weak dependence on the out-of-plane (OP) magnetic field, while the LF mode rises steeply with the field. At even lower fields (0-150 mT, at which magnetic stripes develop), the resonance frequency rises linearly as the field decreases (figure 5.3). Low temperature FMR spectra indicate similar trends with a slight shift towards higher resonant frequencies, originating from an increase in the PMA at lower temperatures.

FIGURE 5.3: (a) Frequency and (b) field sweep resonance spectra of Fe(0.5)Co(0.5) at 300K. Overlaid circles represent absorption maxima, which are extracted by fitting our raw data. Different colours correspond to resonances in the ferromagnetic phase (blue dots at large fields), skyrmion phase (cyan and green dots at intermediate fields) and stripe phase (pink dots at low fields).

The micromagnetic simulations and analytical calculations identify the counter-clockwise (CCW) gyrations of skyrmions and the uniform precession of the ferromagnetic background as the physical mechanisms responsible for our skyrmion resonance spectra. While the LF resonance primarily originates from CCW oscillations of skyrmions, the HF resonance is caused by scattering of uniform precession magnons in the ferromagnetic background off skyrmions. Through our periodic boundary condition (PBC) simulations, modelled in figure 5.4a, we established that interlayer magnetostatic interactions can be tuned via the thickness of the nonmagnetic spacer layer, causing a shift in the resonance frequencies of the LF and HF modes (figure 5.4b). This discovery could be developed for future microwave applications harnessing gyration modes of skyrmions. Resonance spectra simulated using a FM(1 nm)/HM(3 nm) stack are consistent with our experiments (figure 5.4c). Since the spacer thickness of 3 nm is higher than the experimental value

FIGURE 5.4: Summary of our PBC simulations. (a) Material geometry for simulations with variable spacer thickness t . (b) Variation in resonant frequencies for the HF and LF modes with spacer thickness. (c) Experimental data points for Fe(0.5)Co(0.5) are overlaid on simulated resonance spectra, obtained using a [FM(1 nm)/Spacer(3 nm)] model: the experimental and simulated data agree closely. The spacer thickness (3 nm) is higher than the experimental value of 2 nm.

of 2 nm, we suspect the presence of magnetic ‘dead’ layers in our sputter-grown films.

5.2 Future Directions

5.2.1 Skyrmion Dynamics in Thin Films with In-Plane Anisotropy

Both multilayer compositions reported in the previous chapters (Fe(0.5)Co(0.5) and Fe(0.4)Co(0.6)) exhibit PMA. To understand the role of magnetic anisotropy in resonance spectra, we are also experimenting with thin films displaying IP anisotropy. Figure 5.5 displays magnetization and field sweep resonance spectra for a [Ir(1)/Fe(0.2)Co(1.6)/Pt(1)]₁₀ film with IP anisotropy. The microwave absorption in this case is strikingly different from our other films. Typically, in multilayer thin films, the skyrmion resonance is characterised by HF and LF dispersion curves, with slopes lower and higher than the Kittel mode. However, for Fe(0.2)Co(1.6), we only observe one resonance mode corresponding to skyrmions. Moreover, the stripe phase consists of two dispersion modes with distinct slopes. This work is still in progress.

FIGURE 5.5: Magnetization and broadband resonance spectra of Fe(0.2)/Co(1.6) at 300K. (a) Out-of-plane (OP) and in-plane (IP) $M(H)$ curves, indicating a large in-plane anisotropy. (b) Transmission intensity map measured in OP field sweep experiments, indicating four resonance modes, labelled as i, ii, iii and iv. (c) Field sweep line cuts at 3.0 GHz, 2.2 GHz and 1 GHz. The dotted lines mark the positions of the local resonance fields.

5.2.2 Role of Skyrmion Size and Inter-Skyrmion Interactions on Resonance Spectra

The work presented in this thesis is focused on thin films with a high skyrmion density. Moreover, the skyrmion shapes and sizes vary over a wide range due to the sputtered growth technique. The presented results establish the influence of interlayer magnetostatic interactions on skyrmion resonance and suggest a material architecture-based tuning of resonance spectra. However, there are still many questions which remain unanswered. These include (a) the effect of intralayer inter-skyrmion interactions and (b) the influence of skyrmion sizes and shapes on resonance spectra. Possible future extensions of this thesis involve addressing these questions.

In a ferromagnetic monolayer (chosen to avoid the complexity arising from interlayer interactions in multilayers), resonant absorption in the skyrmion phase can have two possible origins: (a) all skyrmions are oscillating individually in a phase-coherent manner, and the observed spectra are due to a collective excitation of all skyrmions, or (b) the skyrmions are oscillating at an identical frequency but in an incoherent manner and the observed signal is a superposition of all the individual resonance signals. In (b) we have not included the frequency variation arising from the size difference, to keep the discussion simple.

FIGURE 5.6: Schematic of the experimental set-up for broadband FMR experiments on nanodot arrays. The wide pink arrows denote the direction of RF wave propagation. The oscillating magnetic field (h_{rf}) acts in-plane on the top surface of the central conductor, and out-of-plane in the spacing between the central conductor and ground plane.

To resolve the queries mentioned earlier, we propose broadband FMR experiments on an array of nano-sized cylinders (dots). The nanodot sizes can be tuned to modulate the skyrmion sizes and configurations [1–3]. Such controllable material geometries will allow us to distinguish between resonance spectra of isolated vs. interacting skyrmions, and would therefore, enable us to determine the effect of inter-skyrmion interactions on skyrmion resonances.

The proposed experiment requires fabrication of a nanodot array using lithography techniques. The size and periodicity of the dots can be tuned to control the skyrmion order (isolated or lattice) and inter-skyrmion interactions (via the array periodicity). We propose FeGe as an ideal material for such experiments, since (a) thin films of FeGe host skyrmions close to room temperature, and (b) the phase diagram of FeGe is well understood and therefore eliminates ambiguity in understanding the experimental results. Figure 5.6 displays a schematic of the proposed experiment for the proposed experiment. The nanodot array is placed upside down onto a CPW. The top layer of the array is coated with a thin resist layer, to avoid an electrical short between the central conductor and ground. The dot diameter is smaller than the spacing between the central conductor and ground planes of the waveguide. Therefore, dots located in this region will experience a perpendicular RF field, leading to breathing mode oscillations of the skyrmions. Such an experimental design also overcomes the limitation of thin

film geometry, which does not allow the excitation of breathing modes in the experiments reported in earlier chapters. A comparison of resonance spectra in arrays with varying dot size and periodicity will allow us to elucidate the influence of inter-skyrmion interactions on resonance spectra. The proposed array geometry is also technologically relevant. A dense skyrmion array can be utilised as a memory device, where data manipulation can be achieved via localised microwave pulses.

5.2.3 Spin-Waves and Skyrmions

Another attractive possibility for nanodevices is the interaction of skyrmions (localised spin excitation) with delocalized spin textures such as domain walls and spin-waves. Understanding such interactions has both fundamental and technological implications. On the fundamental side, it provides an opportunity to understand wave-particle interactions in condensed matter systems. Technologically, spin-waves can reverse the magnetization of skyrmions, which has applications in logic and data storage [4]. Furthermore, recent works [5] have established the role of spin-waves in domain wall depinning, which can be utilised in racetrack memories. It was observed that collisions of neighbouring domain walls with opposing magnetization rotation generates spin-waves. A similar question can be asked for skyrmions: how will two skyrmions with opposite topological charges interact? However, experiments featuring skyrmions of opposite topological charge are not possible in materials hosting DMI-induced skyrmions. The DMI restricts the chirality of the spin rotations, and hence does not allow simultaneous formation of skyrmions with opposite charges. In contrast, the skyrmions generated due to dipolar interactions do not display any chirality preference [6], and can therefore be employed for the proposed experiments. Thick insulator films, which can stabilise dipolar skyrmions and exhibit low damping are suitable materials for such experiments.

In the field of magnonics, various geometric arrangements of skyrmions could be utilised as magnonic crystals. For example, Bloch skyrmions exist in triangular and hexagonal lattices. Also, the skyrmion lattice in nanostructures can be tuned to form a one-dimensional chain of skyrmions, which could be exploited as a one-dimensional magnonic crystal. Furthermore, the size and lattice arrangement of skyrmions can be tuned with external perturbations, suggesting the possibility of developing dynamic magnonic crystals [7].

A particularly challenging task in magnonics is the generation of sub-100 nm wavelength spin-waves. Traditional approaches such as CPW or stripline fabrication, or even relatively modern schemes such as spin-torque oscillators and grating couplers, can only produce spin-wavelengths down to 375 nm. However, recent developments have shown that magnetic vortices can be utilised to generate nanoscale spin-waves [8]. A pair of antiferromagnetically coupled vortices can act as a source of spin-waves due to the gyration of the vortex cores around their equilibrium positions. The spin-wave frequency can be controlled by varying the coupling strength between the vortices. Skyrmions are topological quasi-particles, similar to vortices, which can exhibit in-plane CW and CCW magnetization gyrations. Therefore, skyrmions could also be exploited for spin-wave generation. Moreover, unlike vortices, the size and lattice configuration of skyrmions can be controlled via material composition, which will help to modulate the spin-wave spectra.

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Appendix A

Design and Construction of a Low Temperature Radio Frequency Sputtering Chamber For Growth of Homogeneous Thin Films

A.1 Introduction

The skyrmions observed in Ir/Fe/Co/Pt films display significant variations in their size (up to 20 %) as well as distortions from an axially symmetric structure. Such deviations from the ideal skyrmion structure are caused by local inhomogeneities in the magnetic parameters, originating from the disordered structure of sputtered thin films lacking atomically sharp interfaces. Regional variations in the energy landscape have previously been observed in dynamic experiments, where skyrmions are pinned at random sites [1]. Consequently, a large critical current is required to move pinned magnetic textures, thus reducing the energy efficiency of the process. Furthermore, the strengths of interfacial effects (such as interfacial DMI and spin Hall torques) are strongly influenced by the sharpness of the interface and deteriorate in rough interfaces. Therefore, it is essential to develop new fabrication methods for low-dimensional ultrathin films, which can be exploited to explore novel phenomena in magnetism. In this chapter, we will detail the design and specifications of our home-built low-temperature sputtering apparatus, which allows the possibility of on and off-axis deposition of thin films.

A.2 Theoretical Background to Magnetron Sputtering

In physical vapour deposition (PVD) techniques, a solid material (known as the ‘target’) is vaporised in a high vacuum environment, enabling deposition of the vapours onto another material, referred to as the ‘substrate’. In general, PVD refers to a group of fabrication techniques - all of which utilise physical methods (such as heating, ablation etc.) to release atoms from the target. For example, during thermal deposition resistive heating is employed to heat the target above its boiling point and generate vapours of target atoms. Similarly, high energy collisions are utilised to eject target atoms during the sputtering process. In general, there is no chemical reaction between the target atoms and the substrate surface during PVD growth. Sputtering is an ideal technique for materials with high boiling points which cannot be deposited easily via other techniques.

In a sputtering facility, the target and substrate are placed inside a vacuum chamber and connected to the negative and positive electrodes of a high voltage power supply. An inert gas (typically argon) is injected into the vacuum chamber, which acts as a medium for the plasma. In a typical sputtering geometry, the target and substrate are placed in line with each other. This configuration is referred to as on-axis sputtering. In on-axis geometry, high energy target ions are deposited on the substrate. Films resulting from such bombardment of high energy target ions onto the substrate can be grown quickly, but lack a smooth surface and do not support epitaxial growth. A modified version of on-axis sputtering is used to enhance the smoothness and epitaxial growth of films, and is known as off-axis sputtering. In this geometry, the target normal is oriented at a high angle (up to 90 degrees) to the substrate (figure [A.1](#)). Therefore, only low energy target atoms are deposited onto the substrate. Off-axis deposition has been widely used to grow cuprate superconductors, in which the stoichiometry and morphology of films can be carefully controlled with the target-substrate angle [\[2\]](#). More recently, it has been demonstrated that a combination of on and off axis growth can be utilised to deposit epitaxial oxide layers [\[3\]](#).

The stabilisation of a plasma is central to successful magnetron sputtering. A plasma is an ionised state of matter which contains ions, free electrons and neutral atoms, maintained in equilibrium by a continuous process of ionisation and recombination. Typically, a plasma is generated by confining a pressurised gas between oppositely

FIGURE A.1: Schematic diagram of off-axis sputtering

charged electrodes. In the sputtering process, positively charged Ar ions, accelerated at a few keV, penetrate below the surface of the target (which is at a negative potential) and transfer their momentum to multiple target atoms via elastic collisions. This momentum transfer results in ejection of atoms from the target, along with secondary electrons and photons. The minimum energy required to eject an electron from a gas atom is referred to as the ‘ionisation energy’. For a successful ionisation process, the energy transferred by a fast-moving electron to the gas molecule must be higher than the ionisation energy. The kinetic energy of the electrons and ions is proportional to the electrode voltage. While low energy ions are reflected back from the target, ions with higher energies will result in ion implantation within the target (figure A.2). In both cases, there will be no sputtering. Therefore, the electrode voltage can be used to control the number density of ions and electrons in the plasma. However, a low concentration of gaseous atoms can destabilise the plasma. Therefore, a suitable balance of electrode voltage and gas concentration is required to sustain the plasma.

Growth by sputtering consists of two independent stages. The initial ejection of target atoms and their ensuing motion towards the substrate are controlled by the plasma characteristics. Subsequently, the target atoms are deposited on the substrate, a process which is governed by the adsorption capacity and surface mobility of the target atoms. Several key control parameters for the deposition of the target atoms on the substrate surface include the crystallographic orientation of the substrate, the presence of impurity atoms and the substrate temperature. The temperature dependence of the deposition process can be characterised in terms of the parameter T/T_m , where T is the substrate temperature and T_m is the

FIGURE A.2: Possible outcomes of the collision of high energy Ar ions with the sputtering target. The probability of the various outcomes is tuned by the energy of the ions and the surface characteristics of the target.

melting point of the target material. Figure A.3 displays the characteristics of Cu and Al-alloy thick films ($25\ \mu\text{m}$) deposited at $1000\text{-}2000\ \text{\AA}/\text{min}$ on $T^* \equiv T/T_m$ and the electrode voltage. As is evident from figure A.3, adjusting the parameters allows the granular size and crystalline structure of a sputtered film to be tuned from large grains (resulting in porous films) to fine-grained films with a definite crystal orientation.

The key requirements for thin films in spintronic applications are uniform thickness and high purity. In sputtering growth, the uniformity in the film thickness is controlled by the geometry of the sputtering chamber, the relative separation between the substrate and target, and the relative orientation of the substrate and target. The flux of ejected atoms (from the target) follows a cosine distribution (figure A.4). Consequently, we may write the mass deposited per unit area from a surface source as [5]:

$$\frac{dM_s}{dA_s} = \frac{M_e \cos \phi \cos \theta}{\pi r^2} \quad (\text{A.1})$$

where M_e and M_s are the total ejected and deposited masses, A_s is the area of the substrate, θ is the angular position of the substrate relative to the target, ϕ is the angular deviation of the substrate normal from the flux direction and r is the separation between the target and substrate. For a parallel orientation of target and substrate (i.e. on-axis geometry), $\cos \theta = \cos \phi = 1$, and hence the majority of the incoming flux will be deposited on the substrate.

The maximum deposition thickness d_0 is expected for $\theta = 0$, as the target atoms flux flow is maximised in this direction. However, the thickness uniformity falls as

FIGURE A.3: Thornton zone diagram [4] depicting the effects of substrate temperature and the electrode voltage on the structure of the deposited film. The figure is reproduced from [4].

θ rises. For the planar source (in which a wide area of the target is accessible to the plasma), the thickness variation across the substrate surface is estimated by

$$\frac{d}{d_0} = \frac{1}{(1 + (\frac{l}{h})^2)^2} \quad (\text{A.2})$$

where h is the distance between the target and substrate centres, and d is the thickness at distance l away from the substrate centre. Although the uniformity in the film thickness increases with the separation between target and substrate, this increases wastage of the target material. Furthermore, non-uniformity in thickness can also appear due to anomalies in the spatial distribution of incoming flux. Such irregularities can be normalised by continuous rotation of the substrate during the deposition.

The purity of the deposited film is an important criterion for establishing film quality. The impurities in a sputtered film can have two possible origins: (1) contaminants present in the target itself, or (2) residual gases present in the

FIGURE A.4: The amount of flux received at the substrate is proportional to the relative position (defined by θ) and the relative orientation of the surface (measured by ϕ) from the target. This relation is quantified by equation A.1. (b) For a parallel orientation of target and substrate, the thickness decreases at larger distances from the substrate centre, as described by equation A.2.

vacuum chamber. The first cause can be eliminated by using high purity targets. The second reason, however, requires a detailed explanation of the sputtering process. During the deposition, the substrate experiences an incoming flux of target atoms as well as gaseous atoms and ions constituting the plasma. Although the primary constituent of the plasma is an inert gas (Ar), which is not adsorbed by and does not react with the substrate, other gaseous impurities such as H_2O , CO_2 , O_2 and N_2 are often present inside the deposition chamber in low quantities. These gases can be deposited alongside the target atoms, hence altering the stoichiometry and homogeneity of the deposited film. The concentration of impurities in the deposited film is directly proportional to the partial pressure of these gases and decreases at high deposition rate.

Moreover, the target atoms collide with the gaseous ions in the plasma on their way to the substrate. Such collisions alter the velocities (both in direction and magnitude) of the target atoms. Consequently, the target atoms can reach the substrate from various orientations and with varying energy, resulting in non-uniform deposition. A low plasma pressure maximises the probability of unobstructed and long mean free paths for the target atoms. Therefore, minimising the chamber pressure is a recommended practice for deposition by sputtering. However, as explained earlier, a sufficiently high pressure is necessary to sustain the plasma in

the chamber. At such low pressures, the plasma is sustained by effectively confining the secondary electron trajectories over the target using permanent magnets, a process referred to as ‘magnetron sputtering’.

The design of a magnetron sputtering source consists of a magnetic pole at the centre of the target, with a ring of magnets forming the other pole at the target perimeter, resulting in a parabolic magnetic field between the centre and edge of the target [6]. This field distribution allows effective focussing of the plasma beam over the substrate, even at low chamber pressures. A perpendicular magnetic field results in circular electron trajectories. The strength of the required magnetic field is limited by the cyclotron radius of the electron, equivalent to $r_g = \frac{m_e v_e^\perp}{eB}$, where m_e and $v_e^\perp$ are the electron mass and velocity perpendicular to the magnetic field B . To focus the electrons over the target, the radius r_g should be smaller than the target dimensions.

A.3 Instrumentation in our Sputtering Chamber

A sputtering tool consists of a target and substrate inside a vacuum chamber. The target is attached to a sputter gun, while the substrate is mounted on a rotatable stage. The vacuum chamber is connected to a pump assembly (comprising a scroll pump and a magnetically levitated turbomolecular pump) via gate valves. A pure inert gas (Ar) or a combination of gases (e.g. Ar + O₂, Ar + N₂) is used to create a plasma. A schematic diagram of our sputtering chamber is shown in figure A.5. The flow rate of each gas is independently regulated via mass flow controllers connected between the chamber and the gas bottles.

Vacuum chamber

The sputtering chamber is a hemispherical chamber made of non-magnetic stainless steel (SS-316) with a mumetal shield. The top lid of the chamber includes three DN63 ports and one CF200 port for the sample stage. The DN63 ports are used as viewports. A turbomolecular pump is attached via a CF200 T-junction on the top lid of the chamber. A pressure gauge and a butterfly throttle valve are also attached to this T-junction.

The hemispherical surface of the chamber consists of six CF100 ports arranged in a confocal geometry, with the focus being the centre of the sphere. Six sputtering guns will be attached to these ports. Side, top and bottom views of the chamber

FIGURE A.5: Schematic of our deposition chamber in on axis sputtering mode

are shown in figure A.6 a, b and c. The central axis passing through the chamber centre is the most suitable position for the substrate, as it is symmetric from all the targets. A particular advantage of this geometry is the possibility of co-sputtering from multiple targets simultaneously, which is beneficial for the deposition of alloys. Four symmetrically-arranged CF40 valves are located close to the bottom of the chamber. Two of these ports host an e-beam evaporator (Specs EBE-4) and an argon ion sputter gun (Specs-IQE 11/35). An intermediate gate valve is connected to each CF40 port to ensure the isolation of the chamber. A quartz crystal microbalance is connected via a 150 mm UHV linear manipulator on a CF40 port located parallel to the sample stage, to record the thickness and homogeneity of the film. The base pressure of the chamber was measured by a Leybold ITR-200 S hot cathode gauge, which operates from atmospheric pressure down to 10^{-10} mbar. The sputtering chamber is pumped by a combination of a $15 \text{ m}^3/\text{h}$ scroll pump and a 600 ls^{-1} turbomolecular pump (Leybold Turbovac Mag W600P) mounted on top of the chamber. The pump assembly is connected to the main chamber via a butterfly valve (MKS-MPE 653B) and a Baratron capacitance manometer. The manometer and throttle valve operate as a monitor and control unit and are connected to an MKS-600 pressure controller.

The sample stage can be accessed from the bottom of the chamber, which contains a horizontally-aligned CF100 port. At this port, a loadlock and wobble stick assembly are connected to the chamber via a second gate valve. The load lock is

FIGURE A.6: A 3D model of the sample stage (a), top lid (b) and underside (c) of the sputtering chamber.

connected to a vacuum pump assembly via a gate valve. The pressure in the load lock chamber is monitored by a combined Pirani/ion gauge.

Before preliminary testing, the chamber was baked at 80°C for five days. This removes water vapour and hence reduces the outgassing from the walls and other components inside the chamber. The chamber was baked using heat tapes wrapped around the chamber surface. A layer of aluminium foil was subsequently applied to ensure uniform heat distribution across all parts of the chamber; K-type thermocouples were used to monitor the temperature. The base pressure of the sputtering chamber after five days of low temperature baking was 10^{-9} mbar.

Sample Holder and Vertical Manipulator

The substrate holder is mounted at the bottom end of a custom-built, variable temperature sample stage manufactured to our design by FerrovacTM GmbH. The substrate holder is connected to a stepper motor, which continuously rotates the substrate during the deposition process, ensuring a uniform distribution of the

target atoms. The sample stage is attached to a vertical manipulator and can hence be moved vertically. The sample stage and its vertical manipulator are shown in figure A.7. The unique feature of this stage is the ability to cool down to 20K using liquid helium, while maintaining continuous rotation *and* electrical connections to the sample. A transfer line port located on top of the stage carries liquid helium to the base of the sample stage. Here the transfer line coils around a copper bath, before rising and eventually exiting at the top of the stage. The copper bath can be cooled down to 20K with a sufficiently high flow rate of liquid helium ($\sim 11/\text{hour}$). The copper bath is thermally linked to a circular plate via thin silver-plated copper beryllium leaf springs. The springs apply pressure to the circular plate, but are not screwed onto it, thus allowing it to rotate while maintaining a high thermal conductance to the copper bath.

The bottom of the sample holder is mounted onto the circular plate and contains a heater and temperature sensor. Substrates are transferred to the sample stage using a custom-built flag style sample holder. These sample-holders clip onto a substrate holder during transfer: three samples $10 \times 10 \text{ mm}^2$ can be carried simultaneously in this design (figure A.8). The sample holders can be moved between the deposition stage and the substrate holder using a wobble stick.

The bottom section of the sample stage is connected to the load lock. A load-lock is a small chamber which isolates the main chamber from atmospheric pressure while changing the substrates. One end of the load lock is connected to the sputtering chamber via a gate valve, and the opposite end contains a linear translator. The load lock chamber is connected to a pump assembly (scroll + turbomolecular pump) and a pressure gauge. In our case, we have attached a portable pumping cart to the load lock. During the substrate change procedure, the load lock is pumped down to a pressure approaching that of the main chamber ($\leq 10^{-8} \text{ mbar}$). For sufficiently close pressures, the gate valve between the load-lock and the main chamber can be opened, and the sample holder inserted by the translator. Once the substrate is back in the load lock after deposition, the main gate valve can be closed, thus isolating the main chamber. The pressure of the load lock is then increased by introducing an inert gas (typically dry N_2). Once the load lock chamber pressure exceeds atmospheric pressure, it can be opened to change the substrate.

Sputter Guns

The sputter guns used for magnetron deposition are manufactured by AJATM (model A-320). The target is located at the end of the gun, surrounded by a conical

FIGURE A.7: (a) Sample stage and vertical manipulator assembly. The top of the sample stage (b) contains the stepper motor as well as ports for the electrical connections and cryogenic liquid transfer. The bottom section (c) contains the cold stage and substrate holder.

gas chimney (figure A.9) which helps to focus the plasma on the target surface and allows deposition at lower chamber pressures. The target can be tilted up to 45 degrees away from its initial position via a manual control rotor, located at gun flange. This tilt functionality in combination with the vertical displacement of the substrate holder allows a range of deposition angles and substrate - target separations, thus allowing *in-situ* on and off axis deposition. This is ideal for

FIGURE A.8: (a) Diagram of the substrate holder, which can hold three $10 \times 10 \text{ mm}^2$ substrates simultaneously. The substrate holder is connected to a linear translator. (b) The actual substrate holder during assembly.

deposition of alloys, where the tilt can be fine-tuned to control the concentration of target atoms at the substrate. The permanent magnets are located directly behind the target and are not shielded: the magnetic field lines hence extend beyond the target, which localises electrons and thus creates a denser plasma. The flange of the gun also includes a pair of VCR ports for chilled water. A pneumatically controlled shutter protects the target while not in use. A major advantage of this model is the ability to inject Ar gas close to the target surface, which provides better control of the plasma characteristics. O_2 and N_2 can also be injected at the gun in a precisely controlled ratio, which is a stringent requirement for the deposition of oxides and nitrides. The O_2 - N_2 ratio is tuned via the mass flow controllers attached to the gas bottles. All the components in the gas handling system are manufactured by Swagelok. The electrical connector supplying the RF power is located at the base flange of the gun, which is connected to a RF power source via a RF matching network. We chose power supplies and matching networks from Paramount due to their high stability at low pressures and low RF power.

Quartz Crystal Microbalance (QCM)

A quartz crystal microbalance (QCM) is a standard instrument for measuring the thickness of deposited films. Its operation is based on the piezoelectric properties of a quartz crystal, where a strain along crystal planes generates a voltage across the crystal and vice versa. An AC voltage applied between opposite crystal faces induces mechanical vibrations in the crystal. If the thickness of the crystal t_q is equal to an integer number of half-wavelengths, a standing wave is stabilised in the crystal. The frequency of the standing wave is referred to as the natural frequency f_0 of the crystal. f_0 is a function of the material characteristics (such as mass and

FIGURE A.9: Image of an AJA-A320-XP sputter gun used in the sputtering chamber (a). The gun flange (b) contains ports for chilled water, the RF power supply and the inert gas. The top section (c) includes a tiltable target holder, and a pneumatically controlled shutter.

density) and the sample geometry. Mathematically, it is given by

$$f_0 = \sqrt{\frac{\mu_q}{\rho_q}} / 2t_q \quad (\text{A.3})$$

where ρ_q is the density and μ_q the shear modulus of the quartz crystal. In the sputtering chamber, the quartz crystal is placed close to the substrate and receives an approximately identical particle flux from the target. This causes an increase in the mass of the crystal and consequently a shift in its resonant frequency. The resonant frequency can be calibrated for various target materials and correlated to the thickness of deposited material. The increment in the resonance frequency of the quartz crystal with additional mass is

$$\delta f = \frac{2f_0^2 n}{A\sqrt{\rho_q \mu_q}} \delta m \quad (\text{A.4})$$

where δf is the frequency variation caused by a quantity δm of additional deposited mass over the quartz crystal and n is the harmonic number.

At present, the sputtering chamber is fitted with two guns, each connected to an independent RF power supply and matching network (figure [A.10](#)). Copper targets have been installed in the guns to test the dependence of deposition rates on chamber pressure, RF power etc.. In our preliminary results, we have sustained a stable plasma down to 10^{-3} mbar. Testing of the base pressure and the thermal stability of the sample stage is currently in progress. We expect the thin films from our sputtering chamber to exhibit lower damping than the films studied in previous chapters, thus allowing us to resolve complex skyrmion resonances in multilayer stacks. Moreover, films with improved uniformity are expected to exhibit lower critical currents for skyrmion motion.

FIGURE A.10: Various components of the sputtering set-up during assembly.

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Appendix B

Fitting Techniques for FMR Spectra

Fitting of Field Sweep FMR data

The following MATLAB function was used to fit single peak field sweep FMR spectra. This function models the magnitude of the resonance signal as a combination of symmetric and anti-symmetric Lorentzian functions [1, 2] superposed on a linear background. A constant has been included in the function to compensate the constant loss during the sweep [3]. A time-dependent drift term (written in terms of the time-dependent magnetic field) compensates the linear drift in the VNA stability during the measurement.

```
function y=S21_lorentz_amplitude(params,field)

H_resonance=params(1);
H_linewidth=0.5*params(2);
Amp_sym = params(3);
Amp_antisym = params(4);
BG = params(5);
Drift = params(6);
denominator=(field-H_resonance).^2+H_linewidth^2;

sym = H_linewidth^2./denominator;
antisym =(H_linewidth.*(field-H_resonance))../denominator;

y= Amp_sym*sym+Amp_antisym*antisym + BG + Drift*field;
```

We have extracted the resonance frequency and spectral linewidth from the fits, which are the only quantities of interest for our experiments. In case of a resonant spectra composed of a superposition of two or more signals, as observed for the

low frequency field sweep spectrum of Fe(0.5)Co(0.5) (chapter-4), we model the resonance response by two independent Lorentzian functions.

Fitting of Frequency Sweep FMR Data

The frequency sweep resonance spectra in previous chapters were fitted with a Dysonian function [4] in MATLAB. The following fitting function incorporates a Dysonian fit superposed on a linearly varying background.

```
function y= Dyson_fit(params,frequency)
Fre_res=params(1);
Fre_lw=0.5*params(2);
Amp=params(3);
Alpha = params(4);
BG = params(5);
Drift=params(6);
Denominator=(frequency-Fre_res).^2+Fre_lw.^2;
Numerator = Amp.*(Fre_lw + Alpha.*(frequency-Fre_res));
Lorentz_fn = Numerator./Denominator;
y= Lorentz_fn+BG+Drift*frequency;
```

FIGURE B.1: Fitting of resonance spectra using commercial PeakFitTM software. The experimental spectra were acquired at (a) 9.6 GHz, (b) 8.6 GHz and (c) 7.8 GHz during a field sweep FMR experiment performed on Fe(0.5)Co(0.5) at room temperature.

Manual Fitting of Resonance Spectra

Since our field sweep spectra often display more than one peak, we also explored the option of manually fitting the resonance data using the commercial PeakFitTM software package (figure B.1). The advantage of this method is that the software offers interactive manual control of all the fitting parameters. This is particularly useful when the background drift varies throughout the experiment and hence cannot be modelled by a single function. Another major advantage in comparison with our automated fitting methods is the ability to selectively discard noisy data, particularly at low frequencies below 5 GHz.

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