Optimization Of Magnetic Thin Films For Generating Stable Chiral Magnetic Structures

by

Alexandra Churikova

B.Sc., Massachusetts Institute of Technology (2016)

Submitted to the Department of Materials Science and Engineering in partial fulfillment of the requirements for the degree of Master of Science in Materials Science and Engineering at the MASSACHUSETTS INSTITUTE OF TECHNOLOGY

September 2018

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Signature redacted

Author ............. ............. Department of Materials Science and Engineering August 17, 2018

Signature redacted

Certified by ............. Geoffrey S. D. Beach

Professor of Materials Science and Engineering Thesis Supervisor

Signature redacted

Accepted by ............. Donald Sadoway

Chairman, Departmental Committee on Graduate Studies
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Abstract

The field of spintronics, or the use of electron spin for information processing, has revolutionized information storage. The long-term stability, low energy and low current in magnetic devices make them attractive for memory storage, sensors and computing. Their effectiveness is limited by the current, density, and device size, which can be optimized by tuning the magnetic materials properties. In this thesis, we show the effectiveness of ferromagnetic materials for the stability of magnetic “bits” numerically, and provide experimental insight into the engineering of material properties of ferrimagnetic and antiferromagnetic films.

Skyrmions are topologically protected chiral spin structures that are highly promising candidates for magnetic bits due to their stability and potential for fast motion. These spin structures can be used to encode 0’s and 1’s in racetrack memory devices. Chiral domain walls and skyrmions have been studied in magnetic thin films sandwiched between non-identical non-magnetic materials which have high spin-orbit coupling and Dzyaloshinskii-Moriya interaction (DMI). Parameters such as layer thicknesses and composition can be tuned for optimal skyrmion stability, speed, and size.

We use micromagnetic simulations to confirm experimental studies where skyrmions have been annihilated systematically with out-of-plane applied fields in the presence of in-plane fields. We show that in-plane magnetic field deforms the skyrmion and its domain wall, increasing the domain wall size and domain wall energy. This effect has been found to be greater in stray-field stabilized skyrmions with zero DMI. We subsequently image ultrasmall skyrmions (of diameters approaching 10 nm) in ferrimagnetic material with a compensated magnetic moment, using a pump-probe X-ray holography technique. Finally, we develop a recipe for synthetic antiferromagnetic materials with perpendicular magnetic anisotropy that have the potential to host skyrmions and detected with X-ray holography imaging.

Thesis Supervisor: Geoffrey S. D. Beach
Title: Professor of Materials Science and Engineering
Acknowledgments

Exploring the world of magnetic materials through fast-paced experiments and simulations has been an exciting journey for me. I’d like to take a moment to thank all of the people who have contributed to the work presented in this thesis.

First and foremost, I would like to thank my advisor, Geoff Beach, for constantly inspiring me to improve in every area of research and scientific work through his guidance and example. Thanks to you, I have learned so much about magnetism and also so much about the importance of building your own equipment, generating your own ideas, sharing knowledge effectively, and always asking the important questions.

I would also like to thank every member of the Beach Group, past and present, who I’ve had the pleasure to work with - thanks to all of your cheerfulness, guidance, and passion, I really cannot imagine myself anywhere else! In particular I would like to thank Felix Büttner for a multitude of stimulating discussions and for taking me on as his apprentice in the experimental arts. Thanks to Can Avci and Kohei Ueda for your extensive help and infinite wisdom in the spin-orbit torque measurements. Thank you to Lucas Caretta for showing me how to be an effective communicator of ideas, and for putting up with all my questions and how-do-I-do-this’s when I was just starting out in the lab (and now). Thank you Max Mann for never being too busy to explain something to me or teach me a measurement trick, and for your endless supply of funny phrases and hats. Thanks Ivan for helping me understand theory and encouraging me to take challenges. I want to thank Mantao Huang and Aik Jun Tan for always willing to help with anything and for always being in great spirits.

I would like to warmly thank David Bono for sharing his infinite knowledge of electronics and equipment design - you have guided me so much in the process of building and creating things without taking any short cuts. It is better to do it the long way, but to do it right. Thanks, David. Thank you Mike Tarkanian, Chris di Perna, and Tara Fadenrecht for teaching me how to use the machine shop, water jet, and more.

I would also like to thank our collaborators abroad who have helped make this
work possible. Thank you to Stefan Eisebitt and Dieter Engel for welcoming me to the Max Born Institute and trusting me with your lab, thanks to Sascha Petz for guiding me with your sputter system, and thanks to Bastian Pfau for your patience in helping me troubleshoot in the lab.

Thank you to Reinoud Lavrijsen and Marielle Meijer for welcoming me to your lab at TU/e and showing me how to work with your MOKE, VSM, and sputtering system. It was a blast.
# Contents

1 Introduction .................................................. 9

2 Background .................................................. 13
   2.1 Origin of Magnetic Order ................................. 13
      2.1.1 Magnetic energy components ...................... 15
      2.1.2 Magnetic Anisotropy .............................. 17
   2.2 Domain Walls ........................................... 19
   2.3 Magnetic Skyrmions .................................... 24
      2.3.1 Origin of chiral magnetic structures ............ 24
      2.3.2 Skyrmion generation and stability ............... 25
      2.3.3 Skyrmion collapse ................................ 27
   2.4 Micromagnetism ......................................... 30
      2.4.1 Micromagnetic Model Assumptions ................. 31
      2.4.2 Landau-Lifshitz-Gilbert Equation ............... 31

3 Methods .................................................... 37
   3.1 Thin film deposition .................................... 37
   3.2 Patternning of films .................................... 39
      3.2.1 Photolithography ................................. 39
      3.2.2 Electron Beam Lithography ....................... 39
   3.3 Magneto-optical characterization ..................... 41
   3.4 X-ray Holography Imaging Technique .................. 43
Chapter 1

Introduction

Magnetic data storage has been around since the 1950’s when IBM developed magnetic disk data storage, with a total capacity of five million characters. Since then, longitudinal recording media in the form of hard-drives has increased come into mass production and consumption. The hard drive combines mechanics and electromagnetism as the reading head reads consecutive bits of data arranged in a circular paths. While very cheap and high capacity, there are problems with bit density, weight, power consumption, and reliability. These issues can be solved with smaller solid-state devices with much faster read speeds and lower power consumption. Research into the transport of spin-up and spin-down (polarized) electrons in a material opened doors to faster and more efficient spintronic solid-state memory devices, such as Magnetic Random Access Memory (MRAM) in particular.

The Nobel Prize-winning discovery of the giant magneto-resistance effect (GMR) by Peter Grünberg and Albert Fert in the mid-1980’s revolutionized data storage. In the GMR effect, a material is able to increase its electrical resistance via a combination of magnetic and non-magnetic layers. This was followed by the development of aluminum oxide magnetic junctions (MTJs) in 1991 and in-plane magnetized magnesium MTJs at the beginning of the century [1]. Meanwhile came the advent of so-called racetrack memory (RM) devices, where information is coded in a magnetic ribbon by a train of up and down magnetic domains (regions of uniform magnetization) separated by domain walls. The train can be moved electrically by spin current.
torques to read or write information. However, these devices are still limited by high critical currents for DW motion, DW size, are very sensitive to external magnetic fields.

Skyrmion-based racetrack memory (RM), where small magnetic vortices called skyrmions store information as data bits, could be the solution to these performance and efficiency issues. It has been shown that these devices would require much lower current densities (approx. $10^6 \text{A} \text{s}^{-2}$) than those needed for domain-wall based RM (approx. $10^{12} \text{A} \text{s}^{-2}$) [2]. Magnetic skyrmions are topologically protected magnetization configurations [2], that have been observed experimentally in various magnetic materials, in bulk systems, thin magnetic films, and magnetic multilayers [3]. The topological nature of magnetic skyrmions is thought to give them enhanced stability and particle-like nature, which makes them suitable as robust information bits in future data storage devices [4]. In fact, a number of theoretical and numerical works have demonstrated that magnetic skyrmions could be essential components for future magnetic and spintronic devices. Most recently skyrmions have been experimentally shown to be stable at diameters as small as 10 nm in bulk ferrimagnetic films, making them attractive as data bits.

Both computational and experimental investigation into the fundamental properties of skyrmions has yielded much insight into the control and nucleation of skyrmions in nanoscale devices. These magnetic states have shown to have various origins, including most commonly the anisotropic exchange interaction, Dzyaloshinskii Moriya interaction (DMI) and stray fields ([5, 8]). DMI can manifest at heavy metal/ferromagnet interfaces, and induces a non-collinear order, leading to new magnetic states. The skyrmion structure arises from competing exchange interactions, occurring due to interfacial symmetry breaking occurring in multilayers [14]. This interaction stabilizes Néel walls (cycloidal rotation of the magnetization direction) with a fixed chirality over the Bloch walls (spiral rotation of the magnetization direction) [9]. The spin Hall effects from the heavy metals in this multilayer material system are due to strong spin-orbit interactions [15]. This leads to spin-orbit torques [60] that can be used to precisely and efficiently control magnetization dynamics.
Figure 1-1: An example of a skyrmion racetrack where skyrmions can be used as information “bits”. [5].

Experimentally, skyrmions have been created, or nucleated, in thin films by shrinking labyrinth domains with magnetic fields [5], as well as created at constriction sites by the injection of a spin current [6, 8]. Studies have shown that there is only a discrete transition between the skyrmionic and ferromagnetic (FM) state. Such annihilation can be modelled using micromagnetic or atomistic simulations. Atomistic simulations have shown that skyrmion collapse via an applied field occurs before any topological change, suggesting that topology plays a minor role in skyrmion stability [18]. Experimentally, skyrmions have been shown to collapse with the application of a magnetic field opposite to the skyrmion core magnetization direction. However, the stability of skyrmions under in-plane or tilted fields has seen limited investigations. Furthermore, ferromagnetic systems are limited in their potential to host ultrasmall magnetic skyrmions, so materials that allow favorable conditions (such as ferrimagnets and antiferromagnets) must be studied.

This thesis presents several investigations into the engineering of ferro-, ferri-, and antiferromagnetic materials for generating stable chiral structures such as skyrmions. In particular:

- In Chapter 2, we present the building blocks behind magnetic materials, with an introduction to domain wall theory and chiral magnetic structures, along with the basics of magnetism and the fundamental energetics that are necessary to consider when investigating magnetic states.
In Chapter 3, we introduce the methods and techniques used to image magnetic textures and characterize magnetic properties.

In Chapter 4, we use micromagnetic simulations to probe the stability of ferromagnetic skyrmions under in-plane and out-of-plane fields. We show that in-plane fields have the effect of domain wall broadening and deformation, and domain wall energy increase for magnetic skyrmions, and use this to explain previous experimental observations.

In Chapter 5, we use the X-ray holography technique to image magnetic skyrmions in a ferrimagnetic material, and determine certain characteristics of skyrmion stability and size in these materials.

In Chapter 6, we optimize the properties of SAF materials with both magneto-optical and electrical measurements, and image magnetic domains in synthetic antiferromagnetic (SAF) material.
Chapter 2

Background

2.1 Origin of Magnetic Order

Magnetic properties of materials arise from the dynamic behavior of quantized magnetic moments. In most materials, magnetic moments are randomly oriented without an external bias field, and the net magnetization is zero (paramagnetic behavior). The origin of magnetism lies in the intrinsic spin-orbit coupling between spin and orbital momentum of individual electrons. The angular momentum and electron spin have separate quantum numbers, but they are coupled by the spin-orbit interaction. The angular orientation of the electron spin is determined by the shape of the electron’s orbital function. The magnitude of this interaction increases with the strength of the electric field. Because two electrons with the same spin of $\frac{1}{2}$ or $-\frac{1}{2}$ are not able to occupy the same energy state due to the Pauli exclusion principle, electrostatic repulsion between two electrons is reduced. This exchange interaction arises from the Coulomb interaction between electrons and the spin symmetry. That is, aligning spins in the same direction is energetically unfavorable because of the large magnetostatic repulsion, but electrons can save energy by forming bonds. The trade-off between these energy terms determines the nature of the long range order in a material. In a lattice of spins, the long-range order will be determined by the Heisenberg exchange
interaction, written as the Hamiltonian:

\[ \hat{H} = - \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j \]  \hspace{2cm} (2.1)

where \( J_{ij} \) is the exchange constant between the \( i \)th and \( j \)th spins.

In transition metal monoxides as well as spinel and garnet structures, the long-range order is governed by the superexchange interaction, resulting from a difference in symmetry of \( p \) and \( d \) orbitals. Due to this interaction in a metal oxide (consisting of an oxygen ion and two metal ions), the \( p \) orbital of the oxygen has an antisymmetric form that maintains the metal \( d \) moments in opposite configuration. This interaction leads to the "antiferromagnetic" structure where moments are aligned antiparallel to one another. The attractiveness of this type of material lie in the effect of its net zero angular moment and magnetization, which leads to fast magnetization dynamics.

![Figure 2-1: Schematic alignment preference of magnetic moments as a result of ferromagnetism, antiferromagnetism, and ferrimagnetism.](image)

In turn, a "ferrimagnetic" material has a crystal structure where antiparallel ions either have a different valence number, such a spinel crystal, or a different atomic number. Two metals of different atomic with a non-zero magnetization form antiparallel sublattices. Each sublattice may have a different temperature dependence and Landau \( g \) factor. This leads to the sublattices having different temperature where net angular momentum is zero (\( T_A \)) and temperature where net magnetization (\( T_M \)) is zero, critical values that are called angular momentum and magnetization compensation temperatures, respectively. At \( T_A \), a ferrimagnet displays the dynamical behavior of an antiferromagnet, and can still interact with magnetic fields and spin current. Meanwhile, the small non-zero magnetic moment occurring near \( T_M \) can potentially stabilize chiral domain walls, as will be discussed in Chapter 5.
Meanwhile, magnetic moments in ferromagnetic materials such as Fe, Co, and Ni exhibit a parallel alignment (spontaneous magnetization) without an externally applied field, causing a nonzero net magnetization. The exchange constants for nearest neighbors are positive to ensure parallel alignment. In transition metals, ferromagnetic behavior can be described by employing the density of states rigid band model. The energy term that prefers parallel spin alignment is the exchange interaction. In this model, spin-up and spin-down electron $s$ and $d$ states are filled from the lowest energy according to the number of electrons per nucleus. The exchange interaction prefers parallel spin alignment. If the exchange energy penalty is greater than the energy of moving to the next higher energy state, then the band in question gets filled. A net spin polarization arises when the two bands have an unequal number of electrons. The Stoner Criterion is a measure for the threshold for this spontaneous magnetization.

### 2.1.1 Magnetic energy components

The magnetic domains in thin films can take different forms, which are determined by factors such as thickness of the film, applied field, and strain. The total energy, disregarding magnetoelastic effects, can be written as:

\[
E_{\text{tot}} = \int \left[ \mathcal{E}_{\text{ex}}(m) + \mathcal{E}_{\text{an}}(m) - M_s H_{\text{ex}} m - \frac{1}{2} M H_d \right] dV
\]

(2.2)

where $M_s$ is the saturation magnetization and $m(r) = M(r)/M_s$ is the magnetization unit vector that aligns in the direction of the local polarization $M$.

**Exchange**

The exchange interaction of neighboring spins that comes from the Heisenberg formulation is used to describe the exchange energy of two adjacent ferromagnetic moments. The exchange energy density of two neighboring electron spins $\mathbf{S}_i$ and $\mathbf{S}_j$ is:

\[
E_{\text{ex}_{i,j}} = -J_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j = -J_{i,j} S^2 \cos \phi_{i,j},
\]

(2.3)
in which $J_{i,j}$ is the exchange term which determines the interaction strength and the degree of spin alignment, while $\phi_{i,j}$ is the angle between the two spins, and $S$ is the spin magnitude. This is a short-range interaction since it originates from wavefunction overlap. Thus, summing over all neighboring electrons, the exchange energy due to a single electron is:

$$E_{ex} = -S^2 \sum_i J_i \cos \phi_i,$$

where $\phi$ is the angle between adjacent pairs of electrons $i$. The corresponding energy density is:

$$\mathcal{E}_{ex} = A(\nabla \mathbf{m})^2 = A \left( \frac{\partial \theta}{\partial x} \right)^2,$$

where $\theta$ is the angle of tilt between adjacent spins, $x$ is the distance between the adjacent spins, and $A$ is the exchange stiffness that depends on the exchange constant and solid structure.

**Zeeman**

The overall energy can be impacted heavily by an external applied field, the Zeeman energy. It describes the interaction with an external field $\mathbf{H}_{ex}$ and depends only on the average magnetization:

$$\mathcal{E}_H = -M_s \int \mathbf{H}_{ex} \cdot \mathbf{m} dV$$

The magnetization tends to align parallel to the Zeeman field in order to minimize the energy of the system. The Zeeman energy thus allows the manipulation of magnetization via applied field, and is often used to switch the magnetization or to move domain walls.

**Magnetostatic energy**

The free magnetic poles at the surface of the material (or the divergence of the magnetization $\nabla \times \mathbf{M}$) induce a stray field (also known as the demagnetizing field $H_d$) which increases the total energy of the material and opposes the orientation of
magnetic moments in a given direction. This shape anisotropy, referred to as the magnetostatic energy, is the energy per unit volume due to the demagnetizing field:

\[ \mathcal{E}_d = -\frac{1}{2} M \cdot H_d. \]  

It is energetically favorable for magnetic moments to align along the longest axis of a sample, so as to minimize free poles and stray fields. Magnetic moments tend to lie in the plane of a magnetic thin film due to magnetostatics.

### 2.1.2 Magnetic Anisotropy

Magnetic anisotropy is commonly derived from the atomic-scale structure of the sample, such as crystal structure and bonding type. This direct connection between structure and magnetization originates in the spin-orbit interaction, or the weak coupling between the spin and angular momenta which contributes to an effective field in the material depending on the orbital profile of the solid. Magnetic moments in a solid may preferentially orient along a certain crystallographic axis, such as \((111)\) in FCC Co gaining magnetocrystalline anisotropy. The energy per unit volume in the simplest case with one easy axis (uniaxial anisotropy) is:

\[ \mathcal{E}_{un} = K \sin^2 \theta, \]  

where \(\theta\) is the angle between the magnetization vector and the easy axis. In general, anisotropy is large in crystals with low symmetry.

Magnetic materials with strong out-of-plane magnetization, i.e. perpendicular magnetic anisotropy (PMA), offer superior qualities compared to the in-plane anisotropy materials for hard disk drive and magnetoresistive random access memory (MRAM) devices and have been successfully commercialized in the last decade.

Magnetostatic anisotropy energy always prefers an in-plane magnetization axis in the conventional magnetic thin film theory. When magnetic moments point in the direction normal to the film plane, a large demagnetizing field is generated, which
causes an unfavorable increase in total energy of the system. This explains the preference of material for in-plane magnetization orientation. However, out-of-plane (or perpendicularly) magnetized films can be created with the introduction of new physics, i.e. spin-orbit interactions. For example, perpendicular magnetization is produced when a thin layer of metallic material (such as Pt or Pd) between a non-magnetic material with large spin-orbit coupling (e.g. an oxide, such as MgO) due to strong interfacial interactions. The degree of perpendicular magnetization can be understood with the uniaxial anisotropy constant of a material, $K_u$, which is defined by:

$$K_u = K_V + \frac{K_S}{t}$$  \hspace{1cm} (2.9)

where $K_V$ and $K_S$ are the volume and surface anisotropy constants, respectively, and $t$ is the film thickness. The $K_V$, or volume anisotropy energy density, originates from the magnetocrystalline, magnetostatic, and magnetoelastic contributions (resulting in a negative energy contribution). Meanwhile, the $K_S$, or surface anisotropy per unit area, derives from spin-orbit interaction at the interface (a positive surface contribution). Thus the anisotropy constant is a result of the system’s tendency to minimize magnetostatic energy competing with the energy minimization via spin-orbit coupling. Net anisotropy must change sign from negative to positive as the film thickness decreases (and the film is perpendicularly magnetized) [36]. Meanwhile the effective interface anisotropy $K_{eff}$ as used in this thesis is:

$$K_{eff} = K_u - \frac{\mu_0 M_s}{2}$$  \hspace{1cm} (2.10)

where $\mu_0$ is the permeability of free space and $M_s$ is the saturation magnetization. The perpendicular anisotropy of a thin film can be identified with a hysteresis loop obtained for example from a magneto-optical Kerr effect measurement (see Section 4). The alignment of magnetization with the hard or easy axis (Fig. 2-2) as an external field is applied will indicate the anisotropy of the film.
Figure 2-2: Demonstration of perpendicular magnetic anisotropy in out-of-plane magnetized thin films when an external magnetic field is applied.

2.2 Domain Walls

Ferromagnetic materials have a spontaneous nonzero magnetization in the absence of field. However, magnetization is not uniformly oriented everywhere in a macroscopic section of a ferromagnetic sample. When a ferromagnetic material is demagnetized such that its net magnetization is small or zero, there are regions of varying magnetization called magnetic domains, where the local magnetization reaches the saturation value. Adjacent domains are separated by boundaries called domain walls, and are classified by the angle between magnetization in neighboring domains. In general, a material would want to break into domains in order to minimize the magnetostatic energies associated with the “free poles” (Fig. 2-3). At the same time, it “costs” energy to form new domains, so the periodicity of a magnetic texture is a direct result of the competition between stray field and domain wall energy terms.

The magnetic moments within the finite width of a domain wall (DW) rotate gradually to provide a continuous switch from the magnetization of one domain to another. A 180° DW separates domains of opposite magnetization (a type of DW called the Bloch wall), while a 90° DW separates domains of perpendicular magnetization (called the Néel wall). In a Bloch wall, the azimuthal angle, defined by the angle between the magnetic moment and the plane of the domain wall, is \( \phi = 0, \pm \pi \)
during the rotation of magnetization. In contrast, the Néel wall is at an azimuthal angle of $\phi = \pm \pi/2$ and the magnetization rotates perpendicular to the wall plane. The analytical description of the DW profile then can be found.

**Domain wall energy density**

The formation of a DW has an energy cost due to the moments within the DW not aligning perfectly parallel with respect to each other (exchange energy penalty) and deviating from the easy axis (anisotropy energy penalty). Shape anisotropy must also be taken into account since the DWs are in thin films. For a homogenous domain wall in the yz-plane and azimuthal angle $\phi$, the energy density per unit area can be found by integrating the energy terms along a path through the wall. For instance,
in the case of the Bloch wall, the energy density for an infinitely extended thin film is:

\[ E_{DW} = \int \left( A \left( \frac{\partial \theta}{\partial x} \right)^2 + K_{eff} \sin^2 \theta \right) dx, \]  

(2.11)

where the first integrated term is the exchange term and the second is the uniaxial anisotropy energy per volume. Considering the azimuthal angle \( \phi \) to be constant, the exchange energy density can also be written as:

\[ A \left( \nabla \frac{M}{M_s} \right)^2 = A(\nabla \theta)^2 + A \sin^2 \theta(\nabla \phi)^2 = A(\nabla \phi)^2 \]  

(2.12)

The second term corresponds to the effective anisotropy energy with magnetocrystalline and shape anisotropy contributions. To formulate the equilibrium condition, the energy density functional is minimized with respect to the polar angle \( \theta \) of the magnetization, yielding:

\[ 2A \left( \frac{\partial^2 \theta}{\partial x^2} \right) = K_{eff} \sin \theta \cos \theta \]  

(2.13)

which, after integration, produces:

\[ A \left( \frac{\partial \theta}{\partial x} \right)^2 = K_{eff} \sin^2 \theta \]  

(2.14)

Therefore, the contributions of the exchange and anisotropy energy to the domain wall are equivalent at energetic equilibrium. The magnetization equilibrates between fast rotation (high exchange energy) and slow rotation (high anisotropy energy) to reach an equilibrium DW width which depends on material parameters. Solving Eq. 2.13, the dependency of the magnetization angle \( \theta \) on position along the Bloch wall can be derived:

\[ dx = \sqrt{\frac{A}{K_{eff}}} \sin^{-1} \theta d\theta \]  

(2.15)
The energy density per unit area of the domain wall can then be found by substituting
Eqn. 2.15 and Eqn. 2.14 into Eqn. 2.11,

\[ \sigma_{DW} = 2\sqrt{AK_{eff}} \int_0^\pi \sin \theta d\theta = 4\sqrt{AK_{eff}} \]  \hspace{1cm} (2.16)

The energy density \( \sigma_{DW} \) accounts for both Bloch and Neel wall since it has no de-
pendency on the azimuthal angle.

**Domain wall profile and width**

The DW profile is the characterization of the magnetization angle \( \theta \) as a function
of the position \( x \) inside the domain wall. For a 180° domain wall, Eq. 2.15 can be
integrated with the substitution \( \theta = \theta' + \pi/2 \) to:

\[ \int dx = \sqrt{\frac{A}{K_{eff}}} \int \frac{1}{\sin \theta} d\theta = \sqrt{\frac{A}{K_{eff}}} \int \frac{1}{\cos \theta'} d\theta' \]  \hspace{1cm} (2.17)

Substituting \( \theta \) back in the expression results in the variation of the DW angle as a
function of \( x \):

\[ \theta(x) = \arcsin \left( \tanh \left( \frac{x}{\sqrt{A/K_{eff}}} \right) \right) + \frac{\pi}{2} \]  \hspace{1cm} (2.18)

**Domain wall pinning**

Two models of the pinning of domain walls by microstructural defects have been pro-
posed. The rigid-wall motion model, most applicable to the topologically protected
domain walls in the skyrmion, considers fluctuations of potential energy with posi-
tion. This model has been widely used as a qualitative description of wall energetics
and dynamics [30]. The rigid-wall model considers an inflexible wall whose motion
is activated and restricted by statistical fluctuations of defect density, which modify
the local potential energy landscape. With a uniform distribution of defects on either
side of the wall, the net pinning force on the wall is zero. Otherwise, a net force tends
to move the wall to a more energetically favorable position.
Figure 2-5: Variation of the domain wall energy landscape (depinning field) as a function of position, with energy minima occurring at crystalline defects (pinning sites). Arrows show the progress of a domain wall as the applied “depinning” field increases, figure adapted from [30].

The equilibrium magnetization distribution in any magnetic field is expected to be determined by the minimum energy configuration, when all the contributing energies are taken into account. In a perfectly uniform material the DW energy is the same everywhere and the position and the distance between the DWs is uniform. In real material, material properties differ from point to point, so the the magnetic constants $A$ and $K$ are functions of the position $r$: $A(r)$ and $K(r)$. Thus, DW energy is locally lowered and extra energy is needed to change the magnetization. DWs are therefore pinned by localized crystalline defects at energy minima and can only be released by a corresponding energy input, e.g. by an applied depinning field (Fig. 2-5). Thus the energy of the applied field must overcome the energy of the interaction of the domain wall with the defect sites.
2.3 Magnetic Skyrmions

2.3.1 Origin of chiral magnetic structures

Magnetic spins align parallel to each other to reduce the exchange energy, as shown in Section 2.1. In materials with broken inversion symmetry in the lattice or at the interfaces, an exchange interaction term called the Dzyaloshinskii-Moriya Interaction (DMI) can be present, giving rise to chiral magnetic configurations in some films (Fig. 2.6). DMI originates from large spin orbit coupling, with a Hamiltonian of neighboring spins \( S_i \) and \( S_j \) given by:

\[
H_{DM} = - \sum_{i,j} D_{i,j} \cdot [S_i \times S_j]
\]

(2.19)

where \( D_{i,j} \) is the Dzyaloshinskii-Moriya tensor and is dictated by crystal symmetry. Unlike the exchange interaction, DMI favors a perpendicular magnetization in order to minimize the cross product (with spins normal to \( D_{i,j} \) but perpendicular to each other). In systems governed by Eqn. 2.15, there is no uniaxial symmetry but a directional non-collinear magnetic order of a certain chirality (right-handed or left-handed depending on the sign of \( D_{ij} \)). The DMI and Heisenberg exchange interactions are in competition to determine the long-range magnetization behavior, and chiral magnetic structures can arise when the DMI is stronger than the Heisenberg exchange interaction. Meanwhile the strength of DMI scales with the strength of spin-orbit coupling which in first approximation should scale with \( Z^4 \) (\( Z \) being the atomic number) - for transition metals, this is based on band structure [37]. Thus DMI is usually high in materials with a heavy element layer, which contributes a large spin orbit coupling.

Topologically protected structures called magnetic skyrmions are small particle-like domains in an out-of-plane magnetized film. As shown in Fig. 2-7, the spins inside a skyrmion rotate continuously with a fixed chirality from the up direction at one edge to the down direction at the center, and then to the up direction again at the other edge. They can be found in various systems, and stabilized by either stray field
energies or DMI [26]. In bulk structures, magnetic skyrmions have been observed in Fe/Mn [38], MnSi [39], FeGe [41], Fe$_{1-x}$Co$_x$Si [40] most recently ferrimagnets GdCo and GdFeCo [34], and others. Room temperature magnetic skyrmions have been created in layer structures with CoFeB as the ferromagnetic layer [?], with either spin-orbit torque fields [8, 31] or field-induced nucleation by shrinking a multidomain state [5].

The two types of magnetic skyrmion configurations, the Néel-type and Bloch-type chiralities (Fig. 2-7), can arise from different interaction symmetries between spins (due to bulk or interfacial effects, for instance). This results in different directions of domain wall rotation. Skyrmions can be defined by the topological number $S$ (or skyrmion number), which is a measure of the winding of the normalized local magnetization, $m$. In the two-dimensional limit, the topological number is:

$$S = \int \left( \frac{\partial \hat{n}}{\partial x} \times \frac{\partial \hat{n}}{\partial y} \right) \cdot \hat{n} \, dx \, dy \quad (2.20)$$

where $n(x, y)$ is the topological density computed as $m \cdot (\partial_x m \times \partial_y m)$, with $m$ being the vector field (magnetization for ferromagnetic materials) [19].
2.3.2 Skyrmion generation and stability

 Historically, magnetic skyrmion bubbles have been shown to exist in materials with a certain value of anisotropy quality factor $Q = 2K_u/\mu_0 M_s^2 > 1$ and in the presence of external magnetic fields. Even when $Q > 1$, the energetics may favor a stripe domain structure to minimize the total energy in a PMA material. Applying an out-of-plane external field can transform the domains into isolated magnetic bubbles, and vice versa. If external field $H$ is decreased, the film will tend to increase the volume fraction of the stripe domains and reduce the Zeeman energy. If $H$ is increased, the energetics favor the magnetization of the ferromagnetic ground state, so above a critical field the skyrmion bubbles will collapse. Thus, a perpendicular field can be used as a stabilization mechanism for the skyrmion bubble at room temperature [?].

 In early numerical and theoretical treatments of skyrmion stability in a ferromagnetic material with DMI, skyrmion stability at zero field when $D$ is larger than a critical value $D_c = 4\sqrt{AK}/\pi$ was discussed in limited detail and applied only to the case of zero field skyrmions [24]. A more complete model, based on the universal 360° wall model for skyrmion spin structure [21], developed by Büttner et al. for the stability of an isolated skyrmion in any given infinite film relies on full compliance between a theoretical phase diagram in a multi-parameter space, numerical calculations, and experimental data [26]. This theory shows a sharp transition between the DMI and stray field stabilized skyrmion states, including states where the skyrmion bubble is stabilized by both energy terms [62]. The most physically relevant prediction

Figure 2-7: Schematic of (a) Néel and (b) Bloch skyrmion with cross section shown [23].
is that Co-based ferromagnetic multilayers, which have been extensively studied in experiments, are not suitable for hosting sub-10 nm skyrmions at room temperature, which is the property needed for using skyrmions as data storage bits. The model is based on the calculation of an equilibrium skyrmion energy functional for a given set of material parameters \((Ku, Ms, A, \text{interface and bulk DMI constants, magnetic layer thickness } d)\) and external out-of-plane field.

2.3.3 Skyrmion collapse

The path between the nucleation and annihilation of a skyrmion in an unstable configuration is of particular interest for their implementation in room-temperature devices. Furthermore, understanding the phase space of material parameter values which are required for skyrmion stability helps build efficient and robust skyrmion-based devices. This section will examine the key relevant investigations into the stability and collapse to date, both experimental and theoretical.

Simulations

The stability and collapse of magnetic skyrmions has been studied both theoretically and numerically (with \(ab\text{-initio}\) and micromagnetic simulations). Direct calculation of energy barriers to collapse of a magnetic skyrmion has been carried out micromagnetically in vortex core switching in zero-field and finite applied field configurations. The core reversal field was found to always increase and improve in accuracy upon refinement of mesh size, but the method still proves to be applicable for relative calculation for different film sizes. In the energy and path calculations, finite element micromagnetic code was used to vary the mesh size accordingly as the skyrmion size changes [?].

Néel skyrmion collapse has been investigated by analyzing the energetic collapse path with \(ab\text{-initio}\) calculations and Langévin dynamics. Lowest energy barrier to collapse was found under an out-of-plane destabilizing field, and collapse was shown to occur before any topology change, suggesting that topology plays a minor role in
skyrmion stability [18].

In the theoretical model described earlier, the path from the skyrmion state at an equilibrium radius to the ferromagnetic ground state is through a singular $R = 0$ state [62]. At the point of collapse, all energy terms vanish except for exchange energy, from which the finite energy for any isolated skyrmion at collapse ($R = 0$) is $E_0 = 27.3Ad$. The implication of this model is that all skyrmions are prone to collapse, and skyrmion stability is related to the annihilation energy barrier $E_a = E_0 - E(R)$ which depends on the micromagnetic parameters and not the topology. The size of this collapse barrier at a given temperature indicates the thermal stability of a skyrmion. It is important to note that skyrmions can be deformed in a way not possible to be explained by the 360° wall model, which reduces the energy barrier [18].

Experiment

This section will focus on several phenomena observed in a Pt(3 nm)/Co(0.9 nm)/Gd(1 nm)/GdOx(30 nm) stack under room temperature and the applications of external fields, carried out by L. Caretta. Wide-field Kerr microscopy was used to probe the out-of-plane magnetic contrast of the film surface, using the polar MOKE technique where the in-plane components of the magnetization cancel out. The thickness of the Gd layer was chosen to tune the perpendicular anisotropy field $H_k$. When the ratio of the DMI effective field $H_{DMI} = \frac{D}{\mu_0 M_s \Delta}$ (where $\Delta = \sqrt{A/K_{eff}}$) to the perpendicular anisotropy field $H_k$ is large, bubble domains consisting of coupled 360 degree domain walls can be nucleated [25]. After minimization of the domain wall energy density,

$$\sigma_{DW} = 4\sqrt{AK_u} - \pi|D|,$$

the threshold for skyrmion formation is when

$$\frac{H_{DMI}}{H_k} > \frac{2}{\pi}.$$
The experiment was carried out by first nucleating bubbles with an out-of-plane (OOP) field $H_z$, with a direction opposite to the magnetization of the skyrmion core. The bubbles were then shrunk with an increasing OOP field of this orientation until all of the bubbles were too small for the resolution limit. After the application of an in-plane (IP) field $H_x$, the bubbles were expanded again with an application of an out-of-plane field applied in the opposite direction. The final states with multiple skyrmion bubbles after shrinking and expansion, for varying IP fields, are shown in Fig. 2-8. At each in-plane field, the number of bubbles that collapsed at different out-of-plane fields was recorded, and distributions were constructed 2-9.

![Figure 2-8: MOKE image of skyrmion bubble states after applying an out-of-plane field to expand the remaining skyrmions in the presence of indicated in-plane fields $H_x$ (0 Oe, 65 Oe, and 80 Oe) Adapted from [25].](image)

![Figure 2-9: Distribution of the fraction of annihilated skyrmions as successive OOP fields $H_z$ were applied at three different IP fields $H_x$ [25].](image)

The collapse field skyrmion bubbles was found to be a strong function of the applied in-plane field. That is, increasing the in-plane field reduced the annihilation threshold of the skyrmions (Fig 2-10,a). The skyrmion bubble annihilation field
became deterministic at in-plane fields near the DMI effective field, and the full width half maximum of the distribution of collapse skyrmion decreased consistently with higher in-plane fields (Fig. 2-10,b).

Figure 2-10: (a) Average annihilation (collapse) field for the skyrmions as a function of in-plane field. (b) The distribution width of collapse out-of-plane fields as a function of in-plane field.

The wide distribution of collapse fields for low in-plane fields in Fig. 2-9 is attributed to the skyrmions collapsing mostly due to thermal Arrhenius activation processes. At higher in-plane fields, it appears thermally activated collapse is no longer as prevalent and the distribution of remaining skyrmions with applied out-of-plane fields is sharper. Chapter 4 of this thesis will focus on using micromagnetic simulations of collapse to explain this deterministic collapse at high in-plane fields, as well as the decrease in average collapse field at high in-plane fields.

2.4 Micromagnetism

Micromagnetism is the continuum theory of magnetic moments widely used to analyze magnetic microstructure. The theory of Landau and Lifshitz is based on a variational principle: magnetization profiles are calculated based on energy minimization schemes. The variational principle is used to develop a set of differential equations called micromagnetic equations, first formulated by Landau and Lifshitz in
one dimension. In the following years, the analysis was extended to three dimensions and accounting for stray field effects. Micromagnetic theory is an integral analysis for the magnetic behavior of small particles, calculations of internal structures of domain walls, the description of magnetization dynamics and calculation of magnetic stability limit, and will be used for the numerical study in Chapter 4.

This complex quantum mechanical approach to describing a ferromagnetic material is simplified based on the length scale that describes a given magnetic phenomena. The micromagnetic theory is commonly used on the submicron length scale where the continuum approximation of micromagnetism yields accurate physics. In contrast, atomistic simulations consider the discrete, quantum mechanical description of spin structure and have a length scale of $< 1$ nm.

### 2.4.1 Micromagnetic Model Assumptions

First we describe the continuum assumptions made in the micromagnetic model. In the micromagnetic model of a ferromagnet, the adjacent moments $\mathbf{m}_i$ at positions $\mathbf{r}_i$ are assumed to align almost parallel with no discontinuities. The second assumption is the homogeneous density of electrons, which implies a continuous distribution of magnetic moments $\mathbf{m}_i$ as a vector $\mathbf{M}(\mathbf{r})$ and a constant saturation magnetization $M_s$. Discrete effects are thus ignored and all other magnetization vectors are continuous in distribution. For example, for a volume $V$ the exchange interaction assumes

$$\int_V \mathbf{M}(\mathbf{r})d\mathbf{r} \approx \sum_i m_i(\mathbf{r}_i)$$  \hspace{1cm} (2.23)

Since the electron density is homogeneous, we can express the magnetization vector modulus as a constant $M_s$ with a reduced magnetization vector $\mathbf{m}$:

$$\mathbf{M} = M_s \cdot \mathbf{m} \quad \text{and} \quad |\mathbf{m}| = 1$$  \hspace{1cm} (2.24)

Unlike in discrete approaches such as atomistic models, the energy minimization procedure depends on the initial conditions and the final state.
2.4.2 Landau-Lifshitz-Gilbert Equation

The Landau-Lifshitz-Gilbert (LLG) equation used in the numerical simulations in this thesis is derived from quantum mechanics. A dimensionless intrinsic damping factor in the LLG Equation is introduced.

Derivation of the LLG Equation for Micromagnetism

Since the continuum micromagnetic approximation stems from the consideration of discrete spins in space and position, we will first treat a single spin with the operator \( \hat{S} \). We can use Heisenberg’s equations to describe the time development of the spin operator \( \hat{S} \). Without energy dissipation, the eigenvalue of the Hamiltonian \( \hat{H} = \mu_a \cdot \mathbf{H} \) is conserved, where the atomic magnetic moment is \( \mu_a = -g\mu_B \mathbf{S} \) (\( \mu_B \) is the Bohr magneton, \( g \) is the g-factor). Heisenberg’s equations can be used to describe the motion of spin \( S \) [28]:

\[
\frac{d\hat{S}_i}{dt} = \frac{1}{i\hbar} \left[ \hat{S}_i, \hat{H} \right] = \frac{g\mu_B}{i\hbar} [S_i, S_j] H_j = \frac{g\mu_B}{\hbar} \epsilon_{ijk} S_k H_j \tag{2.25}
\]

where the commutator of the spin is \([S_i, S_j] = i\epsilon_{ijk} S_k\). Inserting this commutator into Eq. 2.25 yields

\[
\frac{d\hat{S}}{dt} = \hat{S} \times \frac{\partial \hat{H}}{\partial \hat{S}} \tag{2.26}
\]

Using the Ehrenfest theorem, the operators are replaced by their expectation values and the spin is expressed in terms of macroscopic magnetic moment:

\[
\mathbf{M} = -\gamma' \langle \hat{S} \rangle \tag{2.27}
\]

where \( \gamma' = g\mu_B/\hbar \), \( g \) is the Landé factor and \( \mu_B \) is the Bohr magneton. With the expectation value of the Hamiltonian equal to the energy \( E \) of the system, we can re-write Eqn. 2.26 as:

\[
\frac{d\mathbf{M}}{dt} = \gamma' \mathbf{M} \times \frac{\partial E}{\partial \mathbf{M}} \tag{2.28}
\]

In the classical limit, the partial differentiation is replaced by the variation derivative.
δE/δM. A change of the energy $E$ of the system as a result of the change of the magnetic moment $M$ is due to the existence of an effective field, since $δE = µ_0 δM \cdot H_{\text{eff}}$. Then the effective field is

$$H_{\text{eff}} = -\frac{1}{µ_0} \frac{δE}{δM} \quad (2.29)$$

Using this expression in Eqn. 2.28, the equation of motion of the magnetic moment, or $M = nµ$ for a ferromagnet with $n$ the atomic density, can be expressed by the Landau-Lifshitz (LL) equation:

$$\frac{dM}{dt} = -ngµ_B \frac{dS}{dt} = -γ_0(M \times H) \quad (2.30)$$

where $γ_0 = gµ_0µ_B/ℏ$ is the gyromagnetic ratio for an atomic spin. This equation describes the magnetization's precession around the effective field $H_{\text{eff}}$ without any energy loss. The right-hand term of Eqn. 2.30 is thus the precessional term. However, due to dissipative effects, the magnetic moments relax in the direction of the effective field $H_{\text{eff}}$.

**Damping**

With the inclusion of dissipation energy, the electron spins gain non-equilibrium statistics and the motion is nonlinear. A phenomenological damping term was added by Landau and Lifshitz to account for this, yielding the LLG equation:

$$\frac{dM}{dt} = -γ(M \times H) - γ \frac{α}{M} M × (M × H_{\text{eff}}) \quad (2.31)$$

where the dimensionless constant $α$ is the Landau damping constant and refers to the energy dissipation in the material, analogous to a frictional coefficient. The damping term was explained by Thomas Gilbert using a Lagrangian formulation and treatment of the LL equation with a dissipation function [28]. The damping term is $α = γ_0 η M$ where $η$ is a "friction" coefficient, while it can be shown that the new gyromagnetic
This factor is larger than that in the LL equation ($\gamma_0$) by a small factor due to damping coefficient $\alpha$.

Figure 2-11: Schematic of temporal evolution of single magnetic moment $\mathbf{M}$ in an effective field $\mathbf{H}_{\text{eff}}$, which shows (a) precessional trajectory of $\mathbf{M}$ around $\mathbf{H}_{\text{eff}}$ and (b) magnetic moment relaxation in a direction parallel to the effective field (non-zero damping).

**Energy relaxation**

To find the way the energy $E$ of the magnetic system behaves over time, we use the variational formulation once again to express the energy density $U$:

$$\frac{dU}{dt} = \frac{\delta E}{\delta \mathbf{M}} \cdot \frac{d\mathbf{M}}{dt} = -\mu_0 \mathbf{H}_{\text{eff}} \cdot \mathbf{M}$$  \hspace{1cm} (2.33)

Using the LLG equation in Eqn. 2.33, and taking the volume integral, the temporal evolution of energy is:

$$\frac{dE}{dt} = -\mu_0 \int_V \mathbf{H}_{\text{eff}} \cdot (-\gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} - \alpha \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{\text{eff}})) \, dr$$  \hspace{1cm} (2.34)

This integral is always negative or zero, and always decreases over time. In a system with non-zero damping ($\alpha > 0$) and dissipation the system energy decreases to an
energy minimum. This precession of magnetic moments and minimization of energy toward the relaxed state is energy relaxation.

The dynamical behavior of the magnetization in a ferromagnetic film can be found with the LLG equation. With the precession and damping term depending on the effective field $H_{\text{eff}}$, the time evolution of $M$ must also be calculated. Changing the magnetization $M$ also changes the energy of the system $E$. Thus, the equilibration of magnetization in an effective field always coincides with an energy minimization.
Chapter 3

Methods

We start this chapter by describing the way we fabricate thin films and the considerations that need to be taken into account regarding favorable material properties for skyrmion generation and motion in these films.

3.1 Thin film deposition

The substrate used for our samples is a commercially available Si substrate. The surface must be as clean as possible due to the sensitivity of magnetic properties due to film quality and uniformity. The samples were cleaned accordingly, following the standard procedure of sonication in a bath of (i) acetone (ii) isopropanol, and blown dry with \( \text{N}_2 \) to ensure that any residue from isopropanol does not remain on the surface. The samples were then fixed on a holder with copper tape and loaded into the deposition chamber.

The ferrimagnetic, ferromagnetic, and synthetic antiferromagnetic (SAF) films discussed in the next sections are deposited using d.c. magnetron sputtering. Sputtering is a widely used growth technique due to efficiency, high growth rates, and precise layer thickness control down to the sub-nanometer range. The deposition also allows for an alloy or compound material deposition from several different targets. A d.c. sputtering set-up is shown in Fig. 3-1. The target material was placed in a pure Argon atmosphere of 3 mTorr. The large potential difference applied between the
target material and a cathode ionizes the Ar gas. The target material is bombarded by highly energized Ar ions, and target atoms are ejected onto the substrate. The magnet below the target enhances the ionization of the noble gas by increasing the mean free path of the ions and electrons in the plasma, and thus increasing the sputtering rate and allowing the Ar plasma to be stable at lower pressure. Continuous rotation of the sample stage allows for material uniformity, and the shutters are used to expose the material to the target atom bombardment.

Some oxides, e.g. MgO, are deposited by radio frequency (RF) sputtering. A rotating sample stage increases the uniformity of the film thickness. The sputtering system is equipped with four sputtering guns so that a structure consists of maximum four different materials can be deposited without breaking vacuum. The nominal sputtered thickness is estimated from the sputtering time and the calibrated rate. The sputtering rate is calibrated by depositing 20 to 40 nm and measuring the X-Ray Reflectivity profile.

![Schematic of sputter deposition.](image)

Figure 3-1: Schematic of sputter deposition.
3.2 Patterning of films

The magnetic films need to be patterned, labeled, and prepared for optical holography experiments. Lithography is used extensively in the patterning sub-micron features on thin-film solid-state devices. I used photolithography to sputter Hall Bars for harmonic measurements and electrical contacts for electrical connection. The electron beam lithography (EBL) process used to pattern hall cross devices for electronic measurements and magnetic wire membranes for the X-ray holography measurement was carried out by F. Büttner at MIT. The patterning of object holes, references, and individual membrane labels was carried out with the focused ion beam (FIB) at MBI by Christian Günther.

3.2.1 Photolithography

We first explain the general process of lithography: it begins with spin coating a thin resist-layer on a substrate. The layer is subsequently exposed, or a chemical change of the resist layer is induced to transfer the pattern onto the resist. The resist layer is then developed by an appropriate chemical which either removes the exposed (positive) or non-exposed (negative) resist. The pattern transfer is then carried out by either a subtractive or additive process (in this case, subtractive, as positive resist will be used). Material is deposited onto the positive substrate, and the unneeded material is removed along with the resist during the lift-off step.

3.2.2 Electron Beam Lithography

In electron beam lithography (EBL), electrons are used to develop an electron sensitive resist layer. A high level of pattern resolution can be achieved with the EBL technique, such as the one used to pattern the samples in question. The following are typical steps of EBL for patterning of magnetic films. First, an Si wafer with a thermally grown SiO₂ capping layer is prepared. A polymeric mask, which is
poly-ethyl-methacrylate (PMMA) in our case, is then spin-coated directly onto the substrate surface. In this process, a 4% PMMA is generally used to generate a 200nm-thick layer at a 5k-rpm spinning speed. The exposure of electron beam is performed using a Raith 150 scanning electron beam writer, with a maximum of 30kV acceleration voltage and 30-120 μm aperture size. The exposed PMMA (a positive resist) is then developed and removed with a solution of methyl isobutyl ketone and isopropanol with a 1:3 volume ratio. Magnetic films are deposited using the magnetron sputtering, and the excess PMMA is removed during lift-off via sample immersion in n-methyl-2-pyrrolidone (NMP) assisted by ultrasound sonication.

Figure 3-2: Nano-structure fabrication sequence using the electron beam lithography technique. Reproduced from [42].

**Focused Ion Beam milling**

Focused ion beam (FIB) milling, a process similar to sputtering, removes material directionally by ion bombardment. In this method, the layer structure is sputtered before spin-coating. A typical mask can be used with negative photoresist (as in this thesis), with the remaining photoresist covering the needed structures. An advantage is that unlike in liftoff, very smooth and defined edges can be obtained. Another advantage of the FIB, especially for the patterning of nano-scale holography object holes as described in this thesis, is the localized working distance of the ion beam (≈ 10 nm).
3.3 Magneto-optical characterization

The magneto-optical Kerr effect (MOKE) is the main technique used in this thesis to measure the magnetization and properties of magnetic thin films. The popularity of the technique originates from its versatility and monolayer sensitivity, which allows the measurement of magnetic hysteresis loops even on ultra-thin magnetic structures [49]. While the MOKE measurement allows acquisition of magnetic hysteresis loops on short time scales (< 1 s), the MOKE signal does not provide a quantitative measure of the sample magnetization.

Principle

The MOKE refers to the change in polarization that occurs when linearly polarized light is reflected off a magnetized surface. Figure 3-3 shows a schematic of the experimental setup typically used for MOKE microscopy. A film is used to linearly polarize the light from a laser source. The film polarized light can be considered as a superposition of two circularly polarized components with two different helicities. When the linearly polarized light is reflected from a magnetic thin film, interaction between the light and the magnetization results in a rotation of the polarization axis (i.e., the Kerr rotation) and a slight ellipticity (i.e., the Kerr ellipticity). Before light is reflected from a magnetic medium, it travels a small distance into the medium (≈ 20 nm) where the two modes of light polarization propagate with different velocities and absorption rates. The polarization will rotate due to differing velocities, and the different absorption rates will cause a finite ellipticity in the reflected beam.

The three types of Kerr effect can be distinguished based on the orientation of the magnetization and the plane of incident light (Fig. 3-3): in the (1) longitudinal MOKE, the magnetization is in the plane of incidence and perpendicular to the surface normal, in the (2) transverse MOKE, the magnetization is perpendicular to the plane of incidence, and in (3) the polar MOKE, the magnetization is in the plane of incidence and parallel to the surface normal.
Building a custom MOKE set-up

I set up the MOKE system with a magnet that reaches fields of 1.5 Tesla (Fig 3-4). The set-up used consists of light from a diode laser ($\lambda = 650$ nm) beam (1) traveling through a Glan-Thompson Calcite polarizer, and deflected onto the sample with an angled mirror (3), with the numbers in parentheses referencing the labels in Fig. 3-4. The sample is held by a custom-made sample holder (4) and is subject to a field from the steel core magnet (5). The beam is then reflected off the sample, and another angled mirror is used to deflect the beam onto the analyzer to produce a measurement of Kerr rotation. The light intensity is then focused with a lens into the photodetector (8) with a filter. This beam then passes through a pre-amplifier, which amplifies the signal into the MCC (Measurement Computing Corporation) data acquisition controller (DAC), which synchronizes the output of applied fields and currents (digital to analog conversion) to the sample and an data input about how the sample responds (analog to digital conversion). I control the field sweep rate cycle time with data acquisition software designed by M. Mann, and sensitivity by modulating the analyzer angle from extinction. The laser spot is of an approximately 5 mm diameter and therefore the measurement is most suitable for large reference samples rather than small devices or wedges with a gradient of thickness.

To measure a magnetic hysteresis loop, I input the desired waveform and field value
magnitude into a data acquisition LabView program. The waveform is then sent to
the input channel of the DAC, and to the field controller, an analog controller for the
electromagnet. The control is set up so that a field program input of 1 V produces
a given field, such as 6000 (0.6 T). The field controller incrementally increases the
signal sent to the amplifier, changing the magnetic field, until the field measured by
the Hall probe attains the setpoint value. During the field controller calibration, I
tune the damping and feedback time constant to have the fastest response without
overshooting the setpoint field value. An amplifier is used to program the voltage
output in accordance with the voltage signal from the field controller.

3.4 X-ray Holography Imaging Technique

Holography for soft X-ray imaging was first demonstrated in a lensless setup in 2004
[51], and is an approach that is continued to be actively developed. The basic ap-
proach involves the set-up in Fig. 3-5, where the optics of the imaging process involves
a mask in the X-ray beam, with circular apertures (the object and reference holes).
The reference holes are at least three times the object hole radius away from the
object hole. The specimen for imaging is placed behind the object hole and attached to the mask to prevent drift of the reconstructed image. The beam is transmitted through the object and reference holes, and the sample is diffracted as a result of the absorption and phase modulation (Fig. 3-5). The interference pattern of the three diffracted beams is the hologram, and is recorded with the camera.

We exploit the circuluar dichroism effect, or the differential absorption of left- and right-handedly polarized light, to tune to a specific wavelength [52]. This wavelength corresponds to energy at which there is a discontinuity in the linear absorption coefficient of X-rays by an element, which occurs when the energy of the photon corresponds to the energy of a shell of the atom. The sample is illuminated with coherent circular x-rays at the Co $L_3$ absorption edge. The $L_3$ shell corresponds to the creation of electron holes in the $2p_{3/2}$ atomic subshells. The scattering pattern of the transmitted light contains the information about the local orientation of the Co magnetization in the sample. This information is encoded in the amplitude and phase of the wavefield. The phase information is conserved in the detection process by virtue of interference with three reference beams from point-like sources. Each reference interference leads to an independent reconstruction of the magnetic image of the sample, an example of which is shown on the computer screen.

![Figure 3-5](image)

Figure 3-5: a, Pulse injection setup, showing the microstrip with a notch through which we inject spin orbit torque current pulses. The real-time current density is measured via an oscilloscope. b, Schematic of x-ray holographic imaging.
Chapter 4

Micromagnetic Simulations of Skyrmion Collapse in Ferromagnets

To investigate the reason for deterministic collapse of skyrmion bubbles in experiments, as well as the effect of the in-plane field on skyrmion bubble stability in ferromagnetic thin films, micromagnetic simulations of collapse radius and DW width have been performed. In this chapter, I show that skyrmion collapse with out-of-plane fields occurs faster in the presence of in-plane fields, and the in-plane field has a greater effect on de-stabilizing the skyrmion when the film lacks DMI. I propose an explanation for the first phenomenon by showing that the in-plane field increases the domain wall width as well as the domain wall energy, which causes the de-stabilization of the skyrmion. I show that the domain wall energy has a marginally larger increase during the collapse process for the zero-DMI skyrmion, which corresponds to the lack of stability. Finally, a possible explanation is presented for the increasingly deterministic collapse with increasing in-plane field as presented in a previous study in Section 2.3.2, Fig. 2-10.

The phenomenological behavior and deformation of skyrmions has been previously observed numerically and experimentally under tilted and in-plane fields [10] [11] [12]. For example, under applied in-plane field, DMI skyrmions have been asymmetrically
(elliptically) deformed in domain wall driving and domain wall creep experiments [10]. These studies have also suggested that the in-plane field breaks the symmetry of the Néel domain wall, so that the magnetic moments on one side of the bubble domain are initially antiparallel, while moments on the other side are parallel, to the magnetic field. However, there have been little systematic investigation into the deformation of the skyrmion domain wall or domain wall width itself as a consequence of in-plane field effects.

4.1 Simulations parameters

The magnetic layer in the simulation is described by a continuous ferromagnetic configuration, with a varying thickness of the epitaxial Co layer (1 nm for Néel skyrmion, 2.8 nm for Bloch skyrmion). In the Néel skyrmion case, we consider an ultrathin film, where DMI originates from the spin-orbit coupling between the high-spin orbit heavy metal and ferromagnetic adjacent layers, and volume charges have a negligible contribution to the demagnetizing energy.

The magnetostatic material parameters were implemented based on [7]: saturation magnetization $M_s = 1.4 \times 10^6 \text{A/m}$, exchange stiffness constant $A = 1 \times 10^{-11} \text{J/m}$, uniaxial anisotropy constant $K = 1.2 \times 10^6 \text{J/m}^3$, damping constant $\alpha = 0.9$, DMI constant fixed at $D = 0.8 \text{mJ/m}^2$ and $D = 0$ for the Néel and Bloch type skyrmion, respectively. A cell discretization of $0.25 \times 0.25 \text{nm}^2$ was used. Periodic boundary conditions were imposed on the sample, and the simulation area is large enough for an isolated skyrmion to remain unaffected by the edges (larger simulation areas produced no configurational or energy changes). The effective anisotropy field $H_k$ of the Néel and Bloch case, is 1400 Oe.

4.2 Skyrmion collapse via unidirectional field

We first consider the simplest case where the skyrmion collapse occurs via solely an in-plane or out-of-plane field, where the starting configuration is obtained from a
stable relaxed skyrmion state at 40 mT out-of-plane field as in Buettner et al. This original state is for both a Néel and Bloch skyrmion (Fig. 4-1).

Figure 4-1: A Bloch (green) and Néel (red) skyrmion of similar radii as a function of applied field. Small data points are theoretical predictions while large data points are micromagnetic simulations.

An in-plane field is then applied to the relaxed state in the +\( y \) direction (Fig. 4-2) in incremental steps of 100 Oe (10 mT) until collapse. At a certain threshold value of the field, \( H_T \), we see the emergence of a Bloch point in both the case of a Néel and Bloch skyrmion, after which the skyrmion bubble keeps shrinking with increasing in-plane field steps until collapse.

The Bloch and Néel skyrmions are then equilibrated at 400 Oe (40 mT) out-of-plane field at a range of in-plane fields (from 0 to 800 Oe), as shown in Fig. 4-3 a,b. Successive out-of-plane field is applied stepwise until a collapse field with a precision of 5 mT is reached, and each bubble is annihilated.

We numerically find the equilibrium radius of each type of skyrmion as a function of OOP field. The radius is defined as \( R = \sqrt{A/\pi} \), where \( A \) is the skyrmion area weighted by the \( z \)-component of magnetization. We approximate the skyrmion to be spherical with radius \( R \), although the skyrmion shape becomes more elliptically deformed with higher IP fields. The collapse path is simulated for several different values of IP field, including no field at all. We observe that in the case of the Bloch skyrmion, collapse occurs at much smaller OOP fields and the skyrmion collapses.
easier. This reflects the role of the DMI term in stabilizing the skyrmion.

![Figure 4-2: Plot of (a) Bloch and (b) Néel skyrmion radius \( R^* \) as a function of applied OOP field at different applied in-plane fields.](image)

We therefore show that the collapse path follows a qualitatively similar behavior for the Néel and Bloch skyrmion apart from the high in-plane case. The general difference in the collapse field magnitude for the different in-plane fields is also significantly larger for the Bloch skyrmion. This difference can be attributed to the effect of \( H_{DMI} \) as the stabilizing field (and corresponding energetic term) that counteracts the increase in Zeeman energy and in-plane deformation. The 800 Oe in-plane field is so close to the collapse field that there must be only slight amount of out of plane field applied for spontaneous annihilation (420 Oe for Bloch skyrmion annihilation, only a 20 Oe increase from the initial state, as opposed to 1300 Oe for the Néel skyrmion). The reason for this observed qualitative stability is presented later in the section.

At collapse, the total energy of a skyrmions should approach the universal energy of collapse, \( 27Ad \), as has been theoretically shown [62]. The total skyrmion energy for the Bloch and Neel cases was obtained by subtracting the uniform state \( E^0 \) energy from the film and skyrmion total \( E \) energy as calculated in Mumax:

\[
E_{sk} = E - E^0
\]
The $E_{sk}$ was then plotted in units of $Ad\ (E/Ad)$, where $d$ is the film thickness and $A$ is the exchange constant, consistent with previous theoretical treatments of skyrmion energy barriers [46]. We find that the energy rises faster for the Bloch skyrmion as IP field is increased (Fig. 4-3), which corresponds to the smaller collapse fields for the Bloch skyrmion.

![Figure 4-3: Total skyrmion energy of Neel and Bloch skyrmions scaled by the $Ad$ factor.](image)

### 4.2.1 Skyrmion deformation during collapse under IP fields

In order to isolate the impact of the in-plane field on the skyrmion deformation and collapse, we begin with equilibrium Bloch and Néel skyrmion states at zero in-plane field, and an $H_{OOP} = 40$ mT, as shown to be stable in Fig. 4-1. The skyrmion state is relaxed at this field, after which a successive in-plane field is applied in 10 mT increments. The skyrmion state is relaxed at each increment and each magnetization vector field is extracted, and plotted as a vector map in Fig. 4-4. The red and blue colormap corresponds to a $+1$ and $+1$ out-of-plane direction of magnetization, respectively, according to the out-of-plane direction of the magnetization (that is, specifically showing the magnitude of the component $m_z$ in a standard film geometry).
Figure 4-4: Successive collapse of (a) Bloch and (b) Néel skyrmion by an increasing in-plane field, each image is an equilibrated state. The field was ramped up from 0 Oe to the collapse field (within 5 Oe) and the skyrmion state was relaxed at each step.

In both cases of collapse via in-plane field, we observe a high degree of boundary transition region (white color) of the $m_z$ vector, indicating a highly deformed domain wall. We may also compare the spin structure along the domain wall, which is indicated by the black arrows in Figs. 4-4 and 4-5. In the collapse in Fig. 4-4, in Bloch case we see a difference in the domain wall structure under the highest in-plane field compared to the lower in-plane fields - the appearance of a highly deformed domain wall and $360^\circ$ spin rotation radially, and a localized $180^\circ$ rotation at one side of the skyrmion (as seen in the inset of the last panel of Fig. 4-4,a). The highly deformed, broad domain wall (white part of the magnetization in Fig. 4-4) also corresponds with the steep collapse curve at this field. Plotting the final equilibrium state of each of the skyrmions in Fig. 4-2, we observe that the skyrmion is subject to a similar deformation right before collapse (Fig. 4-5). In contrast to the Bloch case, the Néel skyrmion, under high OOP fields, does not deform as significantly before collapse (Fig. 4-5 b). This is reflected in the moderately increasing difference in collapse field
for the curves in Fig. 4-2(b).

Figure 4-5: (a)-(b) shows the Bloch and Néel skyrmion states, respectively, right before collapse, with a zoom into the magnetization rotation at the highest OOP field before collapse.

Is there a topological transition or key change in spin structure that can be observed at the critical collapse field? While there is no transient topological state between the skyrmion state and the ground state ([62]), it has been shown that the energy barrier associated with the collapse mechanism is proportional to the number of spins that need to be flipped in this process and, therefore, scales with the effective skyrmion area $\pi R^2$ [27]. Previous studies, which use multiscale or atomistic methods, show that the barrier goes to zero as the skyrmion size approaches the size of the saddle point excitation - that is, Bloch point-like defect [18]. Thiaville et al. have shown that the modeling of singularity is mesh dependent within the continuum description of micromagnetism [44]. As the Bloch point appears as a singularity of continuum theory, it is always located between mesh points and causes the mesh-dependent effects [20]. While we do not make any claims about the nature of Bloch point formation due to this reason, we do observe a greater deformation of the Bloch skyrmion just before collapse, with the spins rotating 360 degrees through the domain wall line structure. To understand the deterministic nature of collapse that was observed experimentally (in Chapter 2), we characterize not just the size of
the skyrmion but also the domain wall profile.

4.3 Domain Wall Considerations

In previous studies of in-plane field effects on domain walls, a straight domain wall is considered. Furthermore, it is usually assumed that IP field should act uniformly on a bubble and it should retain its original DW structure, but we observe a preferred direction of domain wall deformation, which provides a new, interesting insight into skyrmion deformation under fields and several stepping points to further studies.

4.3.1 Domain Wall Width

To characterize the domain wall profile and maximum domain wall width of a skyrmion (which indicates its degree of asymmetric deformation), we take magnetization profile around the center of the magnetization vector field obtained from each equilibrium skyrmion state. Since there is no exact analytical expression to describe non-circular Skyrmion profiles, we approximate the cross section of a Skyrmion using a standard 360° domain wall profile [35]:

\[
\theta(r) = 2 \arctan \left( \exp\left( \frac{r - r_0}{\Delta} \right) \right)
\]  

for a single domain wall located at position \(r_0\), where \(\theta\) is the polar angle of the magnetization (\(\theta\) shown in Fig. 4-5 c,d). The DW magnetization function is then:

\[
m_z(r) = \cos \left( 2 \left[ \arctan(\exp(\frac{r + r_0}{\Delta})) + \arctan(\exp(\frac{r - r_0}{\Delta})) \right] \right)
\]

at position \(r\), and \(r_0\) and \(\Delta_0\) are the position and width of two overlapping 180° domain walls, respectively. This magnetization function \(m(r)\) was obtained for each skyrmion profile and fitted with the theoretical domain wall profile. The domain wall width was then found from the fitting parameter \(\Delta\). The DW width, \(\Delta\), for the Bloch and Néel skyrmions was then found, as shown in Fig. 4-6 (a) and (b), respectively.
Figure 4-6: Domain wall broadening is seen in both the (a) Bloch and (b) Néel skyrmions in the equilibrium state before collapse, with the domain wall width as a function of polar angle (indicated in middle section) shown.

The DW width for a skyrmion at an equilibrium state just before the final collapse field is applied increases linearly with increasing applied in-plane field. As the IP field increases, there is less impact from thermal fluctuations on the broader domain walls (less impact of pinning). This can be seen from considering the effect of the pinning sites within the magnetic film's energy landscape on a thicker DW wall (as discussed in Section 2.2). With less thermally activated skyrmion collapse, the distribution of skyrmions remaining after a certain applied field is applied will have a smaller half width half maximum, and therefore the collapse is more deterministic. This is reflected in the skyrmion collapse in the experimental sample. One important factor to note is that the simulations are carried out at 0K temperature for a single skyrmion, while the experimental values are obtained at room temperature where thermal effects come into play.

4.3.2 Domain Wall Energy

The skyrmion spin structure can be accurately described by four key parameters: radius $R$, DW width $\Delta$, DW angle $\phi$, and topological charge $N$. The magnetization profile $m_z(x,y)$ as described in Chapter 2 is determined by $R$ and $\Delta$, and the radial
or azimuthal nature of the in-plane component of the domain wall spins is specified by \( \phi = 0 \) (Néel) or \( \phi = \pi/2, \pi/3 \) (Bloch) [46]. We consider the skyrmions treated in this film as bubble skyrmions with a large \( \rho = R/\Delta \) with a large out-of-plane domain bounded by a narrow circular DW. The energy of such bubble skyrmion can be derived from the wall-energy model [13]:

\[
E = 2\pi d \sigma_{DW} R + aR - bR \ln(R/d) + cR^2
\]

where \( 2\pi d \sigma_{DW} \) is the DW energy, and \( \sigma_{DW} \) is the energy density of an isolated DW, and the two terms on the right are the Zeeman-like surface stray field energy and the Zeeman energy, respectively. In this large-radius approximation, the first term is the expression for total domain wall energy. This model is valid for skyrmions above the radii of \( 10d \) (where \( d \) is the film thickness), and we make the approximation in this case that the skyrmions are not undergoing domain wall repulsion (and henceforth adding a contribution to the magnetostatic energy) at radii under \( 10d \).

In the simple case of an isolated 180° domain wall in a single, uniform layer of magnetic material as seen in Chapter 2, there is an intrinsic thickness dependence of domain wall width \( \Delta \) that is caused by the magnetostatic interactions, which changes the local value of anisotropy \( K \) in the proximity of the domain wall. In the theoretical description of the domain wall profile, there are two contributions to the magnetostatic energy: (1) the stray field energy associated with the volume charges inside of the isolated domain wall, and (2) the stray field energy of surface charges surrounding the domain wall. In the case of the domain wall of a magnetic skyrmion, there are two contributions to the magnetostatic energy of the domain wall (due to the volume and surface charges). To properly account for DW energy we need to consider both. The total output of the Mumax magnetostatic energy includes these contributions and the magnetostatic energy of the film. We do not know the surface charges (which dominate in a thin film) so we subtract the total magnetostatic energy from the total film energy (including the DW demagnetizing energy). This means that the resulting domain wall energy neglects the magnetostatic contribution. To account for this,
Figure 4-7: Magnetostatic energy contributions to the total film and skyrmion energy include (a) magneto-crystalline film volume charges and (b) cylindrical domain wall volume and surface charges.

one could calculate the magnetostatic contribution theoretically from a cylindrical domain wall with a non-zero ellipticity (Fig. 4-7(b)), a subject for further study.

Noting this consideration, we calculate the domain wall energy of a Bloch and Neel skyrmion. The domain wall energy itself includes the exchange and anisotropy contributions, since the magnetostatic and Zeeman energies must be subtracted. *Mumax* subtracts the magneto-crystalline anisotropy \( K_u \) from the total energy value. The total domain wall energy, for the entire volume of the film, using the energy values from the output of the simulation, is approximated as:

\[
E_{DW} = E_{tot} - E_{zeeman} - E_{demag} - E_{Ku},
\]

where \( E_{Ku} \) is the magneto-crystalline contribution to the anisotropy from the ground state. From the total DW energy \( E_{DW} \), we can then determine the DW energy density through the circular line structure, \( \sigma_{DW} \),

\[
\sigma_{DW} = \frac{E_{DW}}{2\pi Rd}
\]

When calculating the approximate DW energy for the two skyrmions collapsing via in-plane fields, we observe that DW energy monotonically increases with increasing IP field, and is larger for the Bloch skyrmion at high IP fields (Fig. 4-8). The IP field therefore increases the DW energy, and decreases the stability of the skyrmion bubble,
which explains the decrease in collapse OOP field with increasing IP field (Fig. 4-3). The DW energy density increase also corresponds to the broadening of the DW wall at very high IP fields. The larger difference in DW energy for the Bloch skyrmion than the Neel skyrmion DW at high IP fields reflects the gap between collapse fields of the Bloch skyrmion at very high IP fields of 800 and 600 Oe (Fig. 4-2).

![Figure 4-8: Domain wall energy of Neel and Bloch skyrmion, calculated micromagnetically.](image)

We note the presence of error from the circular approximation to the linestructure within the calculation of the domain-wall energy using Eq. 4.0. However, we see that there is a larger rise in the skyrmion energy from Fig. 4-7 for smaller IP fields as well, so there must be another energetic contribution besides the domain wall energy. It is possible that this is a demagnetizing energy contribution that is difficult to account for in a theoretical model for an asymmetric skyrmion.

### 4.4 Conclusion

Using micromagnetic simulations to model skyrmion collapse in ferromagnetic material, I provide possible explanations for the experimental observations of skyrmion
bubble stability in Co/Gd thin film with DMI. We suggest that the decreasing collapse field with increasing in-plane field is a result of increasing the DW energy density as well as the total skyrmion energy, which is consistent with the conditions necessary for bubble collapse \[49\]. Secondly, the collapse field may become more deterministic with higher in-plane field due to a lesser impact of thermal fluctuations and pinning landscape on wider DW walls. I show that the deformation of the skyrmion and the domain wall is more drastic as a result of the in-plane field for the Bloch than the Néel skyrmion (with DMI). To see the impact on this on the skyrmion collapse, we would need to use alternative numerical methods that take into account finite cell-size effects and local magnetostatic energy contributions of the domain wall itself, such as atomistic, density functional theory, or multiscale approaches. For instance, multi-scale spin-lattice simulations that use Langevin dynamics are free from mesh-dependent effects and are able to consider the magnetization behavior under non-zero temperature conditions (closer to room-temperature experimental studies).
Chapter 5

Ultrasmall skyrmions in ferrimagnets

While domain wall and skyrmion motion has been studied extensively in ferromagnets, there still remain key challenges to developing efficient ferromagnetic spintronic devices. While chiral spin textures have been manipulated by spin-orbit torques (SOTs) in perpendicularly-magnetized ferromagnetic films[60], there are inherent limitations to their stability and speeds. In particular, bit sizes are limited by stray field interactions [62][63], and operating speeds are limited by precessional forces[63]. Meanwhile, antiferromagnets are free from stray field effects and have been predicted to allow uninhibited skyrmion motion as well as sub-10 nm sizes of bits. Because it is challenging to magneto-optically image antiferromagnets due to their compensated moments, we turn to ferrimagnetic materials that are able to be studied via differently interacting layer components with optical or electronic excitations.

In this chapter, we show that we can generate and detect ultrasmall skyrmions with high contrast in ferrimagnets at magnetization compensation. ¹ These materials are able to exhibit magnetic skyrmions that approach 10 nm in size at room temperature and zero applied field, which has previously only been observed under

¹Sections of this chapter including figures are accepted for publication in - L. Caretta, M. Mann, F. Büttner, K. Ueda, B. Pfau, C.M. Günther, P. Hessing, A. Churikova, C. Klose, M. Schneider, D. Engel, C Marcus, D. Bono, K. Bagschik, S. Eisbitt, G.S.D. Beach, “Ultrafast domain walls and ultrasmall skyrmions in a compensated ferrimagnet,” Nature Nanotechnology.
cryogenic temperature and under fields of > 1 T [64]. This finding is of great interest to skyrmion-based spintronics, since at compensation temperature, there will be little to no Hall effect [8] and gyration of skyrmions [32]. This can be shown by the direct proportionality of the gyromagnetic term in the Thiele equation to the precession term described in the LLG equation in Section 2 [46]. The result is that at \( T_M \), both terms vanish, leading to ultrafast switching and spin texture movement without precession or gyrotropic motion [61]. To see how this behavior comes about, we note that the magnetization dynamics in ferromagnets follow the LLG equation described in Section 2.4.2,

\[
\frac{dM}{dt} = -\frac{\gamma \mu_0}{1 + \alpha^2} (M \times H) - \frac{\alpha \gamma}{M(1 + \alpha^2)} M \times (M \times H)
\]

where \( H = \frac{\delta E}{\delta M} \) is the effective magnetic field, including applied and external contributions, \( \alpha \) is the Gilbert damping, and \( \gamma \) is the gyromagnetic ratio. The first term induces precession around \( H \) and the second dissipative term rotates \( M \) toward \( H \), imposing a limitation on the switching speeds in ferromagnets to GHz frequencies.

In ferrimagnets, \( \gamma \) and \( \alpha \) are replaced by [54]:

\[
\gamma \rightarrow \gamma' = \frac{M_s(T)}{S(T)}
\]

\[
\alpha \rightarrow \alpha' = \frac{S_0}{S(T)},
\]

where \( S(T) \) is the net spin density and \( M_s(T) \) is the magnetic moment as a function of temperature. The net magnetization of a two-element ferrimagnetic sublattice is then given by:

\[
M_s(T) = M_{s,1}(T) - M_{s,2}(T),
\]

and the net spin density as

\[
S(T) = \left| \frac{M_{s,1}(T)}{\gamma_1} - \frac{M_{s,2}(T)}{\gamma_2} \right|
\]

\[
S_0 = \alpha_0 \left( \frac{M_{s,1}(T)}{\gamma_1} + \frac{M_{s,2}(T)}{\gamma_2} \right),
\]

60
where subscripts refer to the sublattices 1 and 2, \( \alpha_0 \) is the damping constant, and 
\[ \gamma_i = g_i \mu_B / \hbar \] is the gyromagnetic ratio (where \( g \) is the Landé g-factor, \( \mu_B \) is the Bohr magneton, and \( \hbar \) is the reduced Planck constant). At \( T_M \), \( M_s \) and stray fields are zero, which is not usually the same as the compensation temperature for angular momentum, \( T_A \) where the net spin density is zero. Since effective fields generally scale as \( 1/M_s(T) \), applied torques are predicted to be most efficient near \( T_M \). Because \( \gamma \) and \( \alpha \) diverge at \( T = T_A \), the precessional term in the LLG equation disappears and we are left with:

\[
\frac{dM}{dt} = -\frac{M_s(T)}{S_0} M \times (M \times H) \tag{5.5}
\]

Since this equation implies that \( M \) is moving toward \( H \), ultrafast switching and efficient chiral spin texture movement (such as that of skyrmions) without precession or gyroscopic motion is expected at these temperatures.

In fact, it has been shown that reducing the (topological) damping and increasing the current-induced effective field are the two ways to improve the mobility of small skyrmions [62]. Furthermore, due to the low correlation of \( K_u \) and \( M_s \) in ferrimagnetic materials, these parameters can be optimized independently to maximize the skyrmion quality factor with a low \( M_s \) and high \( K_u \). Specifically, in ferrimagnetic rare-earth metal alloys, the high degree of PMA and higher permissible thickness of films allows to reduce the DMI value \( D \) needed to stabilize skyrmions. From previously calculated phase diagrams, these materials also exhibit exchange constant values between 4 pJ/m and 10 pJ/m which correspond to stable skyrmions at small radii and DMI values [62]. This makes ferrimagnetic bulk rare earth metal alloys an ideal candidate for studying current-induced generation of very small skyrmions.

5.1 Experimental Methods

The measurements of skyrmion generation and stability in this Chapter are carried out in Gd\(_{44}\)Co\(_{56}\), an amorphous ferrimagnetic alloy with antiferromagnetically-coupled sublattices having different g-factors. The latter fact allows \( T_A \) to be close to \( T_M \) and the tuning of these compensation temperatures to room temperature.
5.1.1 Fabrication

The Ta(1nm)/Pt(6nm)/Gd$_{44}$Co$_{56}$(6nm)/TaO$_x$(3nm) ferrimagnetic stack with interfacial DMI studied in this chapter was grown with d.c. sputtering by K. Ueda (Fig. 5-1,a). The ferrimagnetic alloy was co-deposited with separate Co and Gd targets, with an Ar sputter gas pressure of 3 mTorr, and a background base pressure of $2 \times 10^{-7}$ Torr at nominal room temperature. The alloy composition was chosen to produce a magnetization compensation at room temperature (Fig. 5-1,b), and was controlled by varying the Gd sputter gun current. Samples were deposited on thermally oxidized Si wafers. Current shunting through the Ta seed layer was neglected as the resistivity of Ta is approximately 10 times higher than that in Pt. It has also been confirmed with a square out-of-plane hysteresis loop that the sample exhibits bulk perpendicular anisotropy.

Figure 5-1: a, Ferrimagnetic stack structure. b, $M_s(T)$ data measured on a continuous film reference sample near $T_M$ using vibrating sample magnetometry by M. Mann and L. Caretta.

I deposited the Cr(5 nm)/Au(100 nm) outer and inner contacts by thermal evaporation and shadow masking. To secure each wafer and the contact geometry shadow mask on inner lid of the thermal evaporation chamber (Fig. 5-2,a), I first designed and machined the sample stage that is specific to the 5mm $\times$ 5mm wafer (shown in Fig. 5-2,b). The gold is deposited on the top of a thin layer of chromium to prevent gold island formation on the film and ensure full adhesion of the gold layer to the substrate. The contact pattern mask was placed on the top of the wafer, shielding the
magnetic membranes but exposing the contact areas. Then, the sample chip, covered with the mask, was secured on top of the sample holder, and attached to the lid of the evaporator chamber. The chamber was then sealed and pumped down to 4e-5 mBar. Both inner and outer contacts were deposited this way.

![Image](image.jpg)

**Figure 5-2:** a, Thermal evaporation chamber used to deposit Cr/Au contacts. b, Custom-designed stage which secures the wafer with samples and contact shadow mask together on the chamber’s inner part.

The sample membranes were prepared by F. Büttner, C. Günther, and Schneider: with FIB by defining a 10μm wide tracks of the same Ta(1)/Pt(6)/GdCo(6)/TaOx(3) material on a 700 nm thick SiN membrane supported by a high purity Si frame, fabricated with EBL and lift-off. These were one object hole (1 μm in diameter) and three reference holes with 30 nm, 40 nm, and 100 nm diameters. A layer of [Cr(5 nm)/Au(100 nm)]<sub>20</sub> was also deposited on the backside of the membrane, with a holography mask that contains four holes made via ion milling. Finally, the 10 μm track was reduced to 900 m, in the region behind the object hole, and a notch was created to assist current induced nucleation of skyrmions.
5.1.2 X-ray holography

We perform fourier transform X-ray holography measurements at room temperature in a custom MAXI endstation at the P04 beamline of the PETRA III synchrotron radiation source, Germany. We use a 777.7 eV photon energy to tune to the Co L₃ absorption edge, to obtain holograms with opposite circular polarization. P. Hessing performed the reconstruction of the holograms (via direct Fourier inversion and propagation to focus the image [17]) to produce magnetic contrast images of the Co magnetization in the 800 nm field of view. On the images, the dark contrast regions shows magnetization pointing out of the plane, and and light contrast showing magnetization pointing into the plane in white.

We use small magnetic tracks of 1 μm and 2μm length. There is a small notch at the center of the wire for a local increase of current density, which has been found useful for skyrmion nucleation via spin orbit torques [31]. During the experiment, we apply bipolar pulses with amplitude up to 1.7 TA/m³ and 10 ns width for an extended period of time with cumulative current flow of 40s, after which we noticed a change in film topography near the notch, suggesting electromigration-induced damage. We thus focus on the regions of the film far away from the notch where magnetic properties are minimally affected. We show this by confirming that the out-of-plane domains in the demagnetized state and a lack of out-of-plane saturation. We confirm that the features are ascribed to skyrmions and not to artifacts from topography, since magnetic contrast can be separated from the topographic features by subtracting holograms with positive helicity from holograms recorded with negative helicity. Skyrmions appear in varying locations during each nucleation sequence, which rules out a confusion with topological artifacts (would be always in the same location).

5.2 Skyrmion generation and collapse

In the published studies to date, room-temperature skyrmions have been shown to be much larger than the 1-10nm skyrmions seen in single-layer ferromagnets at cryogenic temperatures [47] [48], with sizes ranging from 30nm to 2 μm [6] [5]. The larger sizes
relate to the dominant dipolar interactions in the heavy-metal/ferromagnet multilayers commonly used to realize them [46]. The size dependency can be understood from the skyrmion energy versus radius landscape as shown by Büttner.

A typical skyrmion energy as a function of radius curve for a given skyrmion in a high $M_s$ ferromagnet is shown in Fig. 5-3,a. An external field is required to stabilize a skyrmion of any size. The minimum in the energy landscape is due to a balance of stray field, dipolar, and domain wall energies, as previously shown in circular domains in bubble materials [49]. The maximum just after $R = 0$ pushes the equilibrium radius to larger values. The skyrmion is strongly affected by stray fields, as the minimum defines a stray field skyrmion [46], with a characteristic field-dependent size (Fig. 4a), collapse at a finite diameter and finite field, and expansion into stripes at zero field. At room temperature, the these stray field skyrmions collapse at a diameter much larger than 10nm for plausible material parameters [46]. In atomically thin high-DMI ferromagnets where skyrmions have been observed [5], a minimum can exist in the energy landscape with no intermediate maximum, allowing smaller equilibrium skyrmion size at room temperature. However, the depth of the minimum defining these DMI skyrmions scales with the film thickness. At room temperature the thickness must be higher, and this leads to dipolar fields that decrease the skyrmion stability.

For a ferrimagnetic film, the landscape shown in Fig. 5-3,b predicts a zero-field-minimum at small $R$, energetically favorable for room-temperature stability, field-insensitive size, and DMI skyrmions [46]. This is because increasing film thickness does not correspond to an increase in stray fields, allowing the increase of the depth of the minimum stabilizing DMI skyrmions [46].

In the pump-probe holography measurement, we first show that the current pulses effectively demagnetize the system. We do so by magnetizing the film out-of-plane to saturation, and then injecting a train of of nanosecond current pulses of $1.7 \times 10^{12} A/m^2$ amplitude with zero external field applied. We observe a uniform image contrast for the saturated magnetization (Fig. 5-4,a) and a labyrinthian domain state after the current pulses (Fig. 5-4,b). Therefore we conclude that the current
pulse effectively demagnetizes the film due to Joule heating as the temperature rises above the compensation temperature $T_M$. At this point the magnetization is stray field stabilized. The temperature rise is also evidenced by the topographic changes on the film surface after pulse injection.

When current pulses are injected in the presence of an out-of-plane field $B_z$, we generate skyrmions instead of stripes. The following is the procedure used to generate skyrmions. The film is first saturated to the out-of-plane magnetization. We then apply a single up-down-up current pulse of 10 ns width per polarity and $1.7 \times 10^{12} \text{A/m}^2$ amplitude, in the presence of an out-of-plane bias field (31 mT) applied in the direction of the saturated film. Skyrmions have been observed in a sample where current-induced annealing took place after a high current pulse. The topography of the film, and the part of the membrane that is included in the field of view,
is shown in parts (b) and (a) of Fig. 5-5, respectively. The field of view is the large circle that is defined by the holographic mask (with a radius of 900 nm). Damage due to heating is also visible near the notch. The magnetic images reconstructed from the difference of positive and negative x-ray holograms have very weak topographic features (Fig. 5-5c-d).

Figure 5-5: (a) Top-view of the sample geometry acquired using a scanning electron microscope by F. Biittner. (b) Topography of the sample after current-induced annealing, where the wire shows some signs of damage, in particular in a band-like region near the notch. (c)-(d): Clear magnetic contrast in the images (above the noise level and the residual topography data), with three skyrmions marked with green arrows in (d).

The generated skyrmions can be seen as circular regions of dark contrast in Figs. 5-6, 5-5 after single bipolar pulses were injected in the presence of an applied field of $B_z = 30$ mT. We reconstructed each image independently with a Matlab script developed by Hessing. Skyrmions are considered to be skyrmions when they meet a specific criteria. In Figs. 5-6a-c, we observe multiple exposures of the same skyrmion state without changing external conditions. We identify four skyrmions with the colored circles. We are able to distinguish between the skyrmions and the background intensity fluctuations (artifacts) by comparing multiple exposures of the same state (Figs. 5-6a,b). However in Fig. 5-6c, the smallest of the four skyrmions (16 nm in diameter as calculated with Mathematica script designed by M. Mann) has disappeared, from which we conclude that the thermal lifetime of the skyrmion is on the order of minutes. Larger skyrmions have persisted for several hours in this sample. We were not able to reproducibly move the skyrmions, as there is no correlation between the positions of the skyrmions before and after saturation and re-nucleation.
Figure 5-6: (a) Skyrmions nucleated by a current pulse of 10 ns width per polarity and $1.7 \times 10^{12}$ A/m$^2$ amplitude, with an out-of-plane bias field (31 mT) applied in the direction of the saturated film. (b) Same state as in (a) twenty minutes later. (c) Image after another twenty minutes, when one skyrmion has vanished. (d)-(e): Skyrmions at different positions obtained after saturating the film and injecting the same current pulse pattern as in (a), where colored squares indicate the absence of skyrmions in locations present in (a)-(b). (f) Skyrmions under zero out of plane field (nucleated in a bias field which was subsequently reduced to zero).

In Fig. 5-6f, we see that the skyrmions are stable even in zero applied field, and we do not observe a correlation between the position of skyrmions before and after saturation and re-nucleation. (In Fig. 5-6d, colored squares indicate that skyrmions are no longer present (or too small to detect in the resolution limit), in the locations where they were stable in Fig. 5-6a, b. We also observe that the skyrmions generated on the material vary considerably in size, which indicates that magnetic properties are non-uniform on the film (and likely contain a distribution of random pinning sites).

To investigate the possibility of shrinking the larger skyrmions, we then saturate the film and nucleate two visible skyrmions of different sizes (Fig. 5-7 a). As we successively increase this out-of-plane field in consistent increments of around 15 mT, the smaller skyrmion eventually collapses at 99.7 mT while the remaining larger skyrmion collapses at 104.2 mT. We then nucleate a new skyrmion with a 10 ms triangular pulse, and it collapses at a lower out-of-plane field (34.6 mT) than the previous skyrmions. The skyrmion also did not shrink with current and bias field simultaneously applied (Fig. 5-7), and only collapsed at a sufficiently large enough
Figure 5-7: (a) Two skyrmions were nucleated at saturation, and applying successive out-of-plane fields eventually collapses both. (b) Attempt to shrink a skyrmion with a 0.5V pulse results in eventual collapse at a large enough field. (c) Application of consecutive current pulses with 0.5V transmission results in skyrmion annihilation.

field (106.5 mT).

During the measurements, I have carried out preliminary calculations of skyrmion bubble radii through pixel threshold binning method. The method involved setting a threshold pixel intensity (relative to the background “noise” intensity) for which the magnetization is out-of-plane, or is the dark region of the skyrmion core. The region within this intensity threshold would then be rescaled to the field of view, and the size of the skyrmion was found (an example of the output of such algorithm is the skyrmion in Fig. 5-8,a). The smallest skyrmion size found with this preliminary method was approximately 11 nm. The method for the analysis of a large array of skyrmion sizes was developed by M. Mann, where skyrmion in the images have been fitted to a peak intensity profile. This method yielded a distribution of the sizes with the smallest approaching 10 nm in diameter (Fig. 5-8,b), as well as the capability to provide of measure of skyrmion size beyond the resolution limit (Fig. 5-8,c). To achieve this, the intensity peak of the smaller skyrmion can be fit to the same resolution as the larger skyrmion in the line scan. Skyrmions at this length scale are most relevant for spintronic devices, and must be stabilized by DMI, unlike
all other room-temperature skyrmions shown so far [26].

Figure 5-8: (a) A skyrmion analyzed with the preliminary pixel intensity threshold non-adaptive algorithm. (b) Histogram showing the distribution of skyrmion diameters. (c) Magnetic contrast linescans of numbered skyrmions in Fig. 5-6(c) and (e), and corresponding fits (red).

5.3 Conclusion

Using time-resolved X-ray holography measurements, we show that in ferrimagnetic Pt/Gd\textsubscript{44}Co\textsubscript{56}/TaO\textsubscript{x}, where \( T_M \) and \( T_A \) are close to room temperature, very small skyrmions are stable even in zero applied field in ferrimagnets, can be nucleated reliably with current, and collapsed with high magnetic field. The small size of the skyrmion bubbles, near 10 nm in diameter at the smallest, suggests the presence of a strong DMI in these films, as it is the only energy term to stabilize these kinds of reverse domains and lower their domain wall energy [62]. However, no correlation between the position of skyrmions before and after saturation and re-nucleation and no predictive motion has been observed. This suggests the need to possibly quantify the pinning potentials in the film, or use a notch-less sample to ensure “track” uniformity for potential motion. Skyrmion motion can also be improved by optimizing \( K_u, M_s, T_M \) and the DMI of the film, in the aims of generally increasing the quality factor for skyrmion stability (scales inversely with \( M_s \) and proportional to \( K_u \)).
Chapter 6

Optimization of synthetic antiferromagnetic films

While ferromagnetic materials are promising to experimentally study skyrmions in thin films [5], antiferromagnetic materials are even more promising in terms of stability and motion of skyrmion bubbles. Anti-ferromagnetic (AFM) materials consist of two coupled topological spin configurations with opposite winding numbers ($Q = \pm 1$), one for each sub-lattice [53]. Neither sublattice dominates, as the spins are compensated at the skyrmion core (center). There are specific advantages to these materials as alternatives to ferromagnetic skyrmions in magnetic racetrack memory or next generation memory devices. In ferromagnetic skyrmions, there is a Magnus force that acts on the skyrmion and causes a transverse velocity $v_\perp$ relative to an applied current. Because both sublattices generate a magnus force in the AFM, there is no transverse velocity an the AFM skyrmion travels in a perfectly straight trajectory.

The next advantage of the AFM skyrmions is that the longitudinal velocity has been shown to exceed the FM drift velocity. At low damping coefficient $\alpha$ or high $\beta$ (non-adiabatic spin transfer torque parameter), numerical studies have shown AFM skyrmions to move at speeds on the order of $1 \times 10^3 \text{ m/s}$ without losing stability [53].

Furthermore, AFM skyrmions are insensitive to stray fields (having no demagnetizing field). These potential advantages make the study of AFM and synthetic antiferromagnetic (SAF) materials very attractive for potential use of skyrmions in
ultralow-energy data storage and spintronics. However, there are definitely several challenges with working with skyrmion and DW motion in AFM materials: the detection of the AFM spin state is very challenging, but imaging contrast from magnetic moment of individual layers of an SAF achievable with the X-ray holography techniques described in this thesis.

To introduce the materials optimization necessary for the antiferromagnetic (AF) coupling effect, we bring to the attention the best-studied AF-coupled system of \([(Co/Pt)_{x-1}/Co/Ru]_N\) multilayers [58]. In these multilayers, Co layers are ferromagnetically coupled via weakly magnetic Pt layers. It has been shown that the strength of AF coupling between adjacent Co/Pt stacks depends on the thickness of the heavy metal Ru layer. This interlayer exchange coupling is a spin-dependent Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling, a long range exchange interaction that causes spatial oscillations in the spin density of the non-magnetic spacer via the adjacent ferromagnets. This exchange coupling is controlled by the conduction layer electrons, and oscillates as a function of metallic spacer layer thickness. It was first investigated by Parkin et al. between Co layers across a Ru spacer layer [56].

Tuning the thickness of the non-magnetic heavy metal layer between the two magnetic layers can change the interaction from the ferromagnetic to antiferromag-
Figure 6-2: Interlayer exchange coupling strength $J_{12}$ for coupling of Ni$_{80}$Co$_{20}$ layers through a Ru spacer later, with the solid line corresponding to a fit to the data of RKKY form [57].

netic (Fig. 6-2). Synthetic antiferromagnets (SAF’s) are trilayers, multilayers, or superlattices in which the interaction between magnetic layers is antiferromagnetic. In a Co/Pt multilayer material, the maximum exchange field strength for Ir metal coupling layer was approx. 4 kOe and approx. 1.2 kOe for Ru coupling layer. A higher exchange coupling field is most optimal in SAF materials for generation chiral structures, due to the fact there is a competition between dipolar coupling and AF exchange coupling. The higher the AF coupling, the lower the stray field effects and more potential to host stable skyrmions in the material.

6.1 Methods

Fabrication

The multilayer sample for the X-ray holography measurement of SAF structures was grown using d.c. magnetron sputtering at room temperature by F. Büttner, M. Mei-jer, and R. Lavrijsen, with a Ar sputter gas pressure of approx. 7.5 mTorr and a background base pressure of approx. $2.25 \times 10^{-8}$ Torr on a thermally oxidized O$_2$ wafer. The full structure was Ta(4)/Pt(5)/Co$_{80}$B$_{20}$(1)/Ru(0.8)/Pt(0.8)/Co$_{60}$Fe$_{20}$B$_{20}$(0.75)/Ru(0.8)/Pt(0.8)]$_n$/Pt(1.2) where $n=24$. The processing of the samples and magnetic membranes for the holography experiment was done in the same way as in Chapter
5. I have used the identical technique of contact deposition for these samples.

I grew the Ta(5)/Pt(4)/CoFeB(1)/Ir(0.5)/Pt(t Pt)/CoFeB(1)/Ir(0.5)/Pt(2) samples with d.c. sputtering at an Ar gas pressure of approx. 2.5 mTorr and a background base pressure of $7.4 \times 10^{-7}$ Torr on a thermally oxidized Si wafer.

The rest of the ferromagnetic stacks to be tested as SAF components, and discussed in this section, were grown by me and M. Meijer using d.c. magnetron sputtering at nominal room temperature with a Ar sputter gas pressure of 7.5 mTorr and a background base pressure of $6 \times 10^{-8}$ Torr on a thermally oxidized SiO$_2$ wafer.

So-called wedge deposition on several of the films discussed in this thesis was performed to achieve uniformly increasing thickness of a certain layer or layers. During the sputtering, a knife-shaped mask was linearly retracted above the substrate, resulting in a deposit where the layer thickness varies as a function of position. The sample properties can then be probed as a function of thickness.

**Patterning**

I used the negative resist technique to fabricate Hall bars (32 $\mu$m x 6 $\mu$m with two 3$\mu$m wide cross bars) on the pre-existing sputtered SAF film. I spin-coated NR71-100 resist at 3000 rpm for 40 seconds, and heated (baked) the wafer at 150° C for 80 seconds uniformly. The sample was placed under a mask with a Hall bar pattern (using a karl Suss MA-4 Mask Aligner) and exposed to UV light for 8.5 seconds at 50% humidity level. The exposed sample was then post-baked in 100° C and developed with RD6 developer solution for 10 seconds. We confirmed the Hall bar height using profilometry, and the sample was milled to remove 30 nm of material.

To deposit Au contacts on these Hall bars, I used the positive resist lift-off technique, which required alignment of the contact mask with the existing Hall bars. I spin-coated SPR-700 resist on the sample wafer at 3,000 rpm for 30 seconds. I then bake the wafer at 100° C for 60 seconds. After contact alignment, I expose the sample for 10 seconds at 50% room humidity, and develop the resist for 1 minute in developer. I then sputtered the film with Ti(4nm)/Au(50nm) where the Ti is used as an adhesive layer to bind the Au to the substrate. The resist and unnecessary material is removed.
to yield Hall bars and Au contacts by leaving the sample in a 50° C covered beaker in a water bath for 1 hour.

6.2 Optimizing PMA in SAF Films

The SAF stack optimization began with optimizing individual ferromagnetic layers for optimal thickness of the metallic AF coupling layer, the ferromagnetic layer, and the heavy metal layer (Pt) that is a source of strong spin orbit coupling and DMI. The inversion symmetry is broken due to the different layers on the top and bottom of the magnetic layer yielding two types of interfaces. This provides the potential for generating chiral magnetic structures. Meanwhile, Ru metal was chosen as the first type of AF coupling layer to try, along with $\text{Co}_{1-x}\text{B}_x$ and CoFeB as the ferromagnetic layers. We need to optimize at least two ferromagnetic layers of varying element compositions (e.g. one containing Fe and one not) in order to enable high magnetic contrast in the holography images of multilayer stack. The content of Fe in CoFeB can also be varied in order to determine the most Fe-rich FM layer that still gives PMA, since it would also contribute to a high magnetic moment contribution from that layer - yielding high contrast in the X-ray measurement.

Figure 6-3: The ferromagnetic half-stack with two wedge-deposited layers, shown on the center and right, where the thickness $t$ of Ru and $\text{Co}_{80}\text{B}_{20}(t_{\text{CoB}})$ was varied, respectively.

First, we grew a $\text{Ta}(4\, \text{nm})/\text{Pt}(5\, \text{nm})/\text{Co}_{80}\text{B}_{20}(t_{\text{CoB}})/\text{Ru}(t_{\text{Ru}})/\text{Pt}(5\, \text{nm})$ stack, where
the two layers were grown as wedges (Fig. 6-3). The wedge was probed with a scanning MOKE for perpendicular magnetic anisotropy, and PMA was found for a very narrow range of Ru thicknesses (between 0.7 and 0.9 nm). Thus, the $t = 0.8$ nm thickness as optimal for maximum RKKY AF coupling peak was chosen to optimize for the FM layer thickness. The range of thickness for $\text{Co}_80\text{B}_{20}(t_{\text{CoB}})$ was then found to be between 0.9 and 1.6 nm for PMA (Fig. 6-4).

![Figure 6-4: The square hysteresis loops that resulted for a range of thicknesses of $\text{Co}_80\text{B}_{20}(t_{\text{CoB}})$ in the double wedge ferromagnetic stack.](image)

In order conduct harmonic spin-orbit torque measurements on the sample to probe the material's potential for hosting current-driven or current-generated chiral domain walls and skyrmions, we also confirm that PMA exists for layers where the top or bottom Pt layer will be blocked from contributing any signal. We accomplish this by growing 2 nm of MgO either under the top Pt(5 nm) (Fig. 6.5(a)) or growing a Ta(4 nm)/MgO(2 nm)/Pt($t_{Pt}/\text{Co}_80\text{B}_{20}(1 \text{ nm})$/Ru(0.8 nm)/Pt(5 nm) stack and checking for the thinnest possible Pt that still contributes PMA. Probing the magnitude of the spin-orbit torque in the latter stack would also give insight into the impact of the thin Pt spacer layers that would need to be used in a SAF multilayer suitable for beamtime. PMA was exhibited for a narrow range of $\text{Co}_80\text{B}_{20}(t_{\text{CoB}})$ (in the first stack), between 1.1 and 1.5 nm, and a narrow range of Pt for the latter stack (1.2 nm
< \( t_{Pt} < 1.6 \) nm).

![Diagram of magnetic layers](image1)

![Diagram of magnetic layers](image2)

**Figure 6-5:** (a) The square hysteresis loops that resulted for a range of thicknesses of Co\(_{80}B_{20}\)(t\(_{CoB}\)) in the wedged ferromagnetic stack, with a blocked top Pt layer. (b) The non-zero-remanence loops resulting from the Pt-wedge stack.

One last optimization with Ru involved determining the PMA range for the two types of ferromagnetic layer thicknesses. We grew two wedge samples of the structure Ta(4)/Pt(5)/FM/Ru(0.8)/Pt(2) of varying FM layer thickness (Co\(_{80}B_{20}\) and Co\(_{60}Fe_{20}B_{20}\)). We found that the Co\(_{60}Fe_{20}B_{20}\) has a much more narrow PMA range of 0.6 nm < \( t_{CoFeB} < 1 \) nm. Therefore, we would choose the CoB thickness with minimal coercive field, i.e. the \( t = 1 \) nm for the ferromagnetic structure that would be probed with spin-orbit torque measurements.
6.3 Harmonic Measurements of SAF Films

In this section, we introduce the method for measurement and analysis of current-induced spin-orbit torque in ferromagnetic stacks that are used in SAF multilayers. We use this analysis to show that the magnitude of the spin-orbit torque is large enough in these FM layers to cause current-induced domain wall motion, which has not been observed in the X-ray holography measurements. The discrepancy for these two observations will be discussed.

6.3.1 Spin-orbit torque theory

We can probe the electrical properties of thin films with Hall bar structures, as shown in Fig. 6-7. Sending a current in the perpendicular direction in the magnetized layer will generate a transverse voltage through the Anamalous Hall Efect (AHE). The
measured voltage is proportional to the applied current density, the out of plane magnetization component, and certain material properties. This measured voltage is also linearly scaled with the strength of the spin-orbit torques at moderate current densities ($10^6-10^7$ A/cm$^2$). An in-plane current flowing in a layer film as shown in Fig. 6-8 generates two qualitatively different types of spin-torques: a field like (FL) torque and a damping-like (FL) torque, or $T_\perp \approx m \times y$, and $T_\parallel \approx m \times (y \times m)$. These two torques correspond to the effective fields $B_\perp \sim M \times T_\perp$ and $B_\parallel \sim M \times T_\parallel$, respectively, which are perpendicular to the instantaneous magnetization direction (seen in Fig. 6-8). Studies have shown that the damping-like torque $T_\parallel$ is strong enough to reverse the magnetization of high coercivity ferromagnetic layers with both perpendicular and in-plane anisotropy for current densities on the order of $10^7 - 10^8$ A/cm$^2$, as well as move domain walls.

SOTs induce net effects to the magnetization via inversion symmetry, which can
be induced by sandwiching a ferromagnetic layer between two different non-magnetic layers. Spin Hall Effect produces torques which cancel out in symmetric heterostructures. The full expressions to model the action of field-like and damping-like torques can be derived using methods in literature. These torques are odd and even with respect to the inversion of \( \mathbf{m} \), respectively, and in the first order expansion are given by \( \mathbf{T}_\perp = T_\perp^0 \mathbf{m} \times \mathbf{y} \) and \( \mathbf{T}_\parallel = T_\parallel^0 \mathbf{m} \times (\mathbf{y} \times \mathbf{m}) \), respectively, where \( T_\perp^0 \) and \( T_\parallel^0 \) are first order expansion coefficients.

In this section, we probe the current-induced signal with the injection of an alternate current (ac) and harmonic analysis of the signal. The spin orbit torque due to this current will induce periodic oscillations of magnetization about its equilibrium position. This position is defined by the sum of the effective, external, and demagnetizing fields. The ac current, in the form of \( I = I_0 \sin(\omega t) \) creates a time-dependent oscillating voltage:

\[
V(t) = R(t) I_0 \sin(\omega t)
\]

where \( V(t) \) is either transverse of longitudinal resistance, depending on the geometry of the measurement. In general, we use the Hall voltage \( V^H \) to characterize the magnetization as a function of external field \( B_{\text{ext}} \) and current-induced torques, and it is given by:

\[
V^H = IR^H = R_{\text{AHE}} I \cos \theta + R_{\text{PHE}} I \sin^2 \theta \sin 2\theta
\]

where \( I \) is the current and \( R^H \) is the Hall resistance due to the Anomalous Hall Effect (AHE) and planar Hall effect (PHE). The ordinary Hall effect is omitted since it is negligible in ferromagnetic materials. The AHE and PHE are proportional to the first and second term, respectively, where \( R_{\text{AHE}} \) and \( R_{\text{PHE}} \) are AHE and PHE resistances, \( \theta \) and \( \phi \) are polar and azimuthal angles of the magnetization, respectively. Flowing a moderate ac current \( I_{\text{ac}} = I \exp^{i2\pi ft} \) induces small oscillations of magnetization around its equilibrium position \( (\theta_0, \phi_0) \), defined by anisotropy field \( B_k \) and external
field $B_{ext}$. The first harmonic Hall resistance components are given by:

$$R_{\omega}^H = R_{AHE} \cos \theta + R_{PHE} \sin^2 \theta \sin 2\phi$$  \hspace{1cm} (6.3)

and the second harmonic Hall voltage as:

$$R_{2\omega}^H = \left( R_{AHE} - 2R_{PHE} \cos \theta \sin 2\phi \right) \frac{d\cos \theta}{dB_{ext}} \frac{B_{\theta}}{\sin(\theta_B - \theta)}$$

$$+ 2R_{PHE} \sin^2 \theta \cos 2\phi \frac{B_{\phi}}{B_{ext} \sin \theta_B}$$  \hspace{1cm} (6.4)

where $B_0$ and $B_{\phi}$ are the polar and azimuthal components of the total effective field $B \perp + B \parallel$ induced by current, $\theta_B$ is the polar angle of $B_{ext}$, and $\phi \equiv \phi_0$. From Eqs. 6.3 and 6.4, we are able to measure $B_{\theta}$ and $B_{\phi}$ as a function of $\theta$ and $\phi$. If we make the approximation that $R_{PHE} = 0$, we can evaluate $B_{\theta}$ via:

$$R_{AHE} \frac{d\cos \theta}{dB_{ext}} = \frac{dR_{\omega}^H}{dB_{ext}}$$  \hspace{1cm} (6.5)

If not, we can evaluate $B_0$ and $B_{\phi}$ with a recursive procedure by measuring $V^H$ at $\phi = 0^\circ$ and $90^\circ$, and fitting for $R_{2\omega}^H$ [59]. Using the PHE contribution in Eq. 6.4, and using the substitution in Eq. 6.5, we can use the first and second harmonic signals from the SOT measurement to obtain the effective field:

$$B_{\theta} = I b_\theta \frac{R_{2\omega}^H \sin(\theta_B - \theta_0)}{R_{AHE} \frac{dR_{\omega}^H}{dB_{ext}} \theta_0}$$  \hspace{1cm} (6.6)

where $\frac{dR_{\omega}^H}{dB_{ext} \theta_0}$ is the slope of the first harmonic curve evaluated at $\theta = \theta_0$. Meanwhile, the equilibrium magnetization angle, $\theta_0$, is given by:

$$\theta_0 = \arccos \left| \frac{R_{\omega}^{AHE}(B_{ext})}{R_{AHE}} \right|$$  \hspace{1cm} (6.7)

The original effective field equations imply that $b_\theta = B \perp$ at $\phi = 90^\circ$ and $b_\theta = B \parallel$ at $\phi = 0^\circ$. Neglecting the expansion of the torque equations apart from the fourth order
terms, it can be shown that [59]:

\[ I_{b\theta}(\phi = 0^\circ) = T_{\parallel}^0 + T_{\parallel}^2 \sin^2 \theta + T_{\parallel}^4 \sin^4 \theta, \quad (6.8) \]
\[ I_{b\theta}(\phi = 90^\circ) = -\cos \theta(T_{\perp}^0 + T_{\perp}^2 \sin^2 \theta + T_{\perp}^4 \sin^4 \theta), \quad (6.9) \]
\[ I_{b\phi}(\phi = 0^\circ) = T_{\perp}^0, \quad (6.10) \]
\[ I_{b\phi}(\phi = 90^\circ) = T_{\parallel}^0. \quad (6.11) \]

after which the values of the coefficients $T_{\parallel}^{0,2,4}$ and $T_{\perp}^{0,2,4}$ are obtained by fitting $b_{\theta}$ at $\phi = 0^\circ$ and $b_{\phi}$ at $\phi = 90^\circ$ using Eqns. 6.8 and 6.9. Converting the field dependence into a $\theta$ dependence using the AHE, the fits to the torque coefficients can be performed.

### 6.3.2 Spin-orbit torque analysis of SAF multilayer components

For the SOT analysis we use a set of first and second harmonic Anamalous Hall Effect resistance measurements where the ac current magnitude is large enough for tolerable noise in the sensitive second harmonic response, but small enough so that the sample does not break into domains. We first present the measurement for the damping-like torque in which the current is perpendicular to the applied field direction (Fig. 6-9a), and the angle $\theta_B$ that the external field makes with the normal to the film is $\theta_B = 88^\circ$. We tilt the external field slightly off from $\theta_B = 90^\circ$ in-plane direction in order to prevent the formation of domains. We can see that at large current values (such as $j = 2.6 \times 10^6 A/cm^2$), Joule heating is higher and the effect of current on domain walls is increased. This effect can be lessened with a choice of a larger time constant on the lock-in amplifier, as it effectively averages more data at a given field. These jumps at high current can be effectively discarded in the analysis, but we use a smaller current value because the Joule heating has an effect on the curvature as well.

The field-like torque can be extracted from the measurement when the current direction and the field direction are perpendicular, hence the magnetization oscillates
Figure 6-9: (a) Damping-like torque measurement schematic with indication of external field $B_{\text{ext}}$, magnetization vector $M$, and current $I$ along with current direction. (b-c) First and second harmonic signal, respectively, for three different values of injected current $j$, for radial angle $\phi = 0^\circ$ (damping-like torque measurement).

in the direction of the current, at the heavy metal/ferromagnet interface. Similarly, the intermediate current value is chosen for the Anamalous Hall Resistance first and second harmonic measurements (Fig. 6-10).

Using the AHE resistance obtained from the $j = 2.6 \times 10^6 \text{A/cm}^2$ current value, we can use the information from the two plots calculate the damping-like torque to the first approximation as a fit on the $B_\theta$ vs. $\theta$ plot. To be able to use Eq. 6.6 to evaluate the effective field, we must isolate individual components, such as the first derivative of the first harmonic signal and the second harmonic signal, which we show in Fig. 6-10 as an example, in the case of the damping-like torque calculation.

The planar hall effect (PHE) is then accounted for by considering the symmetric and anti-symmetric components of the AHE resistance at $\phi = 45^\circ$ and $\theta_B = 82^\circ$. Due
to the PHE, which is even with respect to magnetization reversal, the endpoints of the hysteresis loop are asymmetric. Since the AHE is odd with respect to magnetization reversal, the first harmonic contributions of the AHE and PHE, were separated by taking the antisymmetric and symmetric components of $R_i^H$. The PHE saturation resistance itself corresponds to the linear fit of $R_{PHE}^H$ vs. $\sin^2 \theta$. The value of the PHE resistance using this method outlined in [59] is found to be $R_{PHE} = 7.1$ m. Eqns. 6.4 and 6.3 can then used to evaluate the damping-like and field-like effective fields, in relation to their dependence on the magnetization angle $\theta$ (Fig. 6-12). The fit to these curves then yields the respective effective torque values.
Figure 6-11: (a-b) First harmonic and its first derivative, as well as the second harmonic signal, respectively, for the damping-like torque.

Figure 6-12: (a) \(B_\parallel\) measured at \(\phi = 0^\circ\) as a function of \(\theta\). The solid line is fit to \(T_\parallel^0 + T_\parallel^2 \sin^2 \theta + T_\parallel^4 \sin^4 \theta\) according to Eq. 6.8. (b) \(B_\perp\) measured at \(\phi = 90^\circ\). The solid line is fit to \(T_\perp^0 + T_\perp^2 \sin^2 \theta\) according to Eqn. 6.9.

The approximated damping-like torque in the first order (\(T_\parallel^0\)) for this sample, taking into account the current density in the Hall cross geometry) is \(T_0 = 4.3 \text{ mT}/(10^7 \text{A/cm}^2)\), which yields a sufficient spin-hall angle for domain wall motion via SOT and is on the same order of magnitude to literature values of SOTs in CoFeB/MgO ferromagnetic materials [59] and Pt/FM/Ru and Pt/FM/MgO structures [65].
6.3.3 X-ray holography measurements of SAF films

During the X-ray holography measurement of SAF stacks, we applied a current pulse to the film after saturation to attempt to move the domains in the ferromagnetic phase. At this point the Co$_{60}$Fe$_{20}$B$_{20}$ layers exhibit domains while Co$_{80}$B$_{20}$ layers are uniformly magnetized. We are able to selectively tune to either the Fe or Co element edge, and observe clear contrast in these structures (Fig 6-13). To attempt to observe domain wall motion, we apply current unipolar pulses of $1.6 \times 10^{12}$ A/m$^2$ (8.6 V transmitted voltage pulse) of alternating polarity (and thus, direction along the film).

![Figure 6-13: Schematic of SAF multilayer structure in X-ray holography measurement in this section, along with the reconstructed images corresponding to imaging at the (left) Co and (right) Gd edges. $J_{AF}$ is the AF coupling coefficient, and $N$ is the number of layers, $N = 24$ in this case.]

After sending more than 20 consecutive such pulses, the domains do not tend to move in accordance to the direction of the applied pulse (as seen in the first seven pulses sent, shown in Fig. 6-14). The motion is entirely random. Thus we conclude that the pulses have no effect other than heating. That is, we can observe domain wall motion after current pulses, but the direction of motion is not (or very little) correlated to the direction of the pulse. We always applied a single unipolar pulse between two consecutive images, and observe a domain wall motion (as indicated by the arrow). The lack of current-induced domain wall motion indicates a low spin-orbit torque through the ferromagnetic layer, and could be due to the thickness of the Pt
layer spacer (0.8 nm) being too low to provide a sufficient spin-orbit torque.

![Figure 6-14: Image (1) shows the original state of the magnetic membrane, and images (2)-(8) show the state right after a unipolar pulse of alternating polarity for each image. The arrow shows the direction of the domain motion. The magnetic structure is a wire (running from top to bottom in the image) of 900 nm width with a notch at the left side.]

6.4 Further studies

A possible way to continue to optimize the Ru-based SAF stack would be to investigate the effect that the small spacer layers have on the net spin orbit torque through the FM layer, and if it is able to be sustained in a stack with a high number of layers.

We can further attempt to optimize the anti-ferromagnetic switching field by changing the Ru heavy metal to Ir, which should have a higher AF exchange field contribution. I grew Ta(5)/Pt(4)/CoFeB(1)/Ir(0.5)/Pt(t_{Pt})/CoFeB(1)/Ir(0.5)/Pt(2) film at two similar thicknesses of Ir, and find that the material has PMA and a AF switching field of about 180 mT (6-15). We can use this information to optimize these stacks for maximum exchange coupling field, or the field at which the loop jump occurs and the AF domains (with M_s = 0) switch to an FM state. Tuning the thickness of Ir around its optimal thickness (0.5 nm) for RKKY coupling can tune the AF coupling field. Subsequently optimizing the thickness of the Pt spacer layer, t_{Pt},
we can modulate the exchange coupling field to a desired value. Greater Pt content will decrease the exchange coupling due to increasing the distance to the Ir layer.

After determining that PMA existed for a Ta(4)/Pt(5)/Co\textsubscript{60}Fe\textsubscript{20}B\textsubscript{20}(t)/Ru(0.8)/Pt(2), we chose the intermediate Co\textsubscript{60}Fe\textsubscript{20}B\textsubscript{20} thickness of 1 nm to grow the full SAF stack of only one repetition. I therefore grew Ta(5)/Pt(4)/Co\textsubscript{60}Fe\textsubscript{20}B\textsubscript{20}(1)/Ir(0.5)/Pt(0.8)/Co\textsubscript{60}Fe\textsubscript{20}B\textsubscript{20}(1)/Ir(0.5)/Pt(2). The resulting AF exchange coupling field, as seen in the hysteresis loops, is 1.8 T—just a little larger than in magnitude than that of one repetition of the SAF stack with the Ru coupling layer that comprised the layers in the holography experiment (Fig. 6-15b,d). The loop exhibits switching from AF to FM domains at positive and negative fields at this value. For further optimization of this stack for spin-orbit torque magnitudes, Fig. 6-16 shows the range of magnetic layer for which the Ir-coupled SAF stack would have PMA. The opposite moments of the FM layers (Co\textsubscript{60}Fe\textsubscript{20}B\textsubscript{20} and Co\textsubscript{80}B\textsubscript{20}) is key for engineering the AF-coupled multilayer that could be imaged with holography techniques.
Figure 6-15: (a) Schematic of single layer SAF structure with Ir as the AF coupling layer, with \( J_{AF} \) the coupling coefficient. (b) VSM switching loop with coupling field of \( B = 1.8 \) T. (c) Schematic of the single layer SAF structure with Ru as the AF coupling layer, with \( J_{AF} \) the coupling coefficient. (d) A switching loop with coupling field of \( B = 1.7 \) T obtained by F. Büttner.
Figure 6-16: (a) The ferromagnetic half-stack with an Ir AF-coupling layer and a wedge-deposited FM layer $Co_{80}B_{20}(t_{CoB})$ showing the range for PMA. (b) (a) The ferromagnetic half-stack with an Ir AF-coupling layer and a wedge-deposited FM layer $Co_{60}Fe_{20}B_{20}(t_{CoFeB})$ showing the range for PMA.
Chapter 7

Summary

In this thesis, we have presented magnetic skyrmions as a subject of study in the interest of possible use as bits in magnetic memory devices, and shown the need for the optimization of magnetic thin film properties in order to host these devices. We show this through the tunability of film properties by optimizing parameters such as film thickness and composition.

To emphasize the importance of predictive simulations in the design of thin film, we have shown micromagnetic simulations that support previous experimental findings of skyrmion bubble collapse under in-plane and out-of-plane fields, and predict a regime for skyrmion stability. The distribution effect on collapse with increasing in-plane fields, as seen in experiments, can be possibly explained by the lesser impact of thermal fluctuations and pinning on as DWs widen. The impact of zero DMI (Bloch skyrmion) is seen in the domain wall broadening and higher domain wall energy change with the application of in-plane field. The larger change in DW energy allows this skyrmion to collapse much easier at high in-plane fields.

To demonstrate high skyrmion stability and small bit size in a real material, we use X-ray holography to generate ultrasmall skyrmion bubbles in ferrimagnetic material. We subsequently grow synthetic antiferromagnetic (SAF) material with a range of magnetic (mainly perpendicular magnetic anisotropy) and electrical properties (spin orbit torque magnitude) that would allow for skyrmion stability and chiral domain wall motion. Furthermore, the ferromagnetic stacks that are grown have potential to
be incorporated into full SAF stacks. The optimization of other parameters such as $K_u$ and DMI could pave the way to generating and efficiently moving spin textures such as skyrmions in these materials.
Bibliography


