

To the Graduate Council:

I am submitting herewith a dissertation written by Nan Tang entitled “Non-collinear Magnetic Textures Studied by Neutron Scattering.” I have examined the final paper copy of this dissertation for form and content and recommend that it be accepted in partial fulfillment of the requirements for the degree of Doctor of Philosophy, with a major in Material Engineering.

Dustin A. Gilbert, Major Professor

We have read this dissertation
and recommend its acceptance:

Philip D. Rack

Haixuan Xu

Lisa M Debeer-Schmitt

Accepted for the Council:

Dixie L. Thompson

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(Original signatures are on file with official student records.)

Non-collinear Magnetic Textures Studied by Neutron Scattering

A Dissertation Presented for the
Doctor of Philosophy
Degree

The University of Tennessee, Knoxville

Nan Tang

May 2023

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to my parents Hongming Nan and Xuping Tang and my wife Zhonghui Li

Acknowledgements

First and foremost, I would like to express my deepest gratitude to my principal advisor, Prof. Gilbert, for his invaluable guidance and mentorship throughout my PhD journey. His expertise, patience and encouragement have been instrumental in shaping my intellectual growth and helping me navigate the complexities of this research. It is even more remarkable that Prof. Gilbert has become not just a principal investigator, but also a friend, contributing to the positive and enjoyable lab environment. I really appreciate it and I will never forget those valuable moments in the past five years.

I extend my heartfelt thanks to the members of my dissertation committee, Prof. Rack, Prof. Xu and Dr. DeBeer-Schmitt, for their constructive comments, thorough reviews, and valuable suggestions. Their combined expertise has greatly enhanced the quality and rigor of this work.

Special thanks go to Dr. Grutter and Dr. Borchers, who have generously offer their expertise that has been essential to the completion of this research. I also appreciate the financial support provided by DOE through Grant No. DE-SC0021344, which has allowed me to focus on my research without undue stress.

I am immensely grateful to my family for their unwavering love, understanding, and support throughout my academic journey. To my parents, Hongming and Xuping, thank you for instilling in me the value of education and the importance of perseverance. To my spouse, Zhonghui, your love, encouragement, and belief in me have been a constant source of strength.

Lastly, I would like to thank my friends Namila, Cameron, Ziang, Dylan and so on, who have stood by me and provided emotional support during the highs and lows of this journey.

Your companionship, laughter, and shared experiences have made this challenging time more manageable and enjoyable.

In conclusion, I dedicate this dissertation to all those who have contributed in some way, big or small, to my academic and personal growth. Without the collective support of my mentors, colleagues, family, and friends, this achievement would not have been possible. Thank you all.

Abstract

Non-collinear magnetic structures, where the magnetic moments do not align along a single axis, can lead to interesting physical phenomena and potential device applications. In this dissertation, two specific non-collinear magnetic textures are studied which includes soft/hard bilayer and skyrmions. Soft/hard magnetic bilayer thin films have been widely used in data storage technologies and permanent magnet applications. Here, we use polarized neutron reflectometry (PNR) to study magnetic configuration in soft-hard bilayer heterostructure thin films designed with different sample geometry and material properties under a range of temperatures and fields. Comparing the PNR results to the micromagnetic simulations reveals that the interfacial magnetic configuration is highly dependent on thickness and saturation magnetization of soft layer materials and external factors (field and temperature) and has a relatively weak dependence on hard layer properties. Magnetic skyrmions, another example of non-collinear structures, exhibit unique, technologically relevant pseudo-particle behaviors which arise from their topological protection, including well-defined, three-dimensional dynamic modes that occur at microwave frequencies. In this work, we use small angle neutron scattering (SANS) to capture the dynamics in hybrid skyrmions and investigate the spin wave structure. Performing simultaneous ferromagnetic resonance and SANS, the diffraction pattern shows a large increase in low-angle scattering which is present only in the resonance condition. This scattering pattern is best fit using a mass fractal model, which suggests the spin waves form a long-range fractal network. Overall, this dissertation contributes to a deeper understanding of the complex behavior and potential applications of non-collinear magnetic structures, particularly in soft/hard bilayer heterostructures and magnetic skyrmions.

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Chapter 1

Introduction

Non-collinear spin textures are fundamentally interesting and offer unique opportunities for a variety of magnetic technologies. One specific example of a magnetic technology which leverages non-collinear magnetism is perpendicularly coupled magnetic soft-hard bilayer thin films. This particular structure plays an important role for permanent magnet application and data storage device. In this system the magnetically soft layer - meaning it has low intrinsic anisotropy - couples to a magnetic hard layer through an interfacial exchange coupling. Away from this interface, the magnetic soft layer can reorient in response to either an applied magnetic field or its own magnetostatic interaction, resulting in a non-collinear magnetic structure. The non-collinear structure can be used to nucleate a domain wall, facilitating premature reversal of the hard layer, improving writability in magnetic hard drives. Another specific example is magnetic skyrmions, which are a specialized magnetic configuration in-which the moment form a closed coplanar looped structure. This magnetic looped structure bestows a topological protection, which in turn gives the skyrmion pseudo-particle behaviors and provides huge potential in spintronics device application.

My dissertation research focuses on these two magnetic structures. In this first chapter I start by providing a general introduction to magnetism and magnetic behavior. Next, the common magnetic interactions are summarized, which are fundamental in determining the micromagnetic structures. I finish the introduction with a brief discussion about magnetic domains and domain wall structures. With this background established, To close out the

chapter I introduce the subjects of this dissertation, magnetic soft/hard bilayer films and magnetic skyrmions.

1.1 Magnetic Moment and Magnetic Ordering

Magnetism, in a simple description, is the collective behavior of the magnetic spin moments within a material. The magnetic moment is a fundamental property of electrons, similar to charge, and is directly related to their angular momentum. The electron angular momentum has two origins: (1) the orbital motion of electrons bounded to a positive-charged nucleus and (2) the intrinsic spin as a unique inseparable property of the electron. In a single hydrogen atom model, an electron (charge $-e$ and mass m_e) is bounded to a proton via Coulomb interaction. From a classical perspective, this electron will process in a path which circulates around the nucleus (for a simplification, the path will be circular, with radius r and velocity v). The angular momentum of this circular motion is $\hbar = m_e v r$. The $\hbar$ is reduced Planck constant, which indicates the angular momentum is quantized. In addition, the moving electron acts as an effective loop of current, $I = -\frac{ev}{2\pi r}$. This current generates a magnetic dipole field, $\mu = I\pi r^2 = -\frac{e\hbar}{2m_e} = -\mu_B$, where μ_B is Bohr magneton. Thus, for the orbital case, the coupling between angular momentum and magnetism is achieved through the electrons motion. The hydrogen atom model demonstrates the origin of magnetic moments in a semi-classical way, while a more accurate way is to consider the “state” in a quantum way. Under spherical symmetry, the state of a bounding electron can be separated into radial, azimuthal and polar components. The state of the azimuthal and polar components is associated with the variables $|l, m_l\rangle$, where l and m_l are the angular quantum number and magnetic quantum number, respectively. Using an orbital angular momentum operator ($\hat{\mathbf{L}}$) and a z component of orbital angular momentum operator ($\hat{\mathbf{L}}_z$) to measure this state, the corresponding equation are $\hat{\mathbf{L}}^2|l, m_l\rangle = l(l+1)|l, m_l\rangle$ and $\hat{\mathbf{L}}_z|l, m_l\rangle = m_l|l, m_l\rangle$, respectively. The magnitude of orbital angular momentum is $\sqrt{l(l+1)}\hbar$ with a z component of $m_l\hbar$, where m_l is the integer between $[-l, l]$. The corresponding magnetic moments of these two angular momentum are $\sqrt{l(l+1)}\mu_B$ and $m_l\mu_B$. From semi-classical and quantum way, we briefly explain the relationship between orbital angular momentum and magnetic moments.

However, in most magnetic materials constructed from $3d$ transition metals, the magnetic moments from orbital momentum are quenched due to the symmetry of electronic wave functions. As a result, the magnetic moments are mostly from electronic spins. Spin is an inherent property of the electron and also corresponds to an effective angular momentum; classical introduction to spin usually makes the analogy of the electron charge on the surface of a spinning sphere, and makes a similar analogy to a moving current, however this is demonstrably false. The spin of an electron can be determined by applying the spin operator ($\hat{\mathbf{S}}$) and the z component spin operator ($\hat{\mathbf{S}}_z$). Using these operators to act on a spin state (ψ), we have $\hat{\mathbf{S}}^2|\psi\rangle = s(s+1)|\psi\rangle$ and $\hat{\mathbf{S}}_z|\psi\rangle = s|\psi\rangle$, where s is a spin quantum number with a value of $\frac{1}{2}$. The corresponding total spin moments are $\sqrt{s(s+1)}g\mu_B$ while the intrinsic spin moment (observed from z direction) is $m_s g\mu_B$, where $g = 2$, $s = 1/2$ and $m_s = \pm\frac{1}{2}$. The g here is gyromagnetic ratio which is a dimensionless number which is ≈ 2 for an electron. Both the orbital and spin angular momentum contribute to the magnetic moments, which are quantized number of μ_B . Compared with the orbital momentum, the spin part plays a more important role in determining the magnetic behaviors of $3d$ materials.

The Hydrogen atom model demonstrates the relationship between the angular momentum and magnetism. Next, the response to an applied magnetic field is considered. We assume the magnetic field ($\mathbf{B}$) is along the z direction, which can be represented as a gauge $\mathbf{A}(\mathbf{r}) = \frac{1}{2}(\mathbf{B} \times \mathbf{r})$. The generalized Hamiltonian of this system can be written as $\hat{\mathcal{H}} = (\frac{[\mathbf{p} + e\mathbf{A}(\mathbf{r})]^2}{2m_e} + V) + g\mu_B \mathbf{B} \cdot \mathbf{S} = \hat{\mathcal{H}}_0 + \mu_B (\mathbf{L} + g\mathbf{S}) \cdot \mathbf{B} + \frac{e^2}{(8m_e)} \mathbf{B} \times \mathbf{r}$, where the first term $\hat{\mathcal{H}}_0$ is the Hamiltonian of the hydrogen atom model and the second and third terms contribute to paramagnetism and diamagnetism, respectively; paramagnetism is the quality that the magnetic polarization aligns with an applied magnetic field while diamagnetism is a magnetic response which counters an applied field. Generally, paramagnetic materials exhibit a much larger magnetic response compared to diamagnetic materials. In both paramagnetism and diamagnetism an external magnetic field is required to achieve polarization.

For the Hydrogen atom model discussed above (without and with an external magnetic field) the magnetism was shown to originate from a single electron in orbit, without interactions with other electrons. In this case, the magnetic orientation (polarization) is determined by an applied magnetic field which breaks the spatial symmetry. However, for

ferro and antiferromagnetism, the interaction between electrons is the key to the magnetic ordering and cannot be ignored. Here, the Helium atom model is used to demonstrate the coupling between two electrons. The wave function of these two electrons can be written as $\Psi = \psi(\mathbf{r}_1, \mathbf{r}_2)\chi(\mathbf{r}_1, \mathbf{r}_2)$, where $\psi(\mathbf{r}_1, \mathbf{r}_2)$ and $\chi(\mathbf{r}_1, \mathbf{r}_2)$ are spatial and spin parts, respectively. Notably, the electrons satisfy Fermi-Dirac statistic; using a permutation operator $\hat{\mathbf{P}}$ to measure this state results $\hat{\mathbf{P}}\Psi = -\Psi$; the eigenvalue equals to -1 which indicates the Ψ is asymmetric. Therefore, either spatial or spin function is asymmetric. For the ground state of a Helium atom, the spatial part is symmetric; two electrons occupy same energy level at $1s$ orbit. As a consequence, the spin part must be asymmetric — the two spins have opposite orientations, resulting in a total spin quantum number $s_{tot} = 0$. In an excited state, the electrons can occupy $1s$ and $2s$ orbitals, respectively, thus the spatial wave is asymmetric. Consequently, the spin part function should be symmetric—two spins are along same direction - the joint spin quantum number $s_{tot} = 1$. Based on $(\hat{\mathbf{S}}_1 + \hat{\mathbf{S}}_2)^2 = (\hat{\mathbf{S}}_1)^2 + (\hat{\mathbf{S}}_2)^2 + 2\hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2$ ($\hat{\mathbf{S}}_1$ and $\hat{\mathbf{S}}_2$ are spin operators for electrons at position $\mathbf{r}_1$ and $\mathbf{r}_2$, respectively), the eigenvalue of $(\hat{\mathbf{S}}_1 + \hat{\mathbf{S}}_2)^2$ equals to $s_{tot}(s_{tot} + 1)$. As mention above, the eigenvalue of $\hat{\mathbf{S}}_1^2$ and $\hat{\mathbf{S}}_2^2$ is $s(s + 1)$, therefore, we can calculate the eigenvalue of $\hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2$ which equals to $-\frac{3}{4}$ (singlet state) or $\frac{1}{4}$ (triplet state). As a result, we can write the Hamiltonian of a Helium atom as $\hat{\mathcal{H}} = \frac{1}{4}(E_{gs} + 3E_{es}) - (E_{gs} - E_{es})\mathbf{S}_1 \cdot \mathbf{S}_2$, where $\mathbf{S}_1 \cdot \mathbf{S}_2$ is the eigenvalue of $\hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2$ and E_{gs} and E_{es} are the ground state and excited state energies, respectively. The first term of the Hamiltonian corresponds to the Coulomb-based repulsive interaction between two electrons, while the second term is caused by the exchange of two electron states. The Hamiltonian term of spins can be written as $\hat{\mathcal{H}}^{spin} = -2J\mathbf{S}_1 \cdot \mathbf{S}_2$, where exchange constant $J = \frac{E_{gs} - E_{es}}{2}$. When $J > 0$ ($J < 0$), the energy favors a parallel (anti-parallel) orientation of the spins, which results in ferromagnetic (antiferromagnetic) ordering. The spin Hamiltonian term demonstrates the exchange coupling between two electrons, which explains the magnetic behaviors in an atom. Moreover, this energy term persists and underpins magnetic interactions between adjacent atoms in solid state systems, resulting in ferromagnetic and antiferromagnetic materials. The exchange coupling plays a fundamental role in determining the magnetic ordering.

The above models illustrate the relationship between electrons and magnetic moments. Under an external field, the electrons intrinsically have both paramagnetic and diamagnetic properties, but the paramagnetism is often dominant. In addition, the exchange interaction between electrons is important for magnetic ordering in ferromagnetic and antiferromagnetic materials. Understanding the behavior of electrons is crucial for studying magnetism and the magnetic properties of materials.

1.2 Magnetic Energy

Within materials, the ultimate spin configuration is determined by the competition between a variety of magnetic interactions. As discussed in the Helium atom model, the exchange interaction is fundamental in determining the local ferromagnetic or antiferromagnetic ordering. This interaction is the result of wave function overlap between adjacent atoms and so plays a dominant role over short distances (several Å) but significantly decreases as the distance increases following an exponential trend. A second energy term called the dipolar or magnetostatic interaction, on the other hand, is long-ranged and decreases more slowly with distance (following a power law), which makes it important in determining the magnetic ordering over larger distances. A third energy term is called the magnetocrystalline anisotropy. This energy originates from the broken symmetry in the crystal lattice, which can determine specific directions favored by magnetization. Beyond the above interactions, the external condition cannot be ignored, which includes the temperature and magnetic field. The temperature disrupts local coherent ordering following standard thermodynamics. In addition, the induced thermal fluctuation can result the change of states, which can excite spin waves. The external field contributes to the Zeeman energy, which breaks the spatial symmetry and creates a favored direction for magnetization to orient. All of these interactions compete with each other, which results in a wide range of magnetic textures and behaviors.

1.2.1 Exchange Interaction Energy

The exchange interaction originates from the exchange of electronic states, which is fundamental of magnetic ordering. The Hamiltonian of this interaction can be written as $\hat{\mathcal{H}}^{spin} = -\sum_{(i,j)} \frac{1}{2} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$, where $\mathbf{S}_i$ and $\mathbf{S}_j$ are spins for atom i and j , respectively, and J_{ij} is the exchange constant between atoms (i and j). Based on whether two magnetic atoms are neighboring or not, the exchange interaction can be direct or indirect.

Direct Exchange Interaction

For direct exchange, the interaction between non-neighboring atoms is ignored. Assuming the exchange constant is uniform between atoms, the Hamiltonian of spin can be rewritten as $\hat{\mathcal{H}}^{spin} = -JS^2 \sum_{(i,j)} \cos\phi_{ij}$, where J is the exchange constant and ϕ_{ij} is the classical spin angle between each of 2 nearest atoms. For $J > 0$, the energy can be minimized when $\cos\phi_{ij} = 1$. The spins align in same direction, which corresponds to a ferromagnetic order. For $J < 0$, the energy is favored when $\cos\phi_{ij} = -1$. The neighboring spins are aligned in an anti-parallel direction, forming an antiferromagnetic ordering. The direct exchange interaction plays an important role to determine the ferromagnetic or antiferromagnetic ordering in metals.

Indirect Exchange Interaction

The indirect exchange interaction is most often considered metal oxides and rare earth metals. In the former case, the nearest neighbors are frequently not magnetic; in the latter the magnetic interactions are mediated by the conduction electrons. The indirect exchange interaction mainly includes super exchange, double exchange, Dzyaloshinsky-Moriya (DM) and RKKY (Ruderman–Kittel–Kasuya–Yosida) interactions.

Super Exchange Interaction For the super exchange interaction, the coupling between two magnetic atoms is achieved through a non-magnetic intermediary. One example is the magnetic order in MnO [Anderson \(1950\)](#). In this material the two Mn^{+2} ions are coupled through an O^{2-} ; this material is an insulator, so the electrons are localized to the metal ions, disallowing direct exchange of spin information. However, through polarization of

the oxygen orbitals, the Mn ions order antiferromagnetically. The oxygen-bridged super exchange interaction is also observed in CuO [Shimizu et al. \(2003\)](#) and Fe₂O₃ [Gilleo \(1958\)](#), while Fluorine [Reger et al. \(2012\)](#) or Arsenic-bridged [Ma et al. \(2008\)](#) ceramics is also reported.

Double Exchange Interaction In some ceramics, distributions in the local cation or anion species results in a distribution in the valence of the other atoms. This distribution in valence encourages delocalization, which is enabled through ferromagnetic ordering. One such example is La_{1-x}Sr_xMnO₃ [Blundell \(2003\)](#), which contains trivalent La and divalent Sr. These cations are ridged in their valence state, as is the oxygen. The mixed cation species leaves a variable valence to be taken by the more flexible Mn ion. Since the charge of the electron cannot be divided, the material contains both Mn⁺³ or Mn⁺⁴ ions to achieve the necessary charge balance. These two ions can be coupled through hopping of electrons. Energy is minimized when the electrons on an atom have the same spin orientation (reducing the wave function overlap, known as Hund’s first law), thus, to encourage hopping, atoms align ferromagnetically. The double exchange interaction provides a ferromagnetic counterbalance to the antiferromagnetic superexchange interaction, the balance of which determines the magnetic ordering in oxides.

DM Interaction The DM interaction often arises from spin orbit coupling and generally takes the form $\mathbf{D} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$, where $\mathbf{D}$ is the Dzyaloshinskii vector. The cross product implies that the minimum energy configuration has the moments oriented perpendicular, in direct competition with the other exchange interactions. Furthermore, the cross product results in a vector, meaning the DM interaction has a directionality and thus requires broken spatial symmetry to manifest. This type of symmetry breaking appears in non-centrosymmetric crystal structures, such as the B20 structure and their magnetic materials MnSi and FeGe. The balance of the direct exchange (colinear) and DM interactions results in the magnetization forming looped structures. These looped structures tend to possess unique magnetic phenomena which vary significantly compared to traditional magnets. One of these looped structures is the magnetic skyrmion, a topologically protected chiral soliton with

potential applications to corruption-resistant data storage applications; the formation and stabilization of skyrmions and skyrmion lattices, chiral magnetic textures, and topological magnetism, which provides new insights to material applications. Therefore, understanding the behavior of magnetic materials under the influence of the DM interaction is essential for the study and development of advanced magnetic materials and technologies.

RKKY Interaction In some rare earth based materials [Yafet \(1987\)](#); [Miyazaki et al. \(2020\)](#), magnetic atoms can be coupled through the conduction electrons. The exchange interaction follows $J_{RKKY}(r) = \frac{\cos(2k_F r)}{r^3}$, where k_F is the Fermi sphere radius. This equation emphasizes the transition between ferromagnetic to antiferromagnetic ordering with the distance between atoms, r . Compared with direct exchange interaction, the RKKY is a long ranged interaction which decreases following a power law. In order to satisfy this oscillatory interaction, the magnetic moment in these materials frequently forms a spin spiral (or spin helix). Therefore, the RKKY contributes to the formation of long-ranged and noncollinear magnetic ordering.

1.2.2 Dipolar Interaction Energy

The magnetic dipolar energy can be written as $E = \sum_{(i,j)} \frac{\mu_0}{4\pi r^3} [\mu_i \cdot \mu_j - \frac{3}{r_{ij}^2} (\mu_i \cdot \mathbf{r}_{ij})(\mu_j \cdot \mathbf{r}_{ij})]$, where μ_i and μ_j are magnetic moments, and $\mathbf{r}_{ij}$ is space vector separating i and j . Assuming two parallel magnetic dipoles with a distance of 1 Å, the corresponding energy is about 10^{-23} J similar to k_B , where k_B is the Boltzmann constant. The dipolar energy is too small to explain the origin of the room temperature ferromagnetic metals (Fe, Co and Ni), but it is important at low temperatures and can explain the ferromagnetic or antiferromagnetic behavior of some 4f ceramics, e.g. the antiferromagnetic behavior of GdPO₄ ($T_N=0.9$ K) [Palacios et al. \(2014\)](#). In addition, the dipole-dipole interaction can be long ranged, which decreases following a power law (exponential law for exchange interaction). This long-ranged interaction is important to determine the magnetic configurations in thin film materials, which contribute to stabilization of magnetic domains [Desautels et al. \(2019\)](#), skyrmions [Desautels et al. \(2019\)](#); [Ezawa \(2010\)](#) and vortex [Liu et al. \(2018\)](#).

1.2.3 Anisotropy Energy

In some bulk or multi-layer thin film materials, the magnetization preferably aligns along specific crystalline directions (easy axes) and resists alignment along other directions (hard axes). The energy difference between the magnetization alignment along the easy and hard axes is defined as magnetic anisotropy. The magnetic anisotropy originates from the spin-orbit interaction; the corresponding Hamiltonian can be written as $\hat{\mathcal{H}} = -\lambda \mathbf{L} \cdot \mathbf{S}$, where $\mathbf{L}$ and $\mathbf{S}$ are orbital and spin angular momentum. This spin-orbit interaction can cause the energy level of orbitals to split and the electrons will fill the low energy level first. This preferential filling of orbitals can lead to an unbalanced distribution of electrons in different directions, which in turn generates magnetic anisotropy.

The generalized form of the magnetic anisotropy is written as $E_{ani} = (K_u + K_s)\sin^2\theta + (K_u + K_s)\sin^4\theta$, where K_u and K_s are the magnetic crystalline and shape anisotropy, respectively, and θ is the angle between the magnetization and the easy axis. The K_s relates to the energy difference of the total dipolar energy along easy and hard axes. A simple example to understand K_s is to consider the energy of two dipoles, shown in Figure 1.1. The energy is minimized when two dipoles and r_{ij} are all parallel, while the energy is maximized for two parallel dipoles vertical to r_{ij} . This is because in the former case the dipolar field reinforce their mutual alignment, while in the latter fir dipolar field resists their mutual alignment. The K_s is based on the arrangement of dipoles while the K_u is largely correlated to the atomic bonding, where the easy axis is usually along stronger bonding directions. However, in most 3d bulk materials, the orbital magnetic moments are quenched due to the spatial symmetry of orbital wave function. At room temperature, the easy axes are [100], [0001] and [111] for Fe (BCC), Co (HCP) and Ni (FCC), respectively, which are based on K_s . For some crystal structures, such as $L1_0$ -FePt, the tetragonal distortion results in a magnetic easy axis; the $L1_0$ structure is a layered face centered tetragonal structure in-which the magnetic easy axis is the (001) direction. Also, in multi-layer heterostructures such as Co/Pt, Co/Pd, or GdFe, an interfacial coupling agains breaks the symmetry resulting in the direction along the film normal direction becoming the magnetic easy axis.

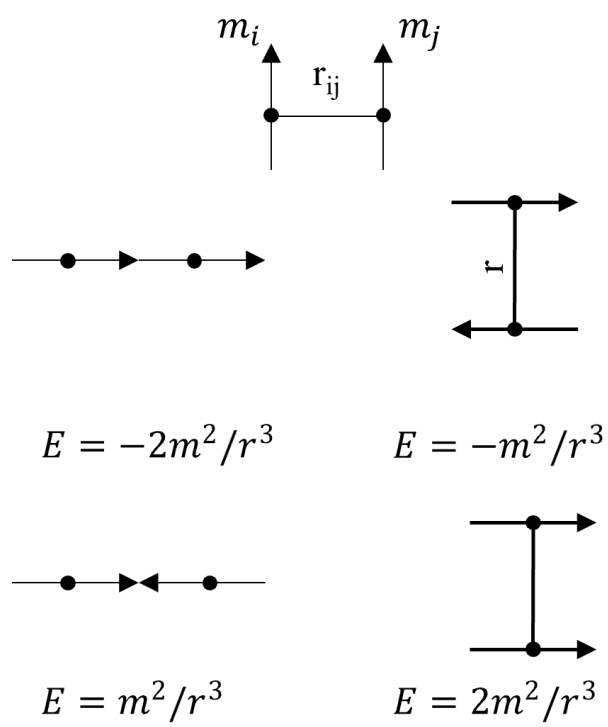


Figure 1.1: two dipoles energy

1.2.4 Thermal Energy

The thermal energy can be calculated as $E = k_B T$. Using classical thermodynamics, the thermal energy causes statistical changes in the state of a system following the Boltzmann distribution function $\exp(\frac{\Delta E}{k_B T})$, where ΔE corresponds to the energy change from initial state to final state. This function demonstrates the possibility of a thermal fluctuation causing a system to change between states with energy difference at a specific temperature. For magnetic materials, the most apparent manifestation of this is the Curie temperature T_C . Above T_C the thermal fluctuations dominate the magnetic orientation, resulting in a random and constantly changing configuration. Cooling through the Curie temperature, the exchange energy dominates and freezes in a stable configuration in-which the magnetic moments are able to align with their neighbors. Another example is the temperature dependent saturation magnetization. In most magnetic materials, as T ($T < T_C$) increases, the saturation magnetization decreases; the increasing temperature induces strong fluctuation to the magnetization, which increases the statistical probability of spontaneous flipping a spin. As a result, the overall magnetic moment of the material is reduced at high T .

1.2.5 Zeeman Energy

The Zeeman energy is written as $E = -\mathbf{H}_{\text{ext}} \cdot \mathbf{M}$, where $\mathbf{H}_{\text{ext}}$ is the external magnetic field and $\mathbf{M}$ is the magnetic moments. The Zeeman energy indicates the energy is minimized when $\mathbf{M}$ and $\mathbf{H}_{\text{ext}}$ are parallel and the energy is maximized when $\mathbf{M}$ and $\mathbf{H}_{\text{ext}}$ are anti-parallel.

1.3 Magnetic Domains and Domain Walls

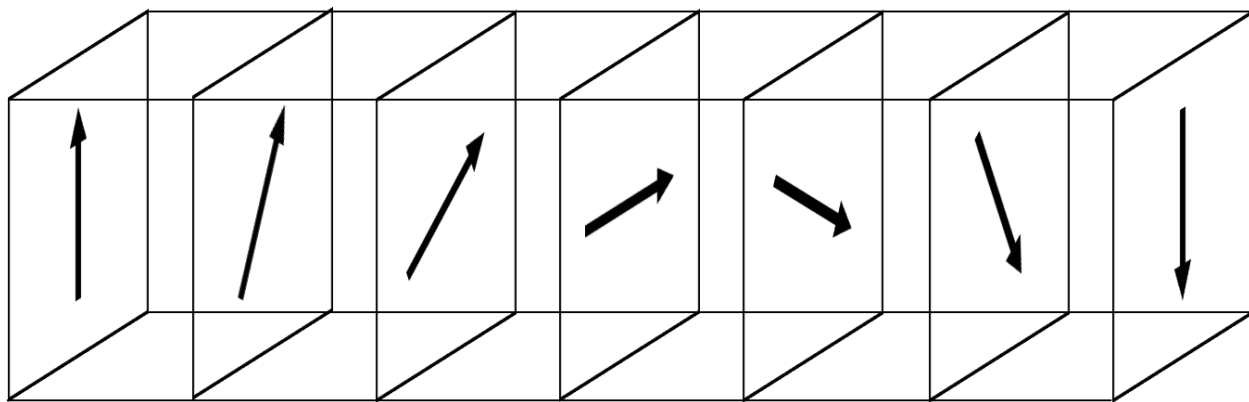
For a ferromagnetic material under a large magnetic field the Zeeman energy tends to dominate the magnetic orientation and form the saturated state, in-which all the magnetic moments are aligned parallel. However, as the field is removed, the balance of these other magnetic energies tends to break the system up into magnetic regions called domains, which are each ferromagnetic and have a saturated orientation within the domain boundary, but the orientation is different between adjacent domains. The boundary between domains is

called the domain wall and is formed due to competition between the magnetic energies. One simple example is the Bloch domain wall, in-which the magnetization rotates vertical to the wall propagation direction, shown in Figure 1.2a. The magnetization can also rotate in the same plane, which forms a Néel domain wall, shown in Figure 1.2b. These two domain wall structures usually originate from the energy competition between the exchange and anisotropy energies and the dipolar interactions. The exchange energy can be minimized when the domain wall width is large, but this increases the anisotropy and dipolar energies. On the other hand, making a very narrow domain wall rapidly increases the exchange energy. The balance of these interactions confines the domain wall length.

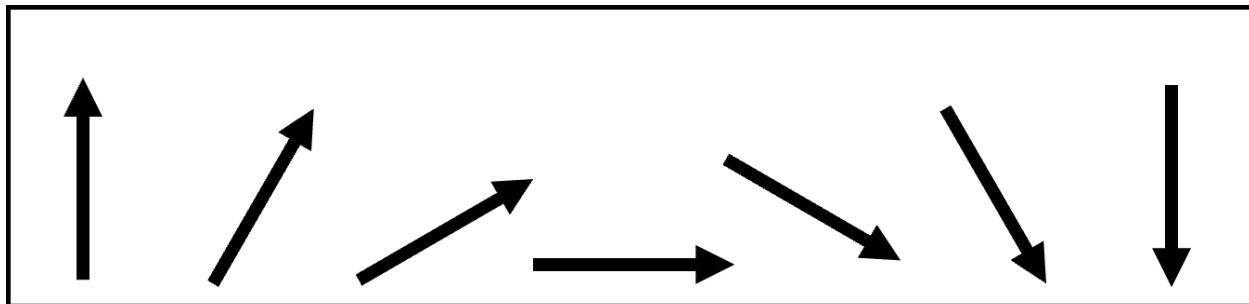
The domain walls in magnetic thin films can be further complicated by reduced dimensionality and short-range proximity effects. Considering a bilayer film consisting of a magnetic soft layer built on a hard layer, the properties (saturation magnetization, anisotropy and thickness) of each layer can be totally different. However, due to the overwhelming strength of the direct exchange interaction at short distances, the magnetic configuration at the interface is rigidly locked. This allows the properties of one layer to influence the behavior of the second. Since the interfaces are highly imperfect – rich in dislocations and vacancies – the resultant properties are an enigmatic mixture of each materials’ properties and can be challenging to predict. Another example of special domain walls is the magnetic skyrmions. Skyrmions are 180° domain walls Jiang et al. (2017) either Néel type or Bloch type (shown in Figure 1.3(a) and (b)), continuously encircling a small domain, thus forming a closed loop. This looped structure cannot be continuously created or destroyed, thus defining a topological class for magnetic skyrmions. The topological protection gives magnetic skyrmions interesting and unique pseudo-particle properties. Overall, magnetic soft-hard bilayers and skyrmions are complex domain wall structures that are of significant interest due to their potential for use in advanced device technologies.

1.4 Magnetic Soft/Hard Bilayers

Heterostructures of coupled, magnetically hard/soft composites Kneller and Hawig (1991); Davies et al. (2005); Fullerton et al. (1998); Lewis et al. (2004); Jiang et al. (1999) have played



(a) 180° Bloch domain wall



(b) 180° Néel domain wall

Figure 1.2: two domain wall examples

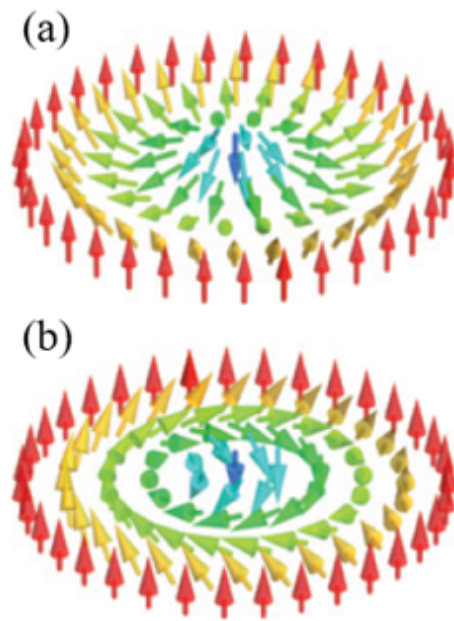


Figure 1.3: (a) Bloch type skyrmion; (b) Néel type skyrmion. The figure is adapted from [Fert et al. \(2017\)](#)

a critical role in permanent magnet [Yang et al. \(2015\)](#); [Roy and Kumar \(2009\)](#); [Quesada et al. \(2016\)](#); [Kneller and Hawig \(1991\)](#) technologies and modern data storage devices [Sonobe et al. \(2001a,b, 2002\)](#); [Victora and Shen \(2005\)](#); [Wang et al. \(2005\)](#); [Choe et al. \(2005\)](#); [Süss \(2006\)](#); [Shiroishi et al. \(2009\)](#); [Kirby et al. \(2010\)](#); [Suess et al. \(2015\)](#); [Weller et al. \(2016\)](#); [Zhu et al. \(2016\)](#). For example, high-anisotropy magnetic materials are coupled to high moment materials to develop a composite magnetic structure with a large maximum energy product for permanent magnet applications [Kneller and Hawig \(1991\)](#); [Zeng et al. \(2002\)](#). In magnetic composite recording media, the high-anisotropy (magnetically hard) region is frequently coupled to a magnetically soft region, which aids in the data-writing process [Litvinov et al. \(2001\)](#); [Suzuki et al. \(2005\)](#); [Liao et al. \(2010\)](#). As the anisotropy of the hard region has been increased to allow for smaller grains, and thus higher recording densities [Weller et al. \(2016\)](#); [Wu et al. \(2013\)](#); [Rottmayer et al. \(2006\)](#); [Gilbert et al. \(2013\)](#), the writability provided by the soft region has become indispensable [Goll and Macke \(2008\)](#); [Casoli et al. \(2008\)](#).

In published examples of magnetic soft-hard bilayers, soft Fe [Varvaro et al. \(2012\)](#) or Al-FePt [Gilbert et al. \(2016\)](#) are deposited onto hard $L1_0$ -FePt thin films. The $L1_0$ -FePt based magnetic recording media has large magnetic anisotropy [Gilbert et al. \(2016\)](#); [Liao et al. \(2013\)](#), which allows long term (10 year) stability at room temperature. However, the high anisotropy materials require a large magnetic field to reverse their orientation, e.g. the writing field, which can be as large as 3 T at room temperature [Suess et al. \(2005\)](#); [Victora and Shen \(2005\)](#). In Fe/ $L1_0$ -FePt and Al-FePt/ $L1_0$ -FePt bilayer thin films, as the thickness of soft layer increases, the field required to reverse the hard layer magnetization is significantly decreased. As a result, the writability is enhanced. Alternatively, the system can be designed with a gradient anisotropy [Dumas et al. \(2011\)](#) which significantly reduces the reversal field. Specifically, the switching field can be minimized by designing the anisotropy constant along the depth of thin films following a $\frac{1}{z^2}$ (z is soft layer thickness) law [Süss \(2006\)](#); [Suess et al. \(2009\)](#), while the hard region provides the thermal stability. This gradient anisotropy has been achieved by depositing thin films under different temperatures [Goll et al. \(2008\)](#) or thicknesses (for heterostructures). For example, Fe is firstly deposited onto FePt film between 200 °C and 400 °C and then the rest at 300 °C sequentially. Changing

the deposition temperature results in the change of the soft layer density and crystalline ordering, generating a gradient of anisotropy. The resulting gradient anisotropy has great potential for use in magnetic recording applications.

In summary, the magnetic bilayer structure can provide a balance between thermal stability and writability, which has great potential in a range of different technological applications and is an active area of research and development.

1.5 Magnetic Skyrmions

Traditionally, skyrmions can be found in some cubic B20 materials, which include MnSi, Cu₂OSeO₃ and FeGe. These skyrmions are formed due to the energy competition between the DM interaction, exchange interaction, Zeeman energy and thermal fluctuations. Skyrmions in B20 materials are stabilized in a small window of magnetic field and temperature parameter space. For example, the MnSi skyrmions [Mühlbauer et al. \(2009\)](#) are formed in a temperature range of 27 to 30 K within a field range of 0.12 to 0.23 T. Another example is the Cu₂OSeO₃ skyrmions [Seki et al. \(2012\)](#) which forms at temperature (56 to 58 K) with a magnetic field of (20 to 40 mT). Both MnSi and Cu₂OSeO₃ skyrmions are formed at low temperatures under an external field. Though the formation of FeGe skyrmions [Yu et al. \(2011\)](#) is near room temperature (276 to 280 K), the external field is still required (20 to 40 mT).

The key to form Skyrmions in B20 bulk materials is the DM interaction. In thin-film multilayers of 3d/5d elements, such as Ir/Fe or Co/Pt, the DM interaction can be generated at the interface due to the strong spin orbit coupling from the heavy metal. This induced interfacial DM interaction plays an important role to realize isolated skyrmions. The first investigation of these skyrmions are from Fe(monolayer)/Ir [Heinze et al. \(2011\)](#) and FePd(bilayer)/Ir [Romming et al. \(2013\)](#) thin film materials. These skyrmions can be stabilized in conditions which are extreme compared to the B20 materials ($T < 10$ K and $H > 1$ T). In later work, the substitution of Fe with Co in magnetic bilayer structures has

been shown to nucleate skyrmions at the interface of Ir/Co and Co/Pt multilayers [Moreau-Luchaire et al. \(2016\)](#). In these system, the skyrmions can be stabilized at room temperature, but the external field is still required.

Without DM itneractions, skyrmions can also be found in some magnetic multi-layer thin film materials (e.g. Fe/Gd [Montoya et al. \(2017a,b\)](#); [Desautels et al. \(2019\)](#)). These skyrmions are stabilized by diploar interactions at room temperature without a magnetic field. The magnetization of skyrmion core is determined by a field sequence. However, without a DM interaction, the winding direction of the skyrmion can be either clockwise or counterclockwise; in B20 materials, recall the DM interaction has a directionality to it, and so all the skyrmions have the same winding direction for a defined core direction. The vector defined by the winding direction and core orientation is called the chirality. The chirality in systems without a DM interaction are distributed with randomly and the total number of skyrmions with clockwise and counter-clockwise chirality will be be statistically equal. In recent works, the chirality of Fe/Gd skyrmions is controlled by inducing interfacial DM interaction, which is achieved by introducing $5d$ heavy metals such as Ir and Pt. The appropriate doping level of these elements balances the energy between the interfacial DM interaction, dipolar interactions as well as exchange interaction, which stabilizes skyrmions with controllable chirality at room temperature.

Most skyrmions are stabilized by DM interaction or dipolar interactions in materials with broken symmetry, which includes B20 materials. However, in some centrosymmetric materials, skyrmions can also be realized due to magnetic frustration effects. One example is Gd_2PdSi_3 [Kurumaji et al. \(2019\)](#): a triangular-lattice of Gd atoms sandwiching a nonmagnetic honeycomb-lattice layer composed of Pd and Si atoms. In a Gd layer, the hexagonal lattice can induce the magnetic frustration, which results in a variety of spin states in Gd layers. In addition, between the Gd layers, the RKKY interaction can introduce a helix ordering in the out-of-plane direction. The geometrically determined magnetic frustration competes with the RKKY interaction, stabilizing the skyrmions in a window of ($0.5 \text{ T} < H < 1.5 \text{ T}$, $T < 15 \text{ K}$).

In summary, skyrmions have been observed in B20 materials, mono-layer and multi-layer thin films as well as the centrosymmetric compound materials. For broken symmetry

materials, the B20 skyrmions can be stabilized by the DM interaction while the multilayer thin film skyrmions can be formed due to dipolar interactions or interfacial DM interactions. The DM interaction, as well as the interfacial DM interaction, are important to determine the skyrmion chirality. In centrosymmetric materials, the skyrmions can be realized by the balance between the geometry defined frustration and RKKY interactions. These skyrmion materials provide great insight into the study of fundamental physics. In addition, by tailoring the material properties of the systems in which skyrmions are formed, researchers can control the size and shape of skyrmions, which motivates skyrmions for future applications in data storage and logic device applications.

Chapter 2

Characterization Methods and Micromagnetic Simulation

In this chapter, I will introduce the characterization methods used in this dissertation. These methods include elastic neutron scattering, vibrating sample magnetometry and ferromagnetic resonance. In addition, micromagnetic simulations are also introduced. The micromagnetic simulations provide unique insights into the magnetic configuration and response by determining the magnetic energies, which often cannot be experimentally extracted.

2.1 Neutron Scattering

2.1.1 The Basis of Elemental Neutron Scattering

Neutron scattering is one of the most powerful tools for the characterization of magnetic microstructures. The neutron is a chargeless fermion with an intrinsic spin, allowing it to interact with nuclear structures and magnetism. Generally, neutrons can be produced by either spallation or fission. For spallation, high energy protons are accelerated by a linear accelerator and then collide with a heavy metal target (often mercury, tantalum or tungsten), which generates neutrons. In fission sources, a radioactive fuel is used which decays to produce neutrons, usually U-235. The core is designed such that the decay of one atom, and

associated neutron emission, triggers subsequent atoms to decay resulting in a steady state generation of neutrons. These generated neutrons usually have high energy, and cannot be directly used for scattering experiments; these 'hot' neutrons also have a small absorption cross section and so cannot be absorbed by other U-235 atoms to propagate the reaction. Therefore, a moderator (usually water, heavy water or graphite) is used to decelerate these neutrons. In both spallation and fission sources, the generated neutrons contain a range of wavelength; using time of flight, a velocity selector or a monochromator, we can select neutrons with a specific wavelength. Diffraction depends explicitly on the wavelength of the probe, in this case the neutron, and so it is important to know the wavelength to high precision.

Neutrons can be scattered by magnetic or non-magnetic atoms; the scattering intensity is directly related to the atomic nuclear (b_n) and magnetic (b_m) scattering lengths. For elastic scattering process, the incident neutron plane wave (wave vector $\mathbf{k}_0$) is scattered by a particle which results a spherical wave (wave vector $\mathbf{k}_1$), shown in Figure 2.1. Notably the elastic scattering condition is only satisfied when $|\mathbf{k}_1| = |\mathbf{k}_0|$. Normally, the observation of scattered neutron is far away from the measured particle, which means the sample diameter ($\mathbf{r}'$) is much smaller than measurement distance ($\mathbf{r}$). As a result, the sample can be considered as a "point"; the realistic scattering space vector $\mathbf{r} - \mathbf{r}'$ can be approximately equaled to $\mathbf{r}$. The scattered neutrons can then be collected by a neutron sensitive detector which has an angular cross section ($d\Omega$). This cross section provides information about the atom positions and magnetic properties.

One problem with the above model is that it uses the Born approximation to treat the incident wave; no distortion is considered for the incident wave before scattering ($\mathbf{k}_0$ is unchanged in different media). The Born approximation works well for most incident angles which makes the problem much easier. However, when the incident angle is small, the Born approximation is no longer satisfied. The incident wave becomes highly distorted due to strong interactions with the sample. Therefore, an improved approximation (e.g. distorted wave Born approximation) is needed, but this is beyond the scope of this dissertation.

The elastic scattering intensity equals to $\frac{1}{N} \sum_i^N f_i(q) \exp(iqr_i) \sum_j^N f_j(q) \exp(-iqr_j)$, where q is the reciprocal space vector, N is the total number of particles, $f_i(q)$ and $f_j(q)$ are

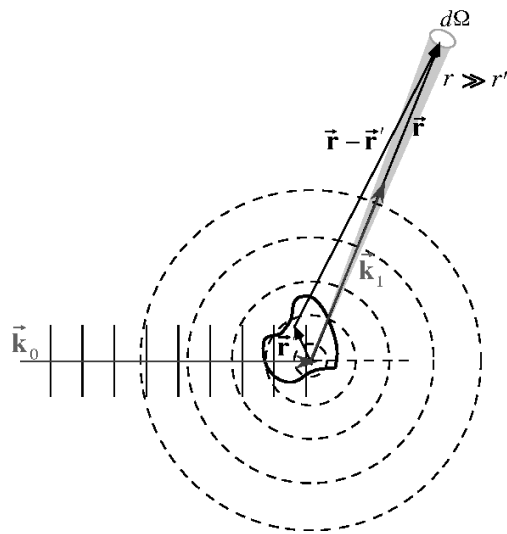


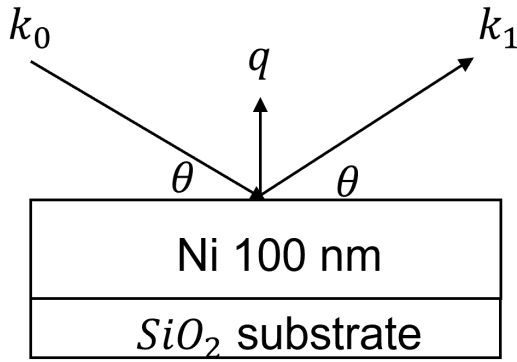
Figure 2.1: diagram of scattering. The figure is adapted from [Carpenter and Loong \(2015\)](#)

the form factors of particles i and j , respectively, and r_i and r_j are space position vectors of particles i and j , respectively. Assuming the particles are identical, then we can rewrite the scattering intensity as $f^2(q)S(q)$, where $S(q) = \frac{1}{N} \sum_i^N \sum_j^N \exp[iq(r_i - r_j)]$ is the scattering structure factor. The $f(q)$ is the Fourier transform of the scattering potential in real space. Assuming the potential is a homogeneous sphere with radius r , the corresponding form factor equals to $f(q) = \frac{3}{(qr)^3}[\sin(qr) - qr \cdot \cos(qr)]$. At small q , $\sin(qr) \approx qr$ and $qr \cdot \cos(qr) \approx 0$, therefore $f(q) = \frac{3}{(qr)^2}$. The $S(q)$ is the averaged configuration of the relative positions between particles of the entire sample. In a 1D periodic lattice with a lattice constant d , the scattering intensity can significantly increase when $q = 2\pi/d$. When the total number of particles is extremely large, the $S(q)$ becomes a delta function, which is an indication of classical scattering. With the form and structure factor, neutron scattering can reveal the shape and ordering of particles.

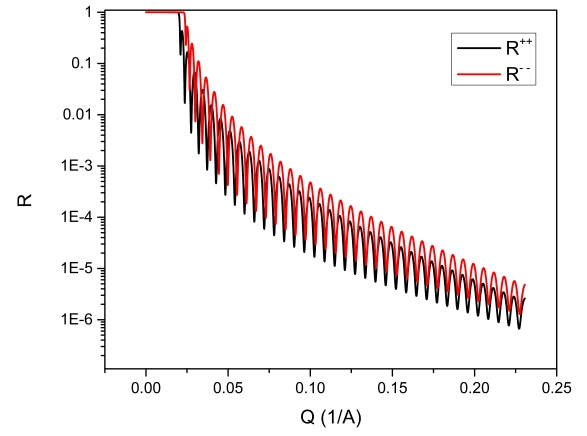
2.1.2 Polarized Neutron Reflectometry (PNR)

Polarized neutron reflectometry (PNR) is a neutron scattering technique which is most commonly used to study magnetic textures in thin films, providing direct insight to the nuclear and magnetic structure along the depth profile. Figure 2.2a shows the reflectometry set up for a Ni (100 nm)/SiO₂ (substrate) thin film. In this measurement, the incident and reflected angles are same, corresponding to the condition for specular reflection. The reciprocal space vector (q) is along the film normal direction (z) and equals to $k_1 - k_0$. During the measurements, the incident angle of the neutrons relative to the surface of the sample, θ , is changed and the reflection intensity is measured. This θ and the reciprocal space vector q are correlated through $q = \frac{4\pi}{\lambda} \sin(\theta)$, where λ is the wave length of the incident neutrons - also the reflected neutrons, since PNR is an elastic scattering technique. As θ changes, the q changes simultaneously, therefore we get the reflectivity (R) vs q data.

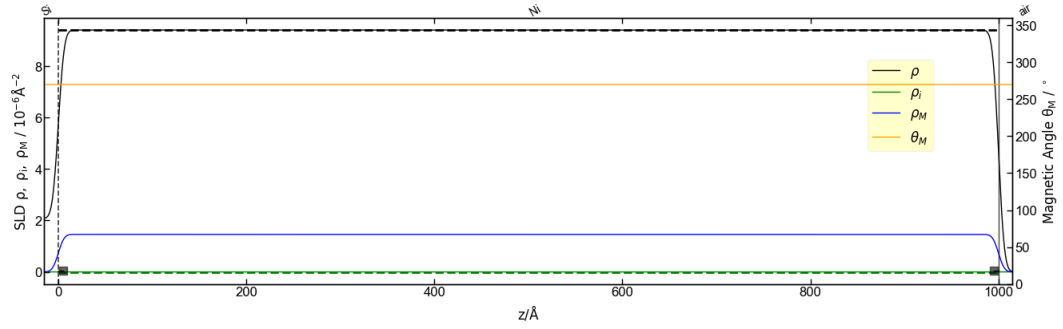
During the experiment the incident neutrons can be polarized either spin up or spin down with respect to a magnetic field at the sample using a supermirror, spin flipper and guide field. During the scattering process the neutron spin can flip if the magnetization of the sample is perpendicular to the incident spin of the neutron. After scattering, another spin flipper and supermirror allows the spin of the scattered neutrons to be probed. This



(a) Reflectivity diagram



(b) Reflectivity vs q



(c) The nuclear and magnetic depth profile

Figure 2.2: Neutron reflectivity of Ni (100 nm) thin film

allows us to measure four scattering "cross sections": R^{++} , R^{+-} , R^{-+} and R^{--} , where + and - indicate the spin of the neutrons; the two superscripts indicate the incident and scattered neutrons, respectively. The R^{++} and R^{--} signals indicate that the incident and scattered neutrons have the same spin and are called the non-spin flip channels. On the other hand, R^{+-} and R^{-+} indicate the spin has flipped its orientation during the scattering process and is thus called the spin flip channel. Notably, the orientation of the spin is set by the guide field and can be pointed any direction (in-plane or out-of-plane). The scattered neutrons will probe the magnetization parallel to the guide field in the non-spin flip channel and orthogonal to the guide field in the spin flip channel. There is one more scattering rule: the neutron is not sensitive to magnetization parallel to the reciprocal space vector ($\mathbf{q}$). This scattering rule, call the Halperin effect emphasizes that the magnetic scattering intensity is related to the Fourier transform of the magnetization in the real space, which is proportional to $\mathbf{q} \times \mathbf{M} \times \mathbf{q}$. When $\mathbf{M}$ is parallel to $\mathbf{q}$, the magnetic scattering intensity becomes eliminated. Simultaneously, all these four reflectivity data is sensitive to nuclear scattering from the sample.

In order to analyze the reflectometry data, a model of the nuclear and magnetic depth profile is fitted to the experimental data using the REFL1D software package. The profile for the Ni (100 nm)/SiO₂ (substrate) thin film is shown in Figure 2.2b. In the Ni (100 nm)/SiO₂ (substrate) thin film, when q is smaller than a critical value (q_c), the reflectivity equals to 1, which indicates the incident neutrons are all reflected. In a more precise way, the energy must be conserved at the interface for elastic scattering. Therefore, the total energy in the system can be written as $\frac{\hbar^2 k_0^2}{2m} = \frac{\hbar^2 k^2}{2m} + \frac{2\pi\hbar^2}{m}\rho$, where k_0 is incident wave vector, k is the wave vector inside the media, ρ is the scattering length which includes both the nuclear (ρ_N) and magnetic (ρ_M) components. For $\frac{\hbar^2 k_0^2}{2m} < \frac{2\pi\hbar^2}{m}\rho$, no wave is allowed in the media which results in total external reflection. The critical q value which corresponds $\frac{\hbar^2 k_0^2}{2m} = \frac{2\pi\hbar^2}{m}\rho$ is identified as q_c . Notably, the q_c of R^{++} is larger than R^{--} , which is due to the different magnetic potential (R^{++} : $\rho = \rho_N + \rho_M$; R^{--} : $\rho = \rho_N - \rho_M$). For $q > q_c$, the reflected intensity oscillates (Kiessig fringes) due to the superposition of the reflected wave from the surface and the interface. At specific angles the pathlength difference between these waves result in their phase being the same, and the waves add constructively, resulting in a maximum in the

reflected intensity. Conversely, if the waves from the surface and buried interface are shifted by half a wavelength, the waves add destructively, resulting in cancellation of the intensity. For a thin film with a single layer, the thickness can be calculated by $d = 2\pi/\Delta q$, where Δq is the distances between two nearest peaks in reciprocal space. In the provided example of a Ni thin film the specular reflection is generated by reflections from the surface of the Ni and the buried Ni/SiO₂. The specular reflection originates from the change of the nuclear and magnetic potential along the film normal direction, e.g. the interfaces, and no scattering occurs from within the layers. At the interface the variation of the potential generates a force, which changes the wave vector direction and causes the reflection. Moreover, when the roughness is large at the surface or interface, the Kiessig fringes become less evident due to diffuse scattering. As q increases, the reflectivity generally decreases as a factor of q^{-4} . This decay law is due to the scattering intensity which is proportional to the $f^2(q)$ as mentioned above. The R vs q data of Ni (100 nm)/SiO₂ (substrate) is a simple example to help understand PNR measurements.

The REFL1D software package uses the simulated structure and calculates the reflectivity. Through an iterative process the model is changed in such a way that the calculated reflectometry matches the experimental data. The simulated profile includes both the nuclear and magnetic scattering length density (SLD) along the film normal direction, shown in Figure 2.2c. The SLD is defined as the sum of the scattering length in a unit molecular volume along the depth of the sample. With SLD along thin film depth profile, the PNR can provide important information about the magnetic structure and composition of the sample.

2.1.3 Small Angle Neutron Scattering (SANS)

Small angle neutron scattering (SANS) can probe material structure on the nanometer scale; structures on this length scale are critical to the performance of advanced engineering materials. For SANS measurements, the sample is typically set up in a transmission geometry, shown in Figure 2.3. The scattered angle θ is linearly correlated to q with a similar relationship as PNR: $q = \frac{4\pi}{\lambda} \sin(\frac{\theta}{2})$, where λ is the wavelength of incident neutrons. According to $d = 2\pi/q$, the q value is proportional to the reciprocal of the real-space scale

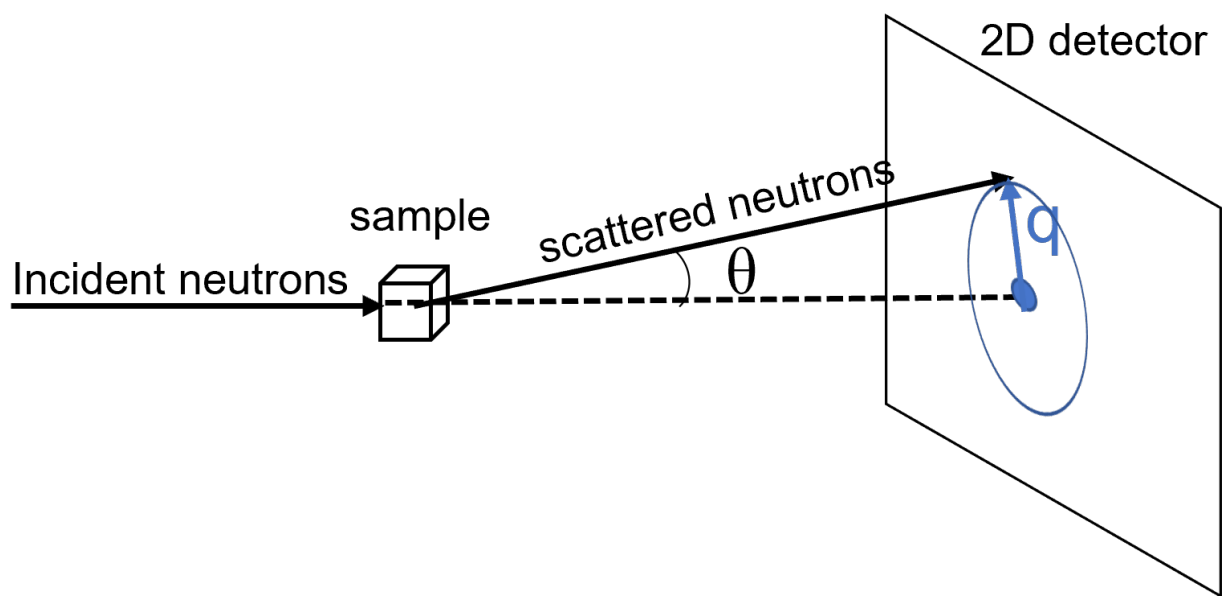


Figure 2.3: diagram of SANS

of the structure being probed, therefore a smaller q value corresponds to a larger real-space scale. This means that in order to probe large-scale domains (such as those larger than 100 nm), it is necessary to measure the scattered neutrons at small values of q , corresponding to a small scattering angle (θ). In SANS measurements, low q can be measured using a long neutron wavelength (λ) and long detector distances (to capture small angle scattering away from the direct beam). A two-dimensional (2D) detector consisting of many pixel-like neutron detectors is then used to count the scattered neutrons, which can provide q vectors with q_x and q_y components. The scattering intensity at these q vectors provides information about the magnetic structure of the sample. Next, we will discuss the scattering pattern of two specific magnetic structures.

Stripe domains Magnetic stripe domains are a domain configuration which commonly forms in magnetic thin films with a perpendicular anisotropy, making the out-of-plane direction the magnetic easy axis. This configuration has a high dipolar energy, and so the film breaks into stripes, oriented in the out-of-plane direction, alternating in the positive and negative out-of-plane directions. Figure 2.4a shows a diagram of a thin film with stripe domain structure along z (film normal) direction. The dimension of domains along the x direction are d_1 and d_2 for magnetization along the positive and negative z direction, respectively. SANS can be used to measure this structure with the direct beam parallel to the z direction. Since we treat the neutron as plane wave, the SANS diffraction pattern is just the Fourier transform of real space structure. For stripe domains, the Fourier transform along the x direction is $F_x = \sum^n f_{up} \exp[inq_x(d_1 + d_2)] + \sum^n f_{down} \exp[i(n + \frac{1}{2})q_x(d_1 + d_2)]$, where f_{up} and f_{down} are form factor of magnetization parallel and anti-parallel to z , respectively, and n is the total number of identical domains. The corresponding scattering intensity equals to $|F_x|^2$, which can be maximized when $q_x(d_1 + d_2) = \pm 2\pi$. As a result, the diffraction pattern of the stripe domains are two spots at $q_x = \pm 2\pi/(d_1 + d_2)$, shown in Figure 2.4b.

Skyrmion lattice The magnetic skyrmion, as discussed above, can emerge as a consequence of competition between the exchange interaction and the DM interaction, dipolar fields, or random anisotropies. The magnetic skyrmion configuration can be described

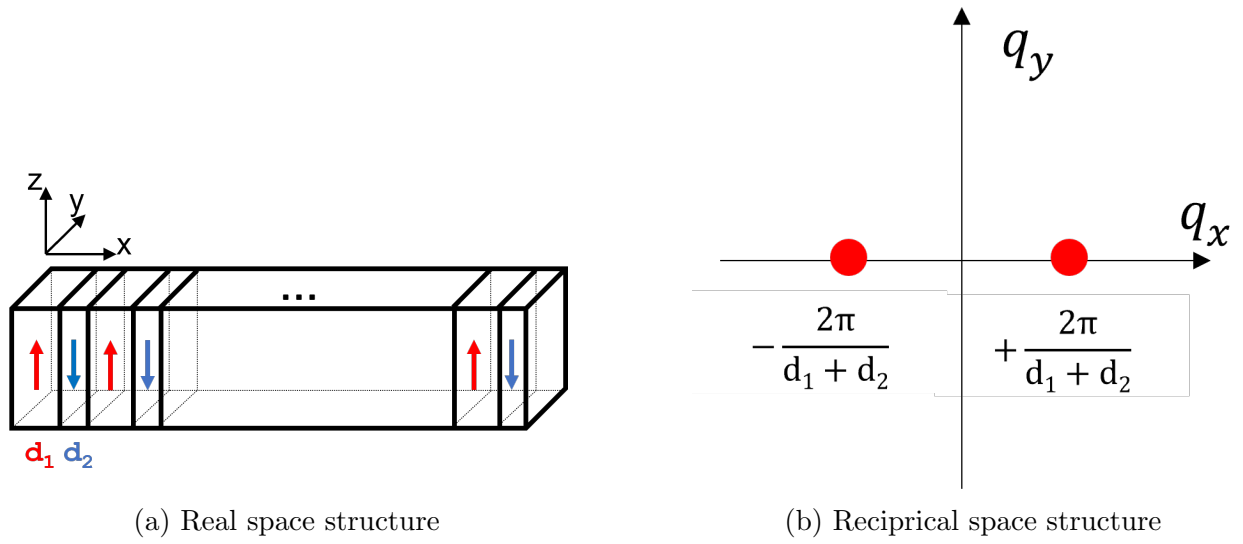


Figure 2.4: diagram of stripe domain

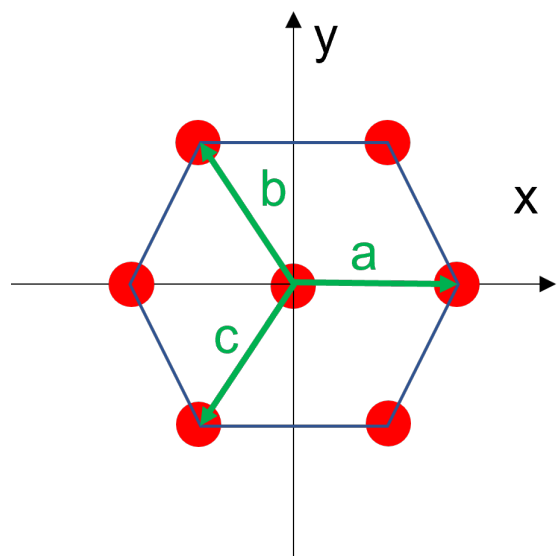
as a coplanar loop of magnetic moments, with a core and perimeter oriented in the out-of-plane direction. This structure has a scattering form factor on its own, however magnetic skyrmions frequently form a hexagonal lattice (a, b and c), shown in Figure 2.5a. The Fourier transform of a is $a^* = \frac{b \times c}{a \cdot (b \times c)}$, which corresponds to a counterclockwise rotation of 30° of a . Similarly, b^* and c^* can be achieved by 30° anti-clockwise rotation of b and c , respectively. Therefore, the diffraction pattern of skyrmion lattice is still a hexagon which is rotated by 30° relative to real space lattice, shown in Figure 2.5b.

2.2 Vibration Sample Magnetometry (VSM)

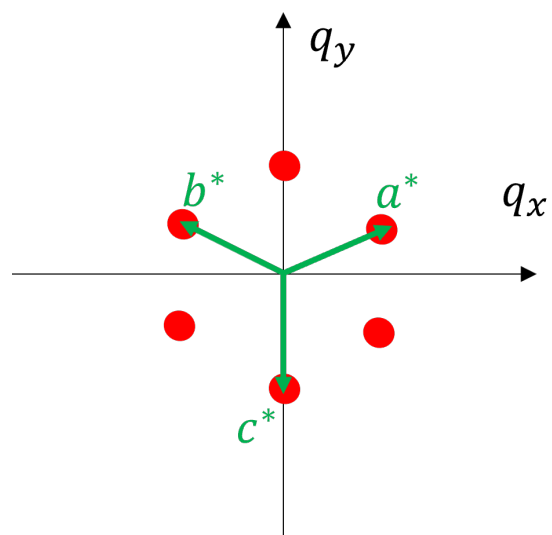
Magnetic measurements were performed using a vibrating sample magnetometer (VSM), set up in a physical properties measurement system (PPMS). In the PPMS system, the magnetic field is generated by a superconductor solenoid located around the sample position. In our lab, this field can range from -9 T to $+9$ T with a maximum changing rate of 22 mT/s. The temperature of the chamber can range from 1.7 K to 400 K with a maximum cooling rate of 12 K/s.

After loading sample, the chamber is evacuated then purged with Helium gas for 3 cycles. This process replaces the air in the chamber with He and reduces the possibility of the contamination from the air as its components can freeze to the sample resulting in erroneous temperature-dependent magnetization. Notably, the measurement pressure of the chamber is around 35 Torr (of helium), which acts as an exchange gas, transporting heat from the sample to the chamber walls.

For VSM, the sample is oscillated at approximately 40 Hz using a linear motor. The sample is centered within a coil of copper wire, moving in the plane perpendicular to the coil. The vibration of the sample constitutes a changing magnetic flux through the coil which results in an induced current. This current is proportional to the total magnetization of the sample, the oscillation amplitude and frequency. The system measures the induced current and calculates the magnetization of the sample. By scanning temperature or magnetic fields, the VSM can generate M vs T or M vs H curves. These curves reveal the details of the magnetic phase transformation (T_C or T_N) as well as the magnetic hysteresis loop (H_c and



(a) Real space structure



(b) Reciprocal space structure

Figure 2.5: diagram of skyrmion lattice

M_s). With this information, the VSM can provide insight into the magnetic behavior of the sample.

2.3 Ferromagnetic Resonance (FMR)

Ferromagnetic resonance (FMR) is a spectroscopy technique which is used to evaluate the magnetic dynamics. It is known that, for a magnetic moment pointed along any particular direction, if a magnetic field is applied anywhere except parallel to the magnetization, the moment will precess around the magnetic field, with a damping term which causes the path to follow a spiral. At the termination of the spiral the moment aligns with the magnetic field. As shown in Figure 2.6, magnetic dynamics are governed by the Landau–Lifshitz–Gilbert equation $\frac{d\mathbf{M}}{dt} = -\gamma\mathbf{M} \times \mathbf{H}_{\text{eff}} - \eta\mathbf{M} \times \frac{d\mathbf{M}}{dt}$, where $\mathbf{M}$ is the magnetization, t is time, $\mathbf{H}_{\text{eff}}$ is the total effective magnetic field, γ is gyromagnetic ratio and η is the damping constant. The first term corresponds to a torque orthogonal to $\mathbf{H}_{\text{eff}}$ and $\mathbf{M}$, which results in the circular precession of the magnetization. The second term corresponds to a damping torque orthogonal to $\mathbf{M}$ and $\frac{d\mathbf{M}}{dt}$, which moves the magnetization towards $\mathbf{H}_{\text{eff}}$. Therefore, the magnetic dynamic includes precession and damping simultaneously. After many precessions, the magnetization becomes static when $\mathbf{M}$ is parallel to $\mathbf{H}_{\text{eff}}$. Notably, $\mathbf{H}_{\text{eff}}$ here is not only the external magnetic field, but also includes the effective anisotropy field, dipolar fields and exchange field. The $\mathbf{H}_{\text{eff}}$ is the sum of these vector fields.

One of the most widely used FMR instruments is based on a vector network analyzers (VNA), shown in Figure 2.7. The VNA provides a spectrum of alternate current (AC) currents between two ports (port 1 and port 2). As the AC currents is transmitted through a wire, a corresponding AC magnetic field is generated circulating around the wire, following from Ampere’s law. The FMR setup uses a special, flat wire patterned on top of a high frequency fiberglass, called a grounded coplanar waveguide. The sample is placed on top of the waveguide and the small AC magnetic field can interact with the magnetic sample. When the frequency of an AC magnetic field matches precession frequency of the magnetic moments (under a specific $\mathbf{H}_{\text{eff}}$), the sample can absorb the microwave energy. The absorbed energy, which corresponds to the magnetic response of the sample, is identified by a reduction

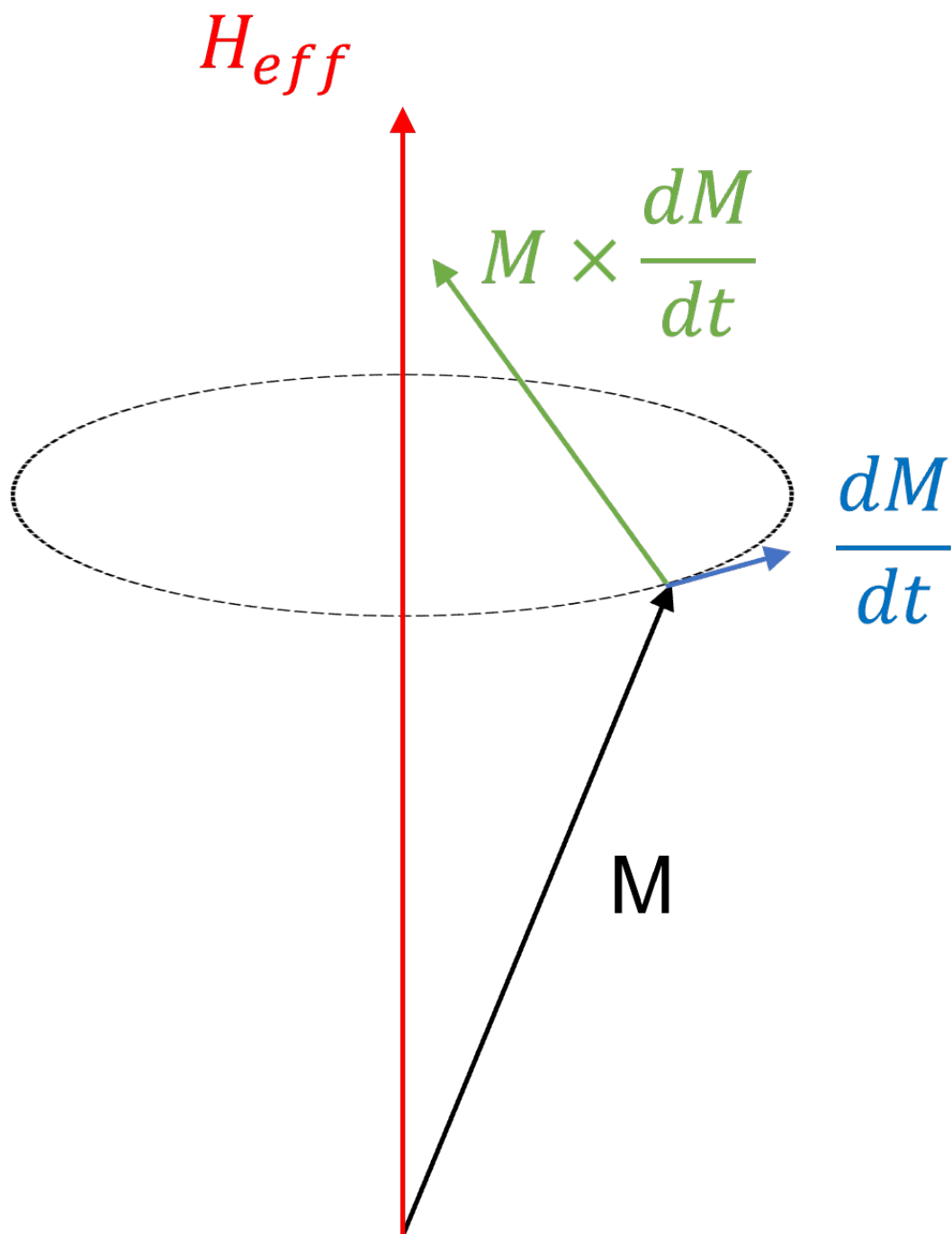


Figure 2.6: Magnetic precession diagram

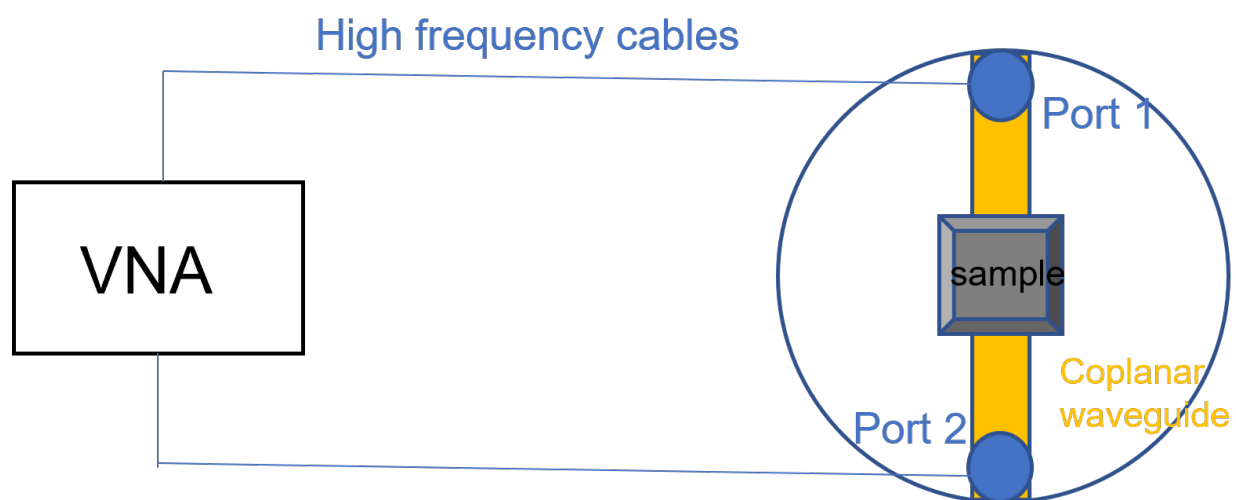


Figure 2.7: FMR diagram

in the power transmission through the waveguide, termed S_{12} (S_{21}), e.g. the transmission signal from port 1(2) to port 2(1). Besides the sample absorption, the microwaves can also be reflected, measured by S_{11} or S_{22} (reflection of port 1 or 2). These reflection data can be used to study the magnetic dynamics of the sample. With these measurement data, the FMR technique is a powerful and versatile tool for studying the magnetic dynamic behavior of materials and understanding the underlying physics of magnetic phenomena.

2.4 Micromagnetic Simulation

Micromagnetic simulations are performed using the object oriented micromagnetic framework (OOMMF) [Donahue \(1999\)](#) simulation platform. This program allows us to define the geometry (Atlases) of the sample which is constructed of nanoscale unit cells (meshes). The mesh is chosen to be small such that the exchange interaction is expected to dominate the orientation of the moments within the cell, called the macrospin approximation. Next, the material properties are input for each of the unit cells, which includes the exchange constant, the magnetocrystalline anisotropy and saturation magnetization, etc. Using these parameters, the magnetic energy terms can be calculated. The sum of these magnetic energies correspond to an effective field. Finally, a time evolver applies the LLG equation to each unit cell using this total effective field. After many circles, the damping term causes the magnetization of each cell to stabilize along a particular direction, which ends the simulation. The field can be changed and the process is repeated. Throughout the simulation the vector magnetization and energy terms are all recorded. The OOMMF simulations can provide insight to the magnetic structures dependent on material properties, which helps understand the experimental results.

Chapter 3

Magnetic Soft-hard Bilayers

This chapter is quoted from the paper "Controlling magnetic configuration in soft-hard bilayers probed by polarized neutron reflectometry" (APL Materials 10, 011107 (2022)), contributed by Nan Tang, Jung-Wei Liao, Siu-Tat Chui, Timothy Ziman, Alexander J Grutter, Kai Liu, Chih-Huang Lai, Brian J Kirby and Dustin A Gilbert. The author of this dissertation was the lead writer of this paper, performing part of data analysis, simulation, interpretation, writing, and figure generation under Dustin A Gilbert's guidance. The thin films sample are fabricated by collaborators Jung-Wei Liao, Kai Liu and Chih-Huang Lai. Brian J Kirby and Alexander J Grutter provided the polarized neutron measurements and helped with the data analysis. Siu-Tat Chui and Timothy Ziman helps for some discussion of the results.

3.1 Motivation

The magnetic interactions play an important role to determine magnetic configurations in hard/soft bilayers with perpendicularly magnetized hard layer. The magnetic configuration can be influenced by sample geometry, material properties and the ambient conditions. For sample geometry, the most important parameter is the soft layer thickness, t_{soft} [Gilbert et al. \(2016\)](#); [Zambano et al. \(2007\)](#). For the case of a thin soft layer with a thickness less than the classical exchange length (l_{ex}), it is typically accepted that the magnetic moments are rigidly coupled to the hard layer by the exchange interaction. Furthermore,

as t_{soft} increases, it is expected that the magnetitic moment relaxes in-plane, especially away from the hard/soft interface, resulting in exchange spring-like behavior [Lavorato et al. \(2016\)](#). In addition to t_{soft} , the magnetic configuration is also influenced by the material properties including the saturation magnetization (M_s^{hard} and M_s^{soft}), exchange stiffness (A_{hard} and A_{soft}), the hard layer magnetocrystalline anisotropy (K_u) and the hard/soft interfacial coupling stiffness (A_{int}) [Fullerton et al. \(1999\)](#); [Asti et al. \(2006\)](#); [Suess et al. \(2005\)](#). While some of these magnetic material parameters have predictable relationships with the magnetic configuration, e.g. K_u and A_{hard} can be used to determine an exchange length ($\approx \pi\sqrt{(A_{hard}/K_u)}$) in hard magnets [Abo et al. \(2013\)](#), the dependence on M_s^{hard} and M_s^{soft} is not as straightforward. For modern applications in heat assisted magnetic recording (HAMR) technologies, understanding the temperature dependence of these properties and its effect on the magnetic configuration is critical to device functionality [Pan and Bogy \(2009\)](#); [Kryder et al. \(2008\)](#); [Victora and Shen \(2005\)](#); [Varaprasad et al. \(2013\)](#); [Dobin and Richter \(2006\)](#). This quality is manifested in HAMR devices where high temperatures are used to reduce K_u , but simultaneously also reduce M_s^{hard} and M_s^{soft} , establishing an optimization problem to achieve the ideal writability. Finally, the applied magnetic field also strongly influences the magnetic configuration; while most of the magnetic energy terms are uniaxial, the field dependent Zeeman energy is unidirectional and establishes a new balance between the various energy terms. Therefore, to better design and control hard/soft magnetic heterostructures, it is important to understand the roles of sample geometry (t_{soft}), materials properties (M_s^{hard} , M_s^{soft}) and external influences (applied in-plane magnetic field, H_{ip} , and temperature, T) in determining the magnetic configuration.

Here, we use polarized neutron reflectometry (PNR) to investigate the magnetic configuration in hard/soft bilayer thin films. Comparing with previous magnetometry-based studies [Fullerton et al. \(1998, 1999\)](#), the PNR reveals the magnetic depth profile within the film, which is determined by the magnetic interactions. A series of bilayer samples with a hard magnetic underlayer (having perpendicular magnetic anisotropy) and a soft top layer is investigated, including $L1_0$ -FePt (hard)/Al-FePt (soft), [Co/Pd] (hard)/CoPd (soft), [Co/Pt] (hard)/FeNi (soft) and $L1_0$ -FePt (hard)/Fe (soft). By designing M_s^{hard} , M_s^{soft} and t_{soft} , the magnetic interactions, and the resultant magnetic configuration, are

systematically varied. Micromagnetic simulations are performed to support the experimental results and reflect similar magnetic behavior. The simulations also reveal the influence of M_s^{hard} and hard layer thickness (t_{hard}) in determining the magnetic configuration. In addition to the intrinsic properties and sample structure, the influence of temperature (T) and magnetic field (H) on the magnetic configuration are also studied by PNR. Polarized neutron reflectometry measurements, together with micromagnetic simulations, provide a comprehensive understanding of the influence of sample structure and external control variables on the magnetic configuration of hard/soft bilayer composite magnets. These results provide insight into the underlying physics correlating bilayer film structure with magnetic configuration.

3.2 Methods

3.2.1 Sample Preparation

Four series of samples covering a range of M_s^{soft} , M_s^{hard} and t_{soft} are synthesized by magnetron sputtering and the details can be found elsewhere [Gilbert et al. \(2016\)](#); [Liao et al. \(2013\)](#); [Greene et al. \(2014\)](#); [Davies et al. \(2004\)](#): 1). The Si/SiO₂ (200 nm)/L1₀-FePt (4 nm)/A1-FePt (t_{soft})/Ti (4 nm) samples were prepared in a vacuum system with a base pressure of 8×10^{-8} Torr. The oriented L1₀-FePt was prepared by rapid thermal annealing, as described previously [Gilbert et al. \(2013, 2016, 2014\)](#). Without breaking vacuum, the A1-FePt soft layer (2 nm or 5 nm) was deposited on L1₀-FePt by co-sputtering of elemental targets, followed by a Ti capping layer. 2). The preparation process of Si/SiO₂ (200 nm)/L1₀-FePt (4 nm)/Fe (9 nm)/Pt (2 nm) is similar to the Si/SiO₂ (200 nm)/L1₀-FePt (4 nm)/A1-FePt (t_{soft})/Ti (4 nm) samples, but sputtering 9 nm Fe and 2 nm Pt as the soft and capping layers, respectively. 3). For Si/SiO₂/[Co (0.4 nm)/Pt (0.7 nm)]₄/FeNi (t_{soft})/Pd(2 nm) samples, 4 repeats of Co (0.4 nm)/Pt (0.7 nm) multilayer stack were first sputtered on Si/SiO₂ substrate. Four different thicknesses of soft Fe_{0.2}Ni_{0.8} (0, 1, 3, 7 nm) layers are then deposited, followed by a 2 nm Pd capping layer. 4). The Si/Pd (20 nm)/[Co (0.3 nm)/Pd (0.9 nm)]₂₅/CoPd (t_{soft})/Pd (5 nm) samples were magnetron

sputtered in an ultrahigh vacuum chamber with a base pressure of 9×10^{-9} Torr. Magnetically hard [Co (0.3 nm)/Pd (0.9 nm)]₂₅ multilayers were sputtered onto Si substrates, with a 20 nm Pd buffer layer. Different thicknesses of t_{soft} (2, 5, 9 nm) were then deposited, followed by a 5 nm Pd capping layer. For all the samples, the underlayer films ($L1_0$ -FePt, Co/Pt and Co/Pd multilayers) have strong perpendicular magnetic anisotropy (PMA).

3.2.2 Experimental Details

Polarized neutron reflectometry was performed at the NIST Center for Neutron Research on both the PBR and MAGIK instruments, which uses 4.75 Å and 5.00 Å wavelength neutrons, respectively. The neutrons were polarized by a 3H spin filter with the magnetic moments parallel or anti-parallel to a guide field direction. For these measurements, this guide field was applied either parallel or vertical to the film normal. For the $L1_0$ -FePt/ $A1$ -FePt samples to study the influence of t_{soft} on magnetic configuration, the film was rotated into a grazing incidence with the neutron beam. The films were first saturated with a magnetic field (700 mT) parallel to the film normal direction and then the field was reduced to 50 mT. Then, this 50 mT field was rotated with a small angle away from the film normal as guide field. Using this configuration, the non-spin-flip PNR cross-sections (R^{++} and R^{--} , where R is the reflection intensity, with the superscript indicating the spin configuration of the incident and reflected neutrons, + (−) indicting the spin-up (spin-down) geometries) provide sensitivity to the nuclear composition, while the spin-flip channels (R^{+-} , R^{-+}) provide sensitivity to any in-plane net magnetization. For all other PNR measurements, the samples were saturated in the out-of-plane direction, then returned to remanence and measured in a small in-plane guide field (1 mT). In this configuration, R^{++} and R^{--} , provide sensitivity to both the nuclear depth profile, and the depth profile of the net in-plane magnetization component parallel to the applied field. Spin-flip scattering for this configuration is not considered here. All the experimental data are then model fitting by REFL1D software Kirby et al. (2012) .

Micromagnetic Simulation Three dimensional micromagnetic simulation is performed on the platform of object oriented micromagnetic framework (OOMMF) Donahue (1999) by using blocks $200 \text{ nm} \times 1 \text{ nm} \times (t_{hard} + t_{soft})$ with a mesh of $0.5 \text{ nm} \times 0.5 \text{ nm} \times 0.2 \text{ nm}$. The

exchange stiffness for hard magnet (A_h), hard/soft interface (A_{int}), and soft magnetic (A_s) are $A_h = A_s = A_{int} = 1.15 \times 10^{-11}$ J/m. The uniaxial anisotropy for hard magnet (K_u) is chosen to be 1×10^6 J/m³ while the anisotropy for soft magnet (K_s) is 0.

3.3 Experimental Results

3.3.1 Effects of Soft Layer Thickness

With the above experimental preparations, the first set of measurements were performed to study the influence of t_{soft} on the magnetic configuration. Using an out-of-plane guide field, reflectometry data for the $L1_0$ -FePt (4 nm)/A1-FePt (t_{soft}) bilayer films with $t_{soft} = 2$ nm and 5 nm are shown in Figure 3.1a and 3.1b, respectively. Data on the same sample but taken at a different time were shown in Gilbert et al. (2016) and reflects a similar profile; differences in the capping layer structure are captured in this work, likely due to the higher q -range measured. For the $t_{soft} = 2$ nm film, the experimental non-spin-flip R^{++} and R^{--} are essentially identical, and the spin-flip R^{+-} and R^{-+} are virtually zero. This indicates that there is little net in-plane magnetization anywhere in the film. In comparison, for the $t_{soft} = 5$ nm film, the R^{++} and R^{--} are still overlapping, but R^{+-} and R^{-+} are now nonzero, indicating the presence of appreciable in-plane magnetization. These interpretations are confirmed by model fitting (solid lines in Figure 3.1a and 3.1b) that yields the scattering length density (SLD) Zhu (2005) depth profiles for the two films, as shown in Figure 3.1c and 3.1d. The profiles have a nuclear component (NSLD) indicative of the nuclear composition and a magnetic component (MSLD) proportional to the in-plane magnetization. Notably, the MSLD is approximately zero for $t_{soft} = 2$ nm and nonzero for $t_{soft} = 5$ nm. These results confirm that t_{soft} can influence the magnetic configuration in the soft layer. Interestingly, this influence extends all the way to the interface; while both the hard and soft layers in the 2 nm sample are oriented out-of-plane, the soft layer in the 5 nm sample is almost entirely in-plane.

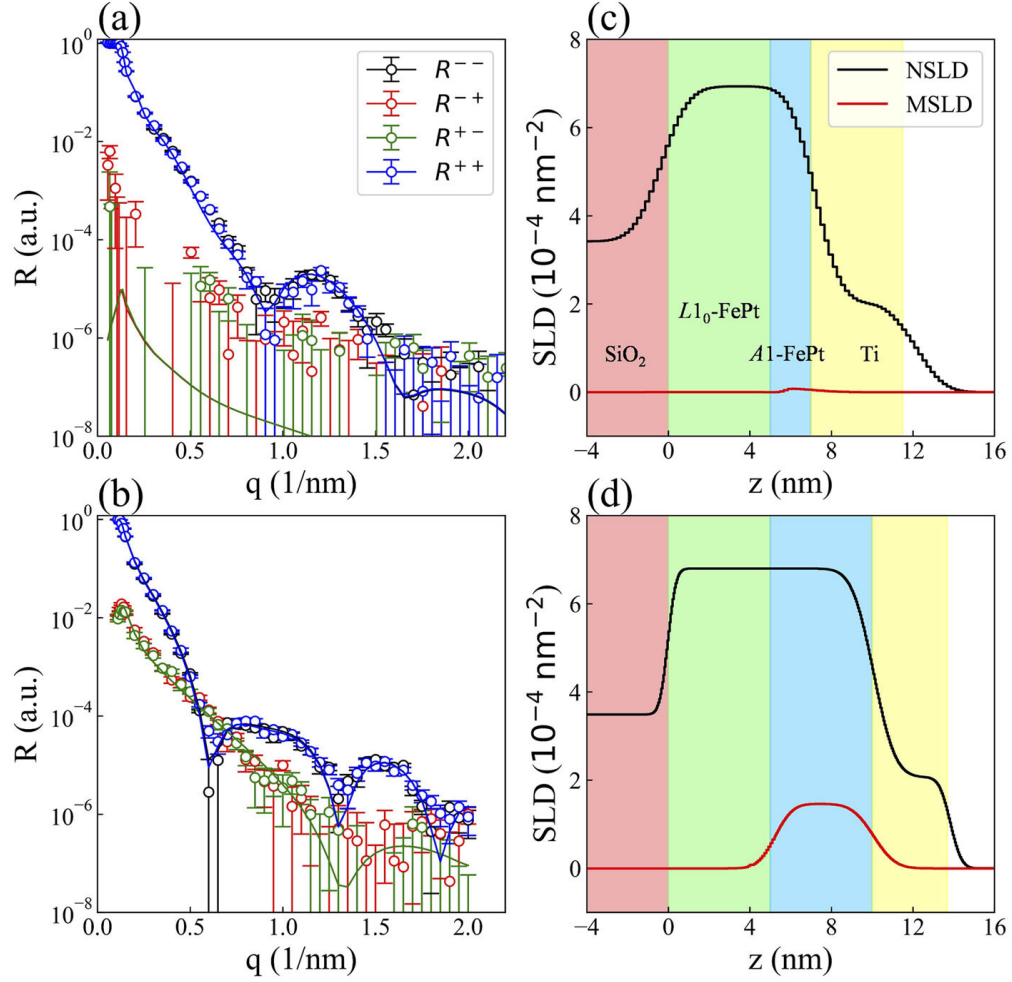


Figure 3.1: Model-fitted PNR data of $L1_0\text{-FePt}$ (4 nm)/ $Al\text{-FePt}$ for (a) $t_{soft} = 2$ nm and (b) $t_{soft} = 5$ nm and the associated scattering length density profiles for (c) $t_{soft} = 2$ nm and (d) $t_{soft} = 5$ nm. The red, green, blue, and yellow regions correspond to the depth profile of SiO_2 , $L1_0\text{-FePt}$, $Al\text{-FePt}$, and capping Ti layers, respectively.

3.3.2 Effects of Saturation Magnetization

Besides t_{soft} , M_s^{hard} and M_s^{soft} are expected to influence the magnetic configurations in these bilayer films. The PNR measurements are also performed on a series of samples with $M_s^{hard} \ll M_s^{soft}$, $M_s^{hard} < M_s^{soft}$ and $M_s^{hard} > M_s^{soft}$, including $L1_0$ -FePt(4 nm)/Fe(9 nm), [Co (0.3 nm)/Pd (0.9 nm)]₂₅/CoPd (9 nm), and [Co (0.4 nm)/Pt (0.7 nm)]₄/FeNi (7 nm), respectively. For the case of $M_s^{hard} \ll M_s^{soft}$ shown in Figure 3.2a, the experimental R^{++} and R^{--} curves are significantly separated, which indicates a net in-plane magnetization. The corresponding magnetic depth profiles, in Figure 3.2b, show a large MSLD in the soft layer (Fe region). Using the MSLD for bulk iron ($4.85 \times 10^{-4} \text{ nm}^{-2}$) Fitzsimmons and Majkrzak (2005), the soft Fe layer is confirmed to be almost entirely in-plane throughout its thickness. For $M_s^{hard} < M_s^{soft}$ and $M_s^{hard} > M_s^{soft}$, the experimental R^{++} and R^{--} curves are strongly overlapping, as shown in Figures 3.2c and 3.2e, respectively. The corresponding magnetic depth profiles show almost no MSLD in Figures 3.2d and 3.2f, respectively. These results suggest that in both the [Co (0.3 nm)/Pd (0.9 nm)]₂₅/CoPd (9 nm) and [Co (0.4 nm)/Pt (0.7 nm)]₄/FeNi (7 nm) films, even at these relatively larger thicknesses, the soft layer magnetization remains mostly in the out-of-plane direction. The PNR measurements are also performed on another five [Co (0.3 nm)/Pd (0.9 nm)]₂₅/CoPd and [Co (0.4 nm)/Pt (0.7 nm)]₄/FeNi samples with thinner soft layer thickness, shown in Appendix A.1 and A.2, respectively. These films also show very little in-plane magnetization.

3.3.3 Effect of In-plane Field and Temperature

The results above implicate a complex competition within these systems to define the soft-layer orientation, which depends on both M_s^{soft} and t_{soft} . Besides these intrinsic material parameters, the interfacial magnetic configuration is also dependent on the external variables (H_{ip} and T). Six PNR measurements were performed on the $L1_0$ -FePt (4 nm)/ $A1$ -FePt (2 nm) bilayer sample with $\mu_0 H_{ip}$ from 100 mT to 700 mT at $T = 295 \text{ K}$ and $T = 500 \text{ K}$, shown in Figure 3.3a and 3.3b, respectively. For each in-plane magnetic field, the corresponding R^{++} and R^{--} curves are separated, which indicates a net in-plane magnetization. This result

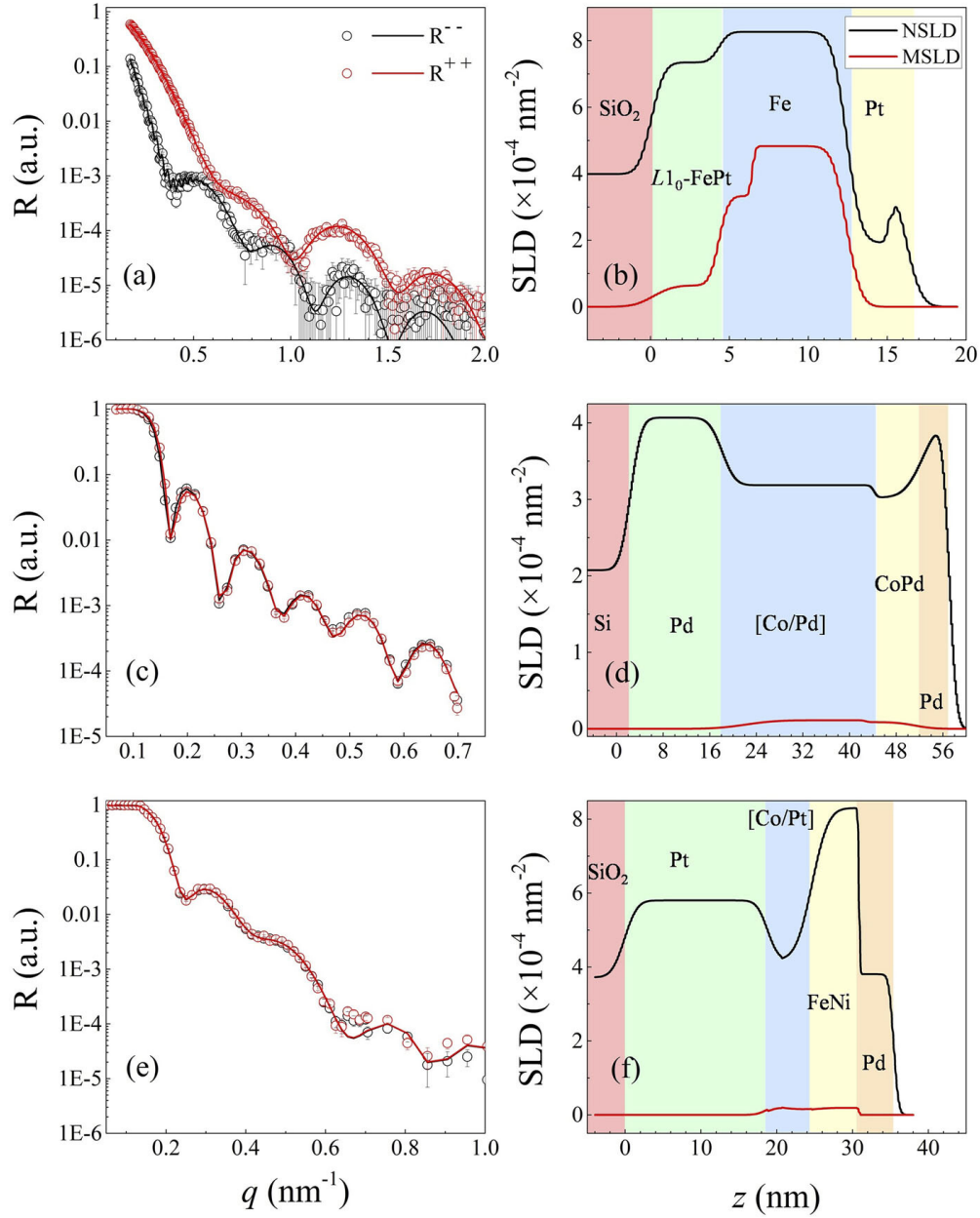


Figure 3.2: $L1_0$ -FePt/Fe (a) fitted R^{++} and R^{--} and (b) the corresponding nuclear (black) and magnetic (red) SLD profiles, where the pink, green, blue, and yellow regions are the SiO_2 , $L1_0$ -FePt, Fe, and Pt capping layers, respectively. [Co/Pd]-multilayers/CoPd (c) fitted R^{++} and R^{--} and (d) nuclear (black) and magnetic (red) SLD profiles, where the pink, green, blue, yellow, and orange regions correspond to the Si, Pd seed layer, [Co/Pd] multilayers, CoPd soft layer, and Pd capping layer, respectively. [Co/Pt]-multilayers/FeNi (e) fitted R^{++} and R^{--} and corresponding nuclear (black) and magnetic (red) SLD profiles with SiO_2 (pink), Pt buffer layer (green), [Co/Pt] multilayers (blue), FeNi (yellow), and Pd capping layer (orange).

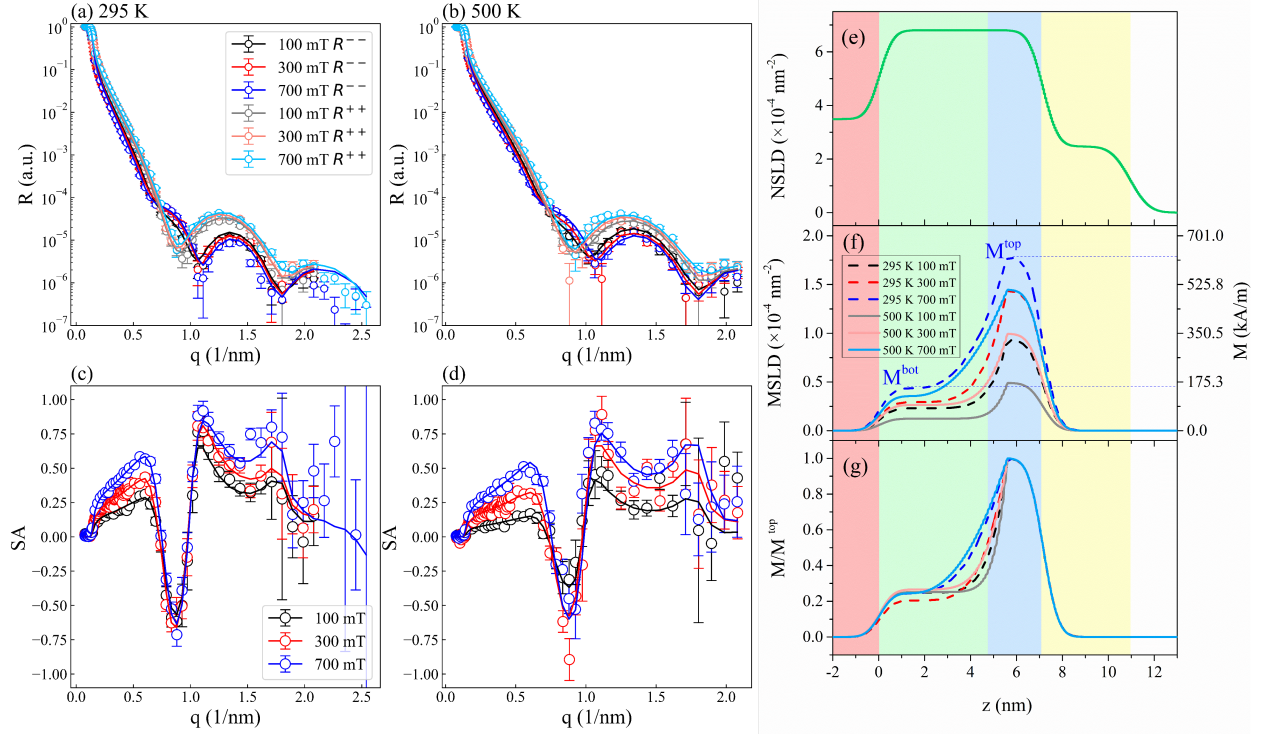


Figure 3.3: Polarized neutron reflectivity for $L1_0$ -FePt (4 nm)/A1-FePt (2 nm) with in-plane magnetic fields of 100 mT, 300 mT, and 700 mT at (a) 295 K and (b) 500 K. The spin asymmetry for each field at (c) 295 K and (d) 500 K. The fitted (e) nuclear and (f) magnetic depth profiles and (g) the normalized magnetic depth profile. M^{top} corresponds to the maximal in-plane magnetization of the soft layer, and M^{bot} is the magnetization of the hard layer.

can be alternatively confirmed by the spin asymmetry (SA) shown in Figure 3.3c and 3.3d for $T = 295$ K and $T = 500$ K, respectively. SA is a quantity expressed as $\frac{R^{++}-R^{--}}{R^{++}+R^{--}}$, which highlights magnetic contributions to the scattering. Both Figure 3.3c and 3.3d show a non-zero SA feature at all H_{ip} and T , indicating the presence of a net in-plane magnetization for all the measurements. Fitted by REFL1D, the nuclear and magnetic depth profiles determined from model fitting are shown in Figure 3.3e and 3.3f, respectively. The fitting constrained the nuclear depth profiles to be the same for all the measurements since the nuclear structure was not expected to change with T and H_{ip} . The magnetic depth profiles here are presented using both MSLD (left axis) and the converted in-plane magnetization (right axis). All the PNR measurements show an appreciable in-plane component of the magnetization. This relaxation follows a power law fall off from the maximal magnetization in the soft layer (M^{top}) to the in-plane component of the magnetization of hard underlayer (M^{bot}). As an example, the position of M^{top} and M^{bot} for $\mu_0 H_{ip} = 100$ mT and $T = 295$ K is demonstrated (dotted blue line). At both $T = 295$ K and $T = 500$ K, increasing H_{ip} results an overall increase of the in-plane magnetization, including M^{top} and M^{bot} , which can be attributed to the increase of in-plane Zeeman energy.

An alternative analysis of the magnetic relaxation is performed by normalizing the magnetic depth profiles by their respective M^{top} , shown in Figure 3.3g. For each H_{ip} , the maximum normalized magnetization ($M/M^{top} = 1$) appears at almost the same position in the soft layer, about 5.8 nm away from the SiO_2 interface. Notably, increasing H_{ip} causes an increase of the interfacial domain wall width, l_{int} , [Li et al. \(2006\)](#) particularly by extending further into the hard layer. The H_{ip} presents an important role in determining the interfacial magnetic configurations by inducing an external Zeeman energy, while increasing T enhances thermal fluctuations in the bilayer, reducing the anisotropy and competing with the exchange coupling. At each H_{ip} , increasing the temperature results in an overall reduction of the total magnetization including M^{top} and M^{bot} . For $\mu_0 H_{ip} = 100$ mT, M^{top} changes from 320 kA/m at 295 K to 180 kA/m at 500 K, more than 40% reduction. This temperature driven reduction of M^{top} decreases with increasing H_{ip} , becoming 30% for $\mu_0 H_{ip} = 300$ mT and only 16% for $\mu_0 H_{ip} = 700$ mT. These results indicate that increasing H_{ip} can compensate the thermal fluctuations, therefore increasing the in-plane soft layer magnetization. The normalized

magnetic depth profiles for each H_{ip} shows that the interfacial magnetic configuration at 500 K is similar to the configuration at 295 K, indicating that there is almost no change in l_{int} or the normalized relaxation rate with temperature between 295 K and 500 K. These PNR experimental results reveal the influence of M_s^{soft} , t_{soft} , H_{ip} and T on the interfacial magnetic configuration, summarized in table 3.1.

3.4 Discussion and Micromagnetic Simulation

The PNR results provide a direct measurement of the magnetic relaxation within the bilayer system; the simulated magnetic depth profiles suggest this relaxation follows a power law from M^{top} to M^{bot} . The PNR measurements show that this magnetic relaxation is dependent on sample geometry (t_{soft}), material properties (M_s^{soft}) and external factors (H_{ip} and T). For t_{soft} larger than a critical thickness, the magnetic configuration changes from a rigid coupling, in which the soft layer has an out-of-plane orientation reflecting the hard underlayer, to an in-plane relaxation, in which the soft layer rotates from out-of-plane to in-plane moving from the interface; this critical soft layer thickness increases with decreasing M_s^{soft} . Increasing H_{ip} is shown to increase the overall in plane magnetization and l_{int} , while increasing T from room temperature to 500 K result in a reduction of the in-plane magnetization, but l_{int} remains almost unchanged. In a simplified model, the in-plane relaxation can be considered as a balance among the magnetostatic energy of the soft layer, perpendicular anisotropy of the hard layer, the interfacial exchange coupling between the soft and hard layers and the Zeeman interaction. The increase of t_{soft} or M_s^{soft} results in the increase of total demagnetization energy, as well as the decrease of energy gap between the ground state to the excited magnon state [Belemuk and Chui \(2013b,a\)](#). When the thermal energy ($k_B T$) is high enough to overcome this energy gap, it will cause the transition from rigid coupling to the underlayer, which favors an in-plane orientation for the soft layer magnetization. As a result, the interfacial magnetic configuration relaxes in-plane. For increasing H_{ip} , the Zeeman energy favors an in-plane orientation parallel to H_{ip} , which increases M^{top} and M^{bot} . However, while increasing H_{ip} , the in-plane magnetic component of the soft layer increases more than the hard layer, due to the much larger saturation magnetization and the absence of

Table 3.1: Summary of PNR experimental results

Sample	t_{soft} (nm)	soft layer MSLD ($\times 10^{-4} \text{ nm}^{-2}$)	Guide field (mT)	Temperature (K)
L1 ₀ -FePt/Al-FePt	2	0.07	50 (out-of-plane)	295
	5	1.46	50 (out-of-plane)	295
	2	0.93	100	295
	2	1.41	300	295
	2	1.77	700	295
	2	0.49	100	500
	2	0.99	300	500
	2	1.44	700	500
[Co/Pd]/CoPd	2	0.01	1	295
	5	0.04	1	295
	9	0.08	1	295
[Co/Pt]/FeNi	1	0.37	1	295
	3	0.22	1	295
	7	0.19	1	295
L1 ₀ -FePt/Fe	9	4.82	1	295

a perpendicular anisotropy. The angle between the magnetization vector of the bottom hard layer and that of the top of the soft layer increases. This increased angle causes the interfacial exchange coupling energy to increase commensurately. To compensate this increase of energy, l_{int} extends further into the hard layer. Increasing T to 500 K, the thermal fluctuations are enhanced which decreases M_s^{soft} , M_s^{hard} and K_u . The reduction of M_s^{soft} decreases l_{int} , while the reduced K_u should increase l_{int} ; these two factors can compensate each other, resulting an almost unchanged l_{int} . Indeed, in the experimental results l_{int} is almost unchanged. Ideally, increasing l_{int} improves writability in HAMR media[51], suggesting that, in this bilayer system, reducing the temperature dependence of M_s^{soft} by e.g. increasing the Curie temperature, would be a promising direction for improvement. The PNR measurements reveal the important roles of t_{soft} , M_s^{soft} , H_{ip} and T in determining the configuration of the magnetic bilayer. The following simulations reinforce these observations and provide a more complete picture which must also include t_{hard} and M_s^{hard} .

The first set of simulations that are presented explore the case of M_s^{soft} (300 kA/m) $< M_s^{hard}$ (900 kA/m), over a range of t_{soft} similar to our experiments with [Co/Pt]/NiFe, shown in Figure 3.4a. For thin t_{soft} (2 nm and 5 nm), the magnetic moments in the soft layer are strictly orientated in the out-of-plane direction. This magnetic configuration can be understood by the classical exchange length, l_{ex} : that for $t_{soft} < l_{ex}$ the interfacial exchange coupling dominates the energy landscape, and rigidly couples the hard and soft layers. Increasing t_{soft} to 9 nm, the in-plane magnetic component of the soft layer begins to increase. Interestingly, the relaxation in the soft layer extends all the way to the interface, challenging the initial idea that the hard/soft exchange coupling is dominant in the interfacial region. Further increasing t_{soft} to 20 nm and 30 nm, the relaxation rate of magnetic configuration in the soft layer decreases, e.g. the soft layer recovers an out-of-plane orientation at the hard/soft interface. These results show a complex and nonmonotonic dependence of the soft-layer orientation on t_{soft} . For completeness, the dependence of the magnetic relaxation on t_{hard} was also investigated in the case of $M_s^{soft}=M_s^{hard}=700$ kA/m, shown in Figure 3.4b. Increasing t_{hard} does not significantly impact the magnetic configuration in the soft layer, indicating that the interfacial exchange interaction dominates the interaction between hard and soft layers.

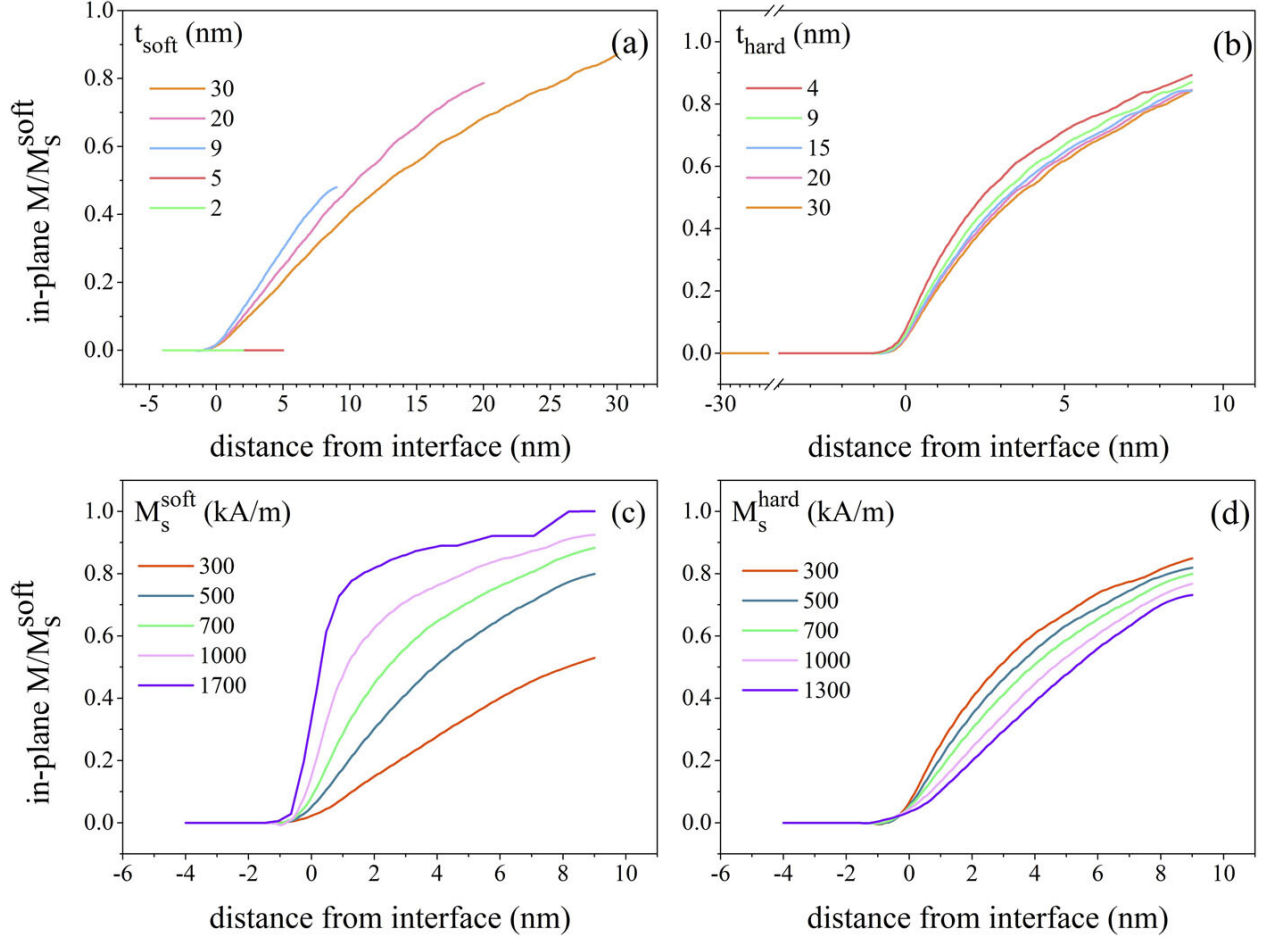


Figure 3.4: OOMMF simulation for the out-of-plane magnetization for (a) $M_s^{hard} = 900$ kA/m and $M_s^{soft} = 300$ kA/m and $t_{hard} = 4$ nm with $t_{soft} = (2, 5, 9, 20, 30)$ nm (b) $M_s^{hard} = 700$ kA/m and $M_s^{soft} = 700$ kA/m and $t_{soft} = 9$ nm with $t_{hard} = (4, 9, 15, 20, 30)$ nm, (c) $M_s^{hard} = 700$ kA/m and $M_s^{soft} = (300, 500, 700, 1000, 1700)$ kA/m (d) $M_s^{soft} = 500$ kA/m and $M_s^{hard} = (300, 500, 700, 1000, 1300)$ kA/m, where $t_{hard} = 4$ nm and $t_{soft} = 9$ nm

These results show a t_{soft} dependent and non- t_{hard} related magnetic relaxation: for $t_{soft} < l_{ex}$, the hard and soft layers are rigidly exchange coupled, while for thicker t_{soft} , the soft layer relaxes in-plane with a relaxation rate which decreases as increasing t_{soft} . The next set of simulations explore the role of M_s^{soft} and M_s^{hard} in determining the magnetic bilayer configuration while fixing t_{soft} and t_{hard} . For a fixed $M_s^{hard} = 700$ kA/m, M_s^{soft} is varied between (300-1700) kA/m, and the out-of-plane magnetization is shown in Figure 3.4c. Increasing M_s^{soft} significantly accelerates the in-plane relaxation of the magnetic soft layer. This acceleration can be seen by evaluating the distance from the hard/soft interface at which the soft layer relaxes in-plane. For $M_s^{soft} = 300$ kA/m, this distance is >30 nm, while for $M_s^{soft} = 1700$ kA/m this distance is only 2 nm. In contrast, models with a fixed M_s^{soft} and variable M_s^{hard} between 300 kA/m and 1300 kA/m, Figure 3.4d, show comparably little change. The bilayer simulations show generally that increasing M_s^{hard} causes the in-plane relaxation to occur over a larger thickness, e.g. the out-of-plane component of the soft-layer magnetization extends further from the hard/soft interface.

These simulations agree with the experimental results. For example, in the FePt/Fe (9 nm) sample the soft-layer was almost fully in-plane, in strong contrast with the [Co/Pd]/CoPd (9 nm) sample which was about fully out-of-plane. The simulations would suggest that the differences in the hard layer have little effect, while the M_s^{soft} dominates the magnetic configuration. Indeed, with a much-larger M_s^{soft} , Fe ($M_s^{soft} = 1700$ kA/m) is expected to be in-plane, while CoPd ($M_s^{soft} \approx 750$ kA/m) could remain out of plane.

3.5 Conclusion

In summary, we have investigated magnetic configurations in perpendicularly-coupled hard/soft bilayer films as a function of their geometric structure (t_{soft}), material properties (M_s^{soft} and M_s^{hard}) and external influences (H_{ip} and T). Using polarized neutron reflectometry, the depth profiles for these coupled systems are experimentally determined and compared to micromagnetic simulations. For very thin t_{soft} , on the order of l_{ex} , the magnetic moments in the soft layer are rigidly coupled to the hard layer throughout their entire thickness, while for thicker t_{soft} the magnetic moments in the soft layer tend

to relax in plane. Surprisingly, the in-plane relaxation extends into regions which were oriented in the out-of-plane direction for smaller t_{soft} , indicating the relaxation is not simply progressive with increasing t_{soft} . The in-plane relaxation is also shown to be dependent on M_s^{soft} , with increasing M_s^{soft} significantly increasing its rate. These results are directly demonstrated in [Co/Pd]/CoPd, [Co/Pt]/FeNi, $L1_0$ -FePt/Fe, and $L1_0$ -FePt/Al-FePt bilayer films using PNR, then confirmed by micromagnetic simulations. Moreover, the simulation results suggest that increasing M_s^{hard} decreases the relaxation rate in the soft layer, while changes in t_{hard} may not significantly influence the magnetic configuration. PNR experiments also reveal the importance of H_{ip} and T on the magnetic configuration. Specifically for the $L1_0$ -FePt/Al-FePt system, increasing H_{ip} or decreasing T results in an overall increase of the in-plane magnetization. This result corresponds to the temperature-dependent balance of the magnetocrystalline anisotropy and the magnetostatic energy. Meanwhile, increasing H_{ip} causes the measured l_{int} to increase, while increasing T leaves l_{int} almost unchanged. The PNR and simulation results together show that the magnetic configuration in the bilayer samples can be modulated by varying t_{soft} , M_s^{hard} and M_s^{soft} as well as H_{ip} and T . Understanding the impact of these parameters is critical for the design of hard/soft bilayer devices.

Chapter 4

Magnetic Skyrmions

This chapter is quoted from "Skyrmion-Excited Spin Wave Fractal Network", contributed by Nan Tang, W.L.N.C. Liyanage, Sergio A. Montoya, Sheena Patel, Elizabeth J. Quigley, Michael R. Fitzsimmons, Sunil Sinha, Alexander J. Grutter, Julie A. Borchers, Eric E. Fullerton, Lisa DeBeer-Schmitt, Dustin A. Gilbert. Project and experiment design was performed by M.R.F., J.A.B., E.F., S.S., L.D.-S. and D.A.G. Sample fabrication was performed by S.M., and ferromagnetic resonance measurements by N.T., W.L.N.C.L. and S.M., SANS measurements were performed over three experiments by N.T., W.L.N.C.L., S.P., L.Q., A.J.G., J.A.B., L.D.-S., and D.A.G. Analysis of the SANS data was performed by N.T., M.R.F., S.S., L.D.-S., A. J. G, J. A. B, and D.A.G. Micromagnetic simulations were performed by D.A.G. The first draft of the text was written by N.T. and D.A.G. All authors made contributions to and approved the final text.

4.1 Motivation

Magnetic skyrmions are chiral spin textures of interest for their fundamental properties which arise from their geometrically-defined topology [Fert et al. \(2013\)](#); [Büttner et al. \(2018\)](#); [Jiang et al. \(2017\)](#); [Sampaio et al. \(2013\)](#); [Nagaosa and Tokura \(2013\)](#). The non-trivial skyrmion topology emerges as a result of the magnetic moments curling into a continuous co-planar loop, bounded on its perimeter and at its core by out-of-plane moments with opposite polarities. This specific geometry gives the skyrmion a non-trivial topology and

associated properties including their specialized dynamic excitations, such as breathing and gyration modes [Montoya et al. \(2017b\)](#); [Dai et al. \(2013\)](#); [Kim et al. \(2014\)](#); [Schwarze et al. \(2015\)](#). Despite intense interest in these excitations for e.g. high frequency skyrmionic devices [Garcia-Sanchez et al. \(2016\)](#), direct, in-situ investigations of skyrmion dynamics have remained challenging due to the combination of their exceedingly small size (10's to 100's of nanometers), high-frequencies (10^9 Hz), and magnetic-only contrast. Even more challenging, the most promising platforms for skyrmionics devices are those with high skyrmion densities which are likely to form ordered lattices or other structures supporting inter-skyrmion coupling and collective dynamics. While the design of any high-frequency skyrmion logic systems will require a detailed understanding of dynamics in high density systems, such information is much more challenging to probe than the skyrmions themselves.

More specifically, the application of high-frequency excitation fields will result in driven skyrmion gyration, where the constantly changing magnetization of the skyrmion causes spin waves to be continuously ejected. These spin waves propagate outward from the skyrmion into the spaces between the skyrmions, where they can interact to form a turbulent magnetic landscape. In most magnetic systems, the wavelength distribution of the spin waves follows a dispersion curve relationship. However, recent works [Satywali et al. \(2021\)](#); [Chen et al. \(2021\)](#); [Srivastava et al. \(2021\)](#) have predicted that the spin waves from skyrmion dynamics manifest with a well-defined periodicity which approximately matches the diameter of the skyrmion. In an ordered skyrmion lattice or other high-density skyrmion system, the spin waves from different skyrmions can constructively and destructively interfere, resulting in an incredibly interactions. Experimental investigations of this dynamic spin wave configuration present enormous challenges, especially in the thin film systems which are by far the most relevant for realizing skyrmionic devices. Nonstandard approaches to the problem are critically needed to obtain insights into the spin wave landscape of a skyrmion lattice, especially in the thin film limit.

To address this issue, we consider neutron scattering techniques which have been critical in the investigation of skyrmions and other magnetic structures. Small angle neutron scattering (SANS), for example, provided the first critical evidence of magnetic skyrmions in MnSi [Mühlbauer et al. \(2009\)](#) and continues to be crucial in their investigation [Desautels](#)

et al. (2019); Meynell et al. (2017); Gilbert et al. (2019). SANS excels as a tool to investigate nanoscale magnetic structures but this success has been challenging to extend to dynamic investigations. To achieve sufficient fluence, SANS instruments operate with a distribution of wavelengths – corresponding to the neutron’s velocity – which limits their time resolution for dynamic investigations to kilohertz. Further, most dynamics studies performed using neutron scattering rely on inelastic scattering in which the energy of the scattered neutrons is resolved. Most SANS instruments are not equipped for this, although recent efforts with MIEZE SANS, SESANS and SEMSANS instruments have shown promise Li et al. (2019); Funama et al. (2021); Li et al. (2021). Other neutron techniques, such as triple axis spectroscopy (TAS), can analyze the energy of the scattered neutrons but struggle to access the small momentum exchange vector (q) necessary to probe the skyrmion length scale Pappas et al. (2011); Bannenberg et al. (2017); Franz et al. (2019); Garst et al. (2017); Weber et al. (2019); Kugler et al. (2015). Instead, we propose to implement SANS-based dynamic investigations by continuously integrating the signal and analyzing the resulting signal as a convolution of all the diffraction patterns.

Specifically, we investigated dynamics in thin film hybrid skyrmion lattices using SANS combined with in-situ and ex-situ ferromagnetic resonance (FMR). Applying a microwave-frequency excitation in the plane of the film with an out-of-plane static magnetic field drives the skyrmions into resonant gyration, causing significant changes to the SANS pattern, including notably the emergence of a new diffraction feature at very small angles. Changes in the SANS pattern are reasonably well fit with a Lorentzian model, representing spin wave excitation. However, a better fit is achieved using a mass-fractal model, which suggests the spin waves form a long-range coherent structure. This spin wave fractal is constructed of fundamental units which match well to the skyrmion diameter and has a cutoff length scale which agrees with the skyrmion lattice spacing. Such an architecture may serve as a unique element in magnon spintronic architectures Chumak et al. (2015); Wang et al. (2021); Kruglyak et al. (2010). Recent works have also shown that dynamic magnonic crystals can also be described as space-time crystals, possessing a periodic structure in both space and time, presenting the possibility that the fractal structure here may be described as a space-time quasi-crystal Träger et al. (2021). These results, interpreted in the context of

a micromagnetic model, provide enhanced insights into skyrmion dynamics and also expand the capabilities of the SANS technique.

4.2 Methods

4.2.1 Sample Fabrication

Thin-films of Ta (50 Å)/[Fe (3.6 Å)/Gd (4.0 Å)]₁₂₀/Ta (50 Å) and Ta (50 Å)/[Fe (3.4 Å)/Gd (4.1 Å)]₁₂₀/Ta (50 Å) were grown on a 1 cm × 1 cm Si (001) substrate using direct current magnetron sputtering. Film growth was performed at room temperature in a 3 mTorr Ar atmosphere. An ultra-high vacuum (UHV) background pressure of $\approx 10^{-9}$ Torr was achieved before deposition to improve the quality of the films. The Ta layers were grown as a seed/adhesion layer and a capping layer against corrosion.

4.2.2 Skyrmion State Preparation

A skyrmion state with local ordering is prepared by the following field sequence: (1) +300 mT out-of-plane static field to saturate the sample, which (2) is then decreased to 0, precipitating a labyrinth domain structure, then (3) apply an out-of-plane field of +195 mT, which precipitates the magnetic skyrmions. Following this sequence the skyrmions form domains with short-range (several micron) hexagonal ordering but no long-range coherent orientation.

A skyrmion state with long-range ordering is prepared by the following field sequence: (1) A saturating magnetic field (+500 mT) is applied at 45° relative to the film normal direction, which (2) is then decreased to 0, resulting in magnetic stripe domains; the orientation of the stripes is determined by the in-plane component of the magnetic field. (3) Then a +195 mT field is applied along the film normal direction, breaking up the stripes to form hybrid skyrmions. The skyrmions form a hexagonally packed lattice, now with a long-range orientation defined by the initial stripe phase.

4.2.3 FMR Measurements Set Up

The FMR measurements were performed using self-built grounded coplanar waveguides (CPWs) connecting to a vector network analyzer (VNA) system. This VNA system can provide an alternating current through the CPWs with a range of frequency from 500 kHz to 32 GHz, thus generating a magnetic field which encircles the stripline. The Fe/Gd thin film is then mounted face to the CPW; the generated magnetic field is thus in the plane of the film. Then the waveguide is inserted into a physical property measurement system (PPMS), which is used to control the temperature (0 to 400 K) and static magnetic field (-9 T to $+9$ T); this static magnetic field is along the film normal direction.

The sample was then placed film side down onto the CPW. Applying the high frequency current through the stripline generates a high-frequency magnetic field in the plane of the film. At resonance, this magnetic field excites the magnetic skyrmions into resonant dynamic modes; the energy transfer from the stripline to sample is identifiable in the electronic transmission through the stripline, $S_{1,2}$. Measurements were performed using a network analyzer to probe frequencies between 1 GHz and 3 GHz, with static out-of-plane magnetic fields between 0 and 300 mT. This measurement concludes in the saturated state. Next, the skyrmion state is reset by the same preparation process and $S_{1,2}$ was again measured at static fields between 0 and -300 mT.

4.2.4 SANS Measurements Set Up

Small-Angle Neutron Scattering (SANS) experiments were performed on the GPSANS instrument at the High-Flux Isotope Reactor and the vSANS instrument at the NIST Center for Neutron Research. Measurements on GPSANS were taken at the maximal sample-detector distance (20 m), using long-wavelength neutrons ($\lambda = 16$ Å, $\Delta\lambda/\lambda = 0.13$). Data from GPSANS were reduced using the GRASP software package; data from vSANS were reduced using the IgorPro Software. A static magnetic field and the neutron beam were aligned with the film surface normal direction. A two-turn solenoid was arranged with one turn on either side of the sample, causing the magnetic component of the excitation field to be in the plane of the film. The solenoid was driven by a 5 W RF amplifier, coupled to

an RF generator. Measurements were performed at room temperature both by fixing the frequency and sweeping the magnetic field, and fixing the field and sweeping the frequency.

4.2.5 OOMMF Simulation Parameters

Three dimensional micromagnetic simulations for Fe/Gd multi-layers were performed for an individual skyrmion in simulation volume of ($x = 304$ nm, $y = 280$ nm, $z = 106$ nm) using a rectangular mesh ($x = 8$ nm, $y = 8$ nm, $z = 2$ nm). The material parameters in the simulated block include the exchange stiffness ($A = 0.7 \times 10^{-11}$ J/m), the saturation magnetization ($M_s = 3.5165 \times 10^5$ A/m) and the uniaxial anisotropy along z direction ($K_u = 1.1 \times 10^5$ J/m³). This exchange stiffness is chosen to be slightly smaller than the one in Ni (0.9×10^{-11} J/m), while M_s is directly measured by magnetometry and K_u is calculated using the hard-axis saturation field. These material parameters resulted in the stabilization of the hybrid skyrmion without an external magnetic field, similar to the experimental results; a significant decrease or increase of the value of these material parameters was found to cause the skyrmion to collapse. The stabilized single skyrmion with these parameters has a diameter of ≈ 150 nm. The static and dynamic states are then prepared by applying magnetic fields ($B_x = 0$, $B_y = 0$, $B_z = 50$ mT) and ($B_x=50$ mT, $B_y=0$, $B_z=50$ mT), respectively.

4.3 Results

4.3.1 Hybrid Skyrmion Thin films

Thin films of Ta(50 Å)/[Fe(3.6 Å)/Gd(4.0 Å)]₁₂₀/Ta(50 Å) were grown by sputtering as described previously [Montoya et al. \(2017a,b\)](#) and in the methods [4.2.1](#). Skyrmions in this system have a hybrid structure which can be described as a Bloch-type skyrmion at the midpoint along the length of the skyrmion tube, e.g. its equator, with flux-closure domains – which emulate Néel skyrmions – at either surface [Montoya et al. \(2017b,a\)](#). The flux-closure domains form to contain the dipole fields from the film, reducing the magnetostatic energy; the orientation of the Néel structure will follow the dipolar fields, which extend in opposite directions on the film’s top and bottom surfaces, defining opposite chiralities to the

skyrmion surface spin structure. The chirality of the Bloch region can be either clockwise or counter-clockwise and occurs with equal probability [Montoya et al. \(2017a\)](#). This structure provides for excellent stability – including at ambient temperatures and zero field - and unique opportunities for sophisticated magnetic dynamics. This system was chosen because it is the only thin film skyrmion which has been successfully probed with SANS to-date [Desautels et al. \(2019\)](#).

The skyrmion state was prepared with either short-range or long-range ordering using the field sequence described in the Methods section. These sequences prepare skyrmions with cores which are anti-parallel to the applied field (e.g., oriented in the negative out-of-plane direction), surrounded by a matrix which is in the positive orientation, parallel to the formerly applied field. These skyrmions have been shown to be stable in the range $-30 \text{ mT} < \mu_0 H < 200 \text{ mT}$ [Desautels et al. \(2019\)](#).

4.3.2 Ferromagnetic resonance

For the FMR measurement the skyrmion state was prepared without tilting the sample, e.g. as domains with local hexagonal ordering, but no long-range orientation. FMR measurements were performed using a ‘flip-chip’ geometry with a grounded coplanar waveguide, generating an in-plane microwave excitation, with an out-of-plane static magnetic field. Figure 4.1 shows a 2D heat map of the transmission spectra $S_{1,2}$ measured as a function of frequency, f , and magnetic field, H . The heat map shows two series of measurements, each of which started from the skyrmion state at remanence: for $H < 0$ the measurement proceeded from $H = 0$ to $\mu_0 H = -300 \text{ mT}$, and for $H > 0$ the measurement proceeded from $H = 0$ to $\mu_0 H = +300 \text{ mT}$; strong absorption, corresponding to resonant magnetic excitations, is indicated by the yellow contrast. The absorption map can be identified by seven regions indicated as (I-VII) in Figure 1. In Region I the skyrmions are stable in their hexagonally close packed lattice. The data show strong resonant absorption, indicated by a drop in the $S_{1,2}$ intensity (yellow in the figure), following a ray which starts at $(H = 0, f = 1.7 \text{ GHz})$, and ends at $(\mu_0 H = 190 \text{ mT}, f = 2.4 \text{ GHz})$. In the above-described measurement configuration, these dynamics are expected to correspond to in-plane gyration modes of the skyrmions. Increasing the magnetic field beyond 190 mT into Region II, the frequency

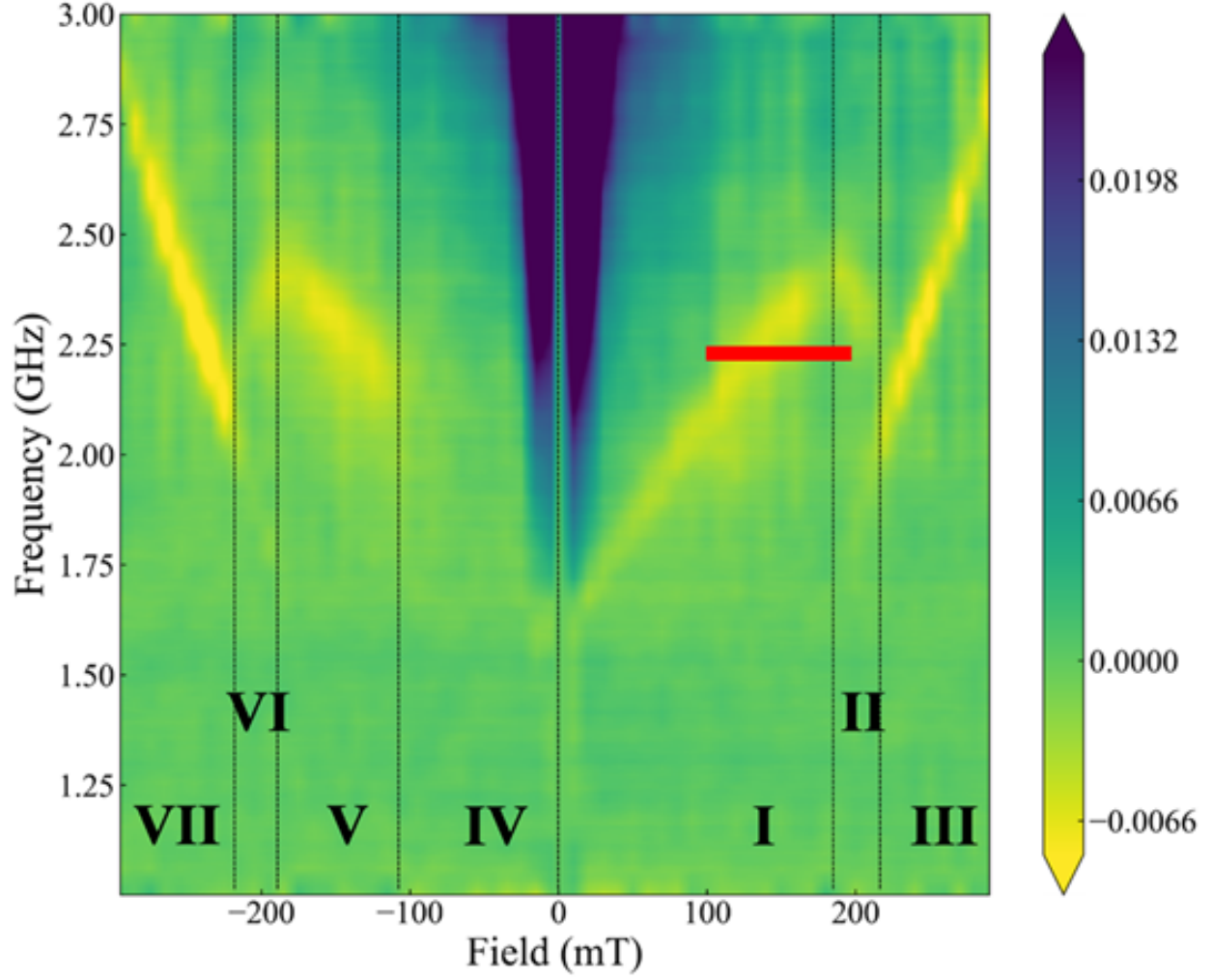


Figure 4.1: Ferromagnetic resonance (FMR) of Fe/Gd multi-layers at room temperature, plotting the transmission coefficient $S_{1,2}$ as a function of frequency and out-of-plane magnetic field. Yellow indicates regions of high absorption. The labeled regions correspond to (I, V) locally ordered skyrmions, (II, VI) destruction of the skyrmion lattice, (III, VII) saturated state, and (IV) labyrinth domains. The red line indicates the nominal range of the SANS measurements.

corresponding to resonance rapidly decreases with increasing field. Further increasing the field beyond 220 mT (Region III), the resonant frequency again increases in response to an increasing static field. In this region the sample is saturated, and resonance corresponds to coherent, long-range spin gyrations, e.g. Kittel resonance. This implies that Region II is a transition region, in-which skyrmions are presumably being annihilated and the lattice destroyed.

In the negative field range $-110 \text{ mT} < \mu_0 H < 0 \text{ mT}$, identified as Region IV, no apparent resonant signal is observed. In this region, the skyrmions are expected to annihilate, forming a labyrinth-like worm domain structure [Montoya et al. \(2017b,a\)](#). The disorder and weak symmetry breaking between the up- and down-oriented domains may result in the suppression of long-range spin dynamics. Decreasing the field to Region V (110 mT to -190 mT), the dynamics reappear, albeit slightly weaker and broader than the positive field analogue (e.g. Region I). This suggests the skyrmions re-emerge in this field range, which is much lower than reported previously with SANS [Desautels et al. \(2019\)](#). However, the SANS results identify the ordered skyrmion lattice, not the emergence of dilute, disordered skyrmions, which is consistent with previous X-ray microscopy results [Montoya et al. \(2017b,a\)](#). Regions VI and VII are largely symmetric with their positive field counterparts (Regions II and III, respectively) and indicate a transition region and saturated state, respectively. These FMR results are consistent with the previous static SANS measurements [Desautels et al. \(2019\)](#) and provide a preliminary understanding of skyrmion resonant modes. This map of the resonant modes is used to guide the field and frequency conditions for the subsequent SANS measurements.

4.3.3 Small Angle Neutron Scattering

The SANS measurements discussed in the main text were performed on the GPSANS instrument at the High-Flux Isotope Reactor and the results discussed in Appendix B.4 were taken on the vSANS instrument at the NIST Center for Neutron Research. For the measurements on GPSANS, five films were stacked for the measurement to increase the signal. These samples were prepared to be in a long-range ordered skyrmion lattice following a magnetic field sequence reported previously [Desautels et al. \(2019\)](#) and in Methods 4.2.2.

The sample was placed in a two-turn solenoid, with the film in-plane direction aligned with the solenoid axis, and the neutron beam and static field aligned with the film normal direction. The resonant frequency of the solenoid was measured to be 2.23 GHz using a network analyzer. A schematic of experimental set up is shown in Figure B.1.

To excite the skyrmion gyration mode, a static magnetic field was applied along the film normal direction, which was also parallel to the neutron flux, and the frequency generator was turned on, energizing the two-turn solenoid. Using a 5 W amplifier, the two-turn solenoid generates a maximal in-plane field of ≈ 0.03 mT. Conceptually, analogous out-of-plane measurements could be performed to excite the breathing mode but were not performed here since the high frequency would induce significant eddy currents and sample heating in the metallic films. Fixing the in-plane excitation frequency, f , at 2.23 GHz, the out-of-plane magnetic field was sequentially increased from 100 mT to 190 mT, marked with a red line in Figure 4.1, with four example SANS patterns shown in Figure 4.2a-d; note that all four panels in Figure 4.2a-d are plotted on the same intensity scale. The SANS plots are discussed in terms of their structure, which consists of the momentum transfer vector – the radial component – and the azimuthal scattering angle – the angle of scattering in the plane of the detector. At $\mu_0 H = 130$ mT, the six-fold diffraction pattern is readily apparent in the SANS data, confirming the skyrmions are stable in their hexagonally ordered lattice with a long-range orientation and that the applied dynamic fields are not distorting the lattice in any detectable way. The background appears as low-intensity diffuse scattering typical of SANS measurements. Increasing the applied field to 150 mT, the six-fold pattern is still easily identified, but the background scattering, especially in the low- q region, starts to increase. Further increasing the applied field to 180 mT, the six-fold pattern is hard to distinguish within the increasing, angle-independent background intensity. As the applied field finally increases to 190 mT, the six-fold pattern becomes apparent again and the intensity of the low- q feature decreases significantly. Notably, the hexagonal SANS pattern has the same orientation after passing through resonance, suggesting long-range order was preserved throughout the measurement sequence. The distinction between these different states is highlighted in Figure 4.2e, which shows the intensity versus azimuthal angle, integrated over a range $q = 0.0029 \text{ \AA}^{-1} \pm 0.0005 \text{ \AA}^{-1}$, capturing the change in scattering

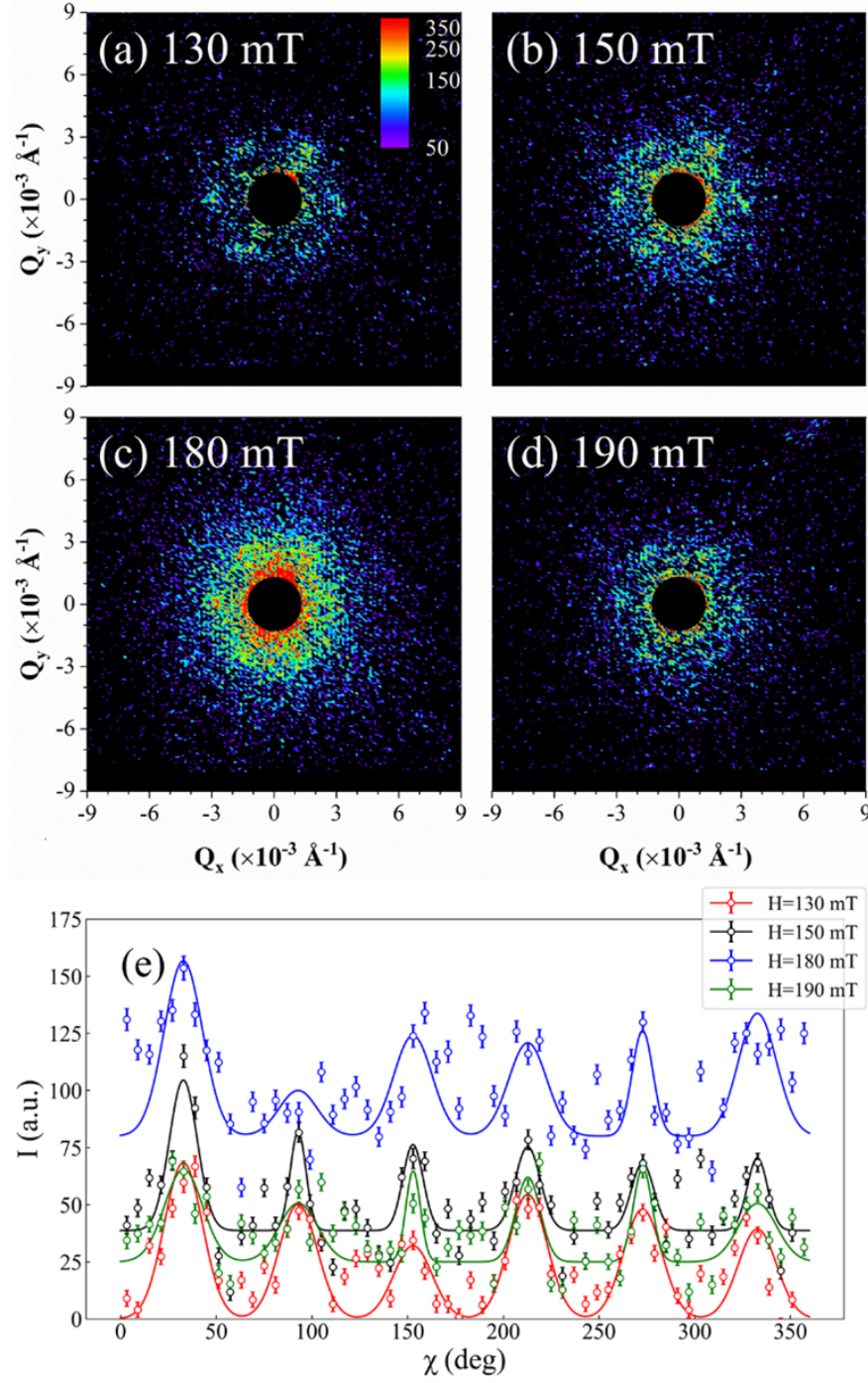


Figure 4.2: 2D SANS pattern of magnetic skyrmions under different magnetic fields and excitation frequencies: (a) off-resonant state ($\mu_0 H = 130$ mT, $f = 2.23$ GHz), (b) initial-resonance state ($\mu_0 = 150$ mT, $f = 2.23$ GHz), (c) on-resonance state ($\mu_0 = 180$ mT, $f = 2.23$ GHz) and (d) post-resonance state ($\mu_0 = 190$ mT, $f = 2.23$ GHz). All 2D images are plotted with the same intensity scale. (e) Intensity vs azimuthal angle ($q = 0.0029 \text{ \AA}^{-1}$, $\Delta q = 0.0005 \text{ \AA}^{-1}$)

intensity of the hexagonal diffraction pattern at different angles. Interestingly, these data show that the resonance increases the total scattering intensity (i.e., integrated intensity in six-fold skyrmion peaks plus the low- q feature). The additional scattered or transmitted neutrons are presumably located at $q \approx 0$ in the static measurements and shift to higher q outside the beamstop in the dynamic case. However, their appearance at low- q in the latter case would tend to suggest large real-space structure, which is not expected from the skyrmion gyrations. Identifying the 180 mT measurement as corresponding to magnetic resonance, the FMR spectrum in Figure 4.1 is suggested to have a small difference in the field calibrations. This series of measurements indicate that the SANS signal is indeed sensitive to the resonant skyrmion dynamics, with clear distinctions between the on-resonance and off-resonance signals.

4.3.4 Micromagnetic Structure of a Hybrid Skyrmion

In the described experimental configuration, the skyrmion lattice exists in the plane of the Gd/Fe thin film with a static magnetic field applied along the film normal direction, parallel to the skyrmion tubes, and an in-plane excitation field. This geometry excites gyration of the skyrmion tube with the orbit in the plane of the film. Previous simulations performed on conventional skyrmion tubes reported that the structure of the skyrmion during its resonant motion can be significantly distorted, especially for higher-order dynamics [Kim et al. \(2014\)](#); [Montoya et al. \(2017b\)](#); [Garst et al. \(2017\)](#); [Seki et al. \(2020\)](#). In the current system, even the lowest energy gyration mode will be complicated by the three part structure of the hybrid skyrmion. As discussed in Section 4.3.1, an individual hybrid skyrmion is comprised of a Bloch type winding at the equatorial band and two Néel skyrmions at the ends of the skyrmion tube; the Néel skyrmions have opposite chiralities as determined by the dipolar fields emanating from the skyrmion core. For the static state shown in Figure 4.3a, the in-plane magnetic components along the $+/-x$ and $+/-y$ directions are balanced, while the core is centered within the chiral winding. Applying the excitation field along the x direction, the structure of the dynamic skyrmion is fixed at each extremum of the driving cycle, with the domains parallel/antiparallel to the excitation field expanding/contracting and displacing the core [Satywali et al. \(2021\)](#); [Weber et al. \(2022\)](#), as shown illustratively in

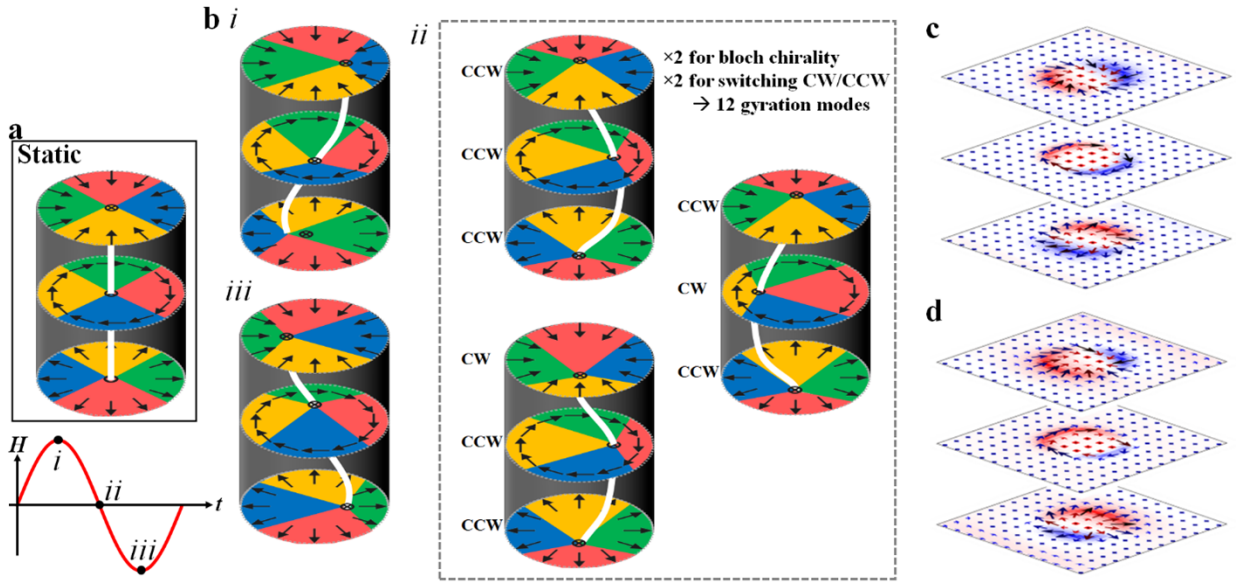


Figure 4.3: Illustrative diagrams showing the (a) static and (b) dynamic gyration of a hybrid skyrmion. At the extremum of the excitation (panel b, i and iii) the magnetic configuration is well defined, however, the gyration between these states can be either clockwise or counterclockwise, shown in panel b(ii). Micromagnetic simulations of (c) the static and (d) dynamic state; the magnetic configurations are analogous to panels (a) and the extremum field shown illustratively in panel b(i).

Figure 4.3b(i) for the positive extremum and Figure 4.3b(iii) for the negative. Connecting the center of the chiral winding from each section reveals that the core has a corkscrew-like shape along the length of the tube. After the extremum of the excitation field, the skyrmion core undergoes damped precession following the Landau–Lifshitz–Gilbert equation [Lakshmanan \(2011\)](#). Without a transverse field (in $\pm y$) or symmetry-breaking energy term (such as the Dzyaloshinskii–Moriya interaction (DMI)) each of the Néel and Bloch regions may gyrate clockwise (CW) or counterclockwise (CCW). Illustrations of three CW/CCW modes are shown in Figure 4.3b(ii); each dynamic structure is expected to have a different energy due to distortions of the core and the overlap of parallel domains in each layer. The resonant frequency of the precession is determined by the material properties (K_U , A , M_s) and applied magnetic field. In the gyration mode, the core is expected to follow an approximately circular orbit around the equilibrium center, with one precession per cycle of the excitation. The structure of the skyrmion in the static and dynamic cases were verified using the object oriented micro magnetic framework (OOMMF), [Donahue \(1999\)](#) shown in Figure 4.3c and 4.3d, respectively. The dynamic model is presented in the extremum state, similar to Figure 4.3b(i). Notice that the regions parallel to the applied field (in $+x$ for this example) in the dynamic models have grown while antiparallel regions have shrunk, similar to the illustrations in Figure 4.3b.

Each of the dynamic modes are doubly degenerate since the gyration direction (CW and CCW) can be exchanged, and double again since the direction of the chiral winding in the Bloch region can be switched (CW in the image). This results in [Boulle et al. \(2016\)](#) unique gyration modes which will exist in the system. Each of these modes will eject a unique time- and depth-dependent collection of spin-waves into the film as their magnetization is continuously changing in time. This distribution causes the local spin wave environment to be disordered between unit-cells of the skyrmion lattice or from skyrmion to skyrmion, depending on the local gyration modes. However, these spin waves will have some common features since each skyrmion is driven by the same excitation field.

4.4 Quantifying the Effects of Gyration on Skyrmion Lattice

4.4.1 Mass-fractal Model

Integrating the SANS pattern across all azimuthal angles at fixed radii (q) generates a plot of intensity (I) versus q , shown in Figure 4.4a. The I vs q data measured away from resonance ($\mu_0 H = 130$ mT) shows a well-defined peak at $q \approx 0.003 \text{ \AA}^{-1}$ corresponding to the hexagonal skyrmion diffraction pattern, but little else. This peak appears in all of the measurements and is modeled by a general Gaussian function, $A_1 \exp(-\frac{(q-\mu)^2}{\sigma})$, where A_1 is the Gaussian amplitude, σ is the peak width, and μ is the central peak position. As the applied field is increased towards 180 mT, a broad scattering feature appears at low- q , which rapidly decays. This decaying feature cannot be reasonably fit by many of the expected functions, including a power law (representing standard Porod-like scattering) in Appendix B.3 or second-order exponential $\exp(-\frac{q^2}{c})$, which would represent quasielastic scattering), shown in Appendix B.3.1. A Lorentzian-squared function of the form $(\frac{A_2}{(1/\xi)^2 + q^2})^2$, which has been previously used to model frozen spin textures and spin waves Motoya et al. (1983); del Moral (1992); Schroeder et al. (2020); Tian et al. (2009); Sachan et al. (2008); Grigoriev et al. (2019), was also tested and again provided a poor fit, discussed in Appendix B.3.2.

The best fitting model, shown in Figure 4.4, was, surprisingly, a mass fractal model Mildner and Hall (1986). This model describes systems which have an extended, chain-like or networked structure constructed out of approximately monodisperse fundamental units. Such structures are commonplace in nanoparticle systems, where the fundamental building blocks (the nanoparticles) aggregate by e.g. van der Waals forces to form extended networks. This model is also commonly used to describe polymeric chains in-which regularly structured monomers form extended, cross-linked networks. Reflecting this physical interpretation, the mass fractal model has four variables: radius of the building block (R), mass fractal dimension (D), cut-off length (γ) and scale factor (A_3). Applying a mass fractal+Gaussian model to the entire dataset, Figure 4.4a shows a good fit, with the converged parameters shown in Figure 4.4b. By subtracting the 130 mT data as an effective off-resonance

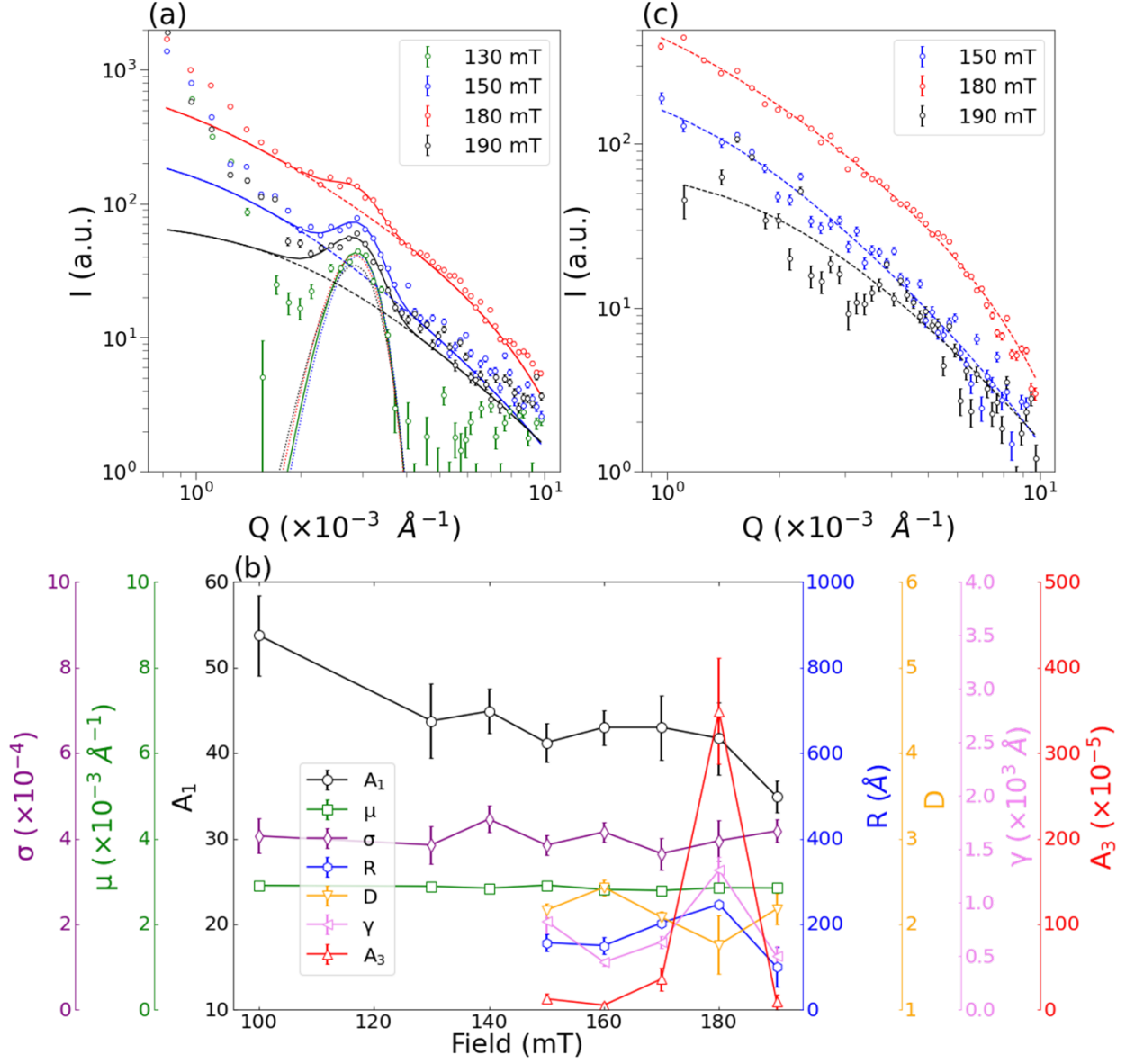


Figure 4.4: (a) I vs q plots for the off-resonant (red), pre-resonance (blue), on-resonance (black) and post-resonance (green) states. The data are represented by points and the fitted models are shown as solid lines; the Gaussian component of the model is shown as a dotted line and the mass fractal model as a dashed line. (b) The total neutrons intensity between $q = (0.0018 \text{ \AA}^{-1}, 0.01 \text{ \AA}^{-1})$ and the corresponding fitting parameters: Gaussian distribution amplitude (A_1), width (w) and central peak position (μ), and radius of the building block (R), mass fractal dimension (D), cut-off length (γ) and scale factor (A_3). (c) I vs q plots for pre-resonance (blue), on-resonance (black) and post-resonance (green) states using the 130 mT data used as a dynamic-independent background, then fitted with the mass fractal + Gaussian model.

background, the mass fractal model is able to achieve a high-quality fit across the entire data range ($0.0007 \text{ \AA}^{-1} < q < 0.01 \text{ \AA}^{-1}$), shown in Figure 4.4c. The parameters for the fits show that the mass fractal behavior is strongly active during resonance, as indicated by the scale parameter, A_3 . The converged model shows that the radius of the fundamental building blocks (R) that make up the fractal structure is approximately $150 \text{ \AA} - 200 \text{ \AA}$, corresponding to a diameter of $300 \text{ \AA} - 400 \text{ \AA}$; this length scale is consistent with the magnetic correlation length identified in the Lorentzian model in the Figure B.4. This length is speculated to correspond to the wavelength of the spin waves. Next, the cut-off length (γ), which indicates the largest distance between points within the fractal, is shown to be $\approx 500 \text{ \AA}$ for most of the measurements, but increases to $1500 \text{ \AA}$ on resonance. The last parameter in the mass fractal model is the fractal dimension, which is between 2 and 2.5, consistent with a structure that is largely confined to the plane of the thin film.

The mass fractal model is conventionally used to describe a system constructed of monodisperse structures ordered into a fractal network. In the current system, the correlation between the mass fractal intensity, A_3 , and the magnetic resonance suggests that spin waves take up this role. However, spin waves generally follow an energy-dependent dispersion curve, with a continuum of wavevectors (and corresponding wavelengths), making them inherently polydisperse; the fractal model implies that the spin waves have a well-defined length scale. It is also notable that this length scale is similar to the radius of the skyrmion, previously measured in similar films to be $\approx 350 \text{ \AA}$ [Montoya et al. \(2017a\)](#). This correlation between the length scale of the spin wave and the skyrmion radius was recently described by Satywali et al [Satywali et al. \(2021\)](#). This work explains that, during gyration dynamics, a magnon-skyrmion bound pair forms with the maximum of the magnon eigenfunction coinciding with the skyrmion radius, and rapidly decaying at both larger and smaller length scales. Considering the data in this context, we revisit the cut-off length (γ), which is $\approx 500 \text{ \AA}$ for the off-resonant conditions, increasing to $\approx 1500 \text{ \AA}$ on resonance. At $500 \text{ \AA}$, the fractal features are smaller than the skyrmion and could be largely bound within it. On resonance, γ is nearly the size of the skyrmion lattice ($2200 \text{ \AA}$), and matches very well with the lattice spacing minus the skyrmion diameter ($1500 \text{ \AA}$), e.g., the interstitial space between skyrmions. This implies that the fractal now fills the interstitial space between the skyrmions. Satywali

et al [Satywali et al. \(2021\)](#). measured two dynamic modes, one low frequency mode which is largely bound to the skyrmion, and one high frequency which results from dynamics between skyrmions. Our data may be probing these two conditions: in the off-resonant case the excitation drives weak dynamics, leaving the skyrmions largely non-interacting (that is, not interacting through magnons) and behaving as Satywali’s low-frequency mode, while in the resonant state the strong dynamics increase the strength of the spin wave emissions, resulting in coupling and the high frequency mode. What was not expected is that these ejected spin waves would form an effective fractal structure of their own.

4.4.2 Micromagnetic Simulations of Skyrmion Lattice Dynamics

Micromagnetic simulations are performed as a real-space complement to the above experimental data. An array of 16 hybrid skyrmions were prepared using the parameters presented in Methods [4.2.5](#). A static magnetic field of 10 mT is along the skyrmion tubes and an oscillatory field applied along the x-axis of the form $A_{field}\sin(Bt^3)$, where A_{field} is the maximal amplitude of the oscillatory field, and B is a constant of order 10^{24} , as shown in Figure [4.5a](#). Using this form, the frequency of the sinusoidal field increases from zero to 5 GHz over a simulation time of ≈ 25 ns. At resonance, the skyrmion undergoes gyration, resulting in a net magnetization in the y-direction, m_y , shown in Figure. [4.5b](#). Applying a Fourier transform to m_y identifies the resonance frequencies as 1.6 GHz, 3.6 GHz, and 4.8 GHz. The resonance at 1.6 GHz agrees well with our experimental observations in Figure [4.1](#) and Figure [B.5](#), [B.6](#) and [B.7](#), which showed that the resonance is ≈ 1.7 GHz for one sample and ≈ 1.3 GHz for the other. Next, the excitation frequency was fixed at 1.6 GHz, driving the system at resonance. After eight cycles the system is presumed to be in a stable dynamic state and the magnetization in the z- and y-directions are exported for visualization, Figure [4.5c](#) and [4.5d](#); videos of these dynamic modes are shown in the Supplemental Material. Similar images taken in the off-resonance condition (frequency of 1 GHz) are shown in Figure [4.5e](#) and [4.5f](#), and videos provided in the supplement, for comparison. For the z-contrasted images, red (blue) indicates magnetization in +z (-z); in the y-contrasted images red (blue) indicates magnetization in +y (-y).

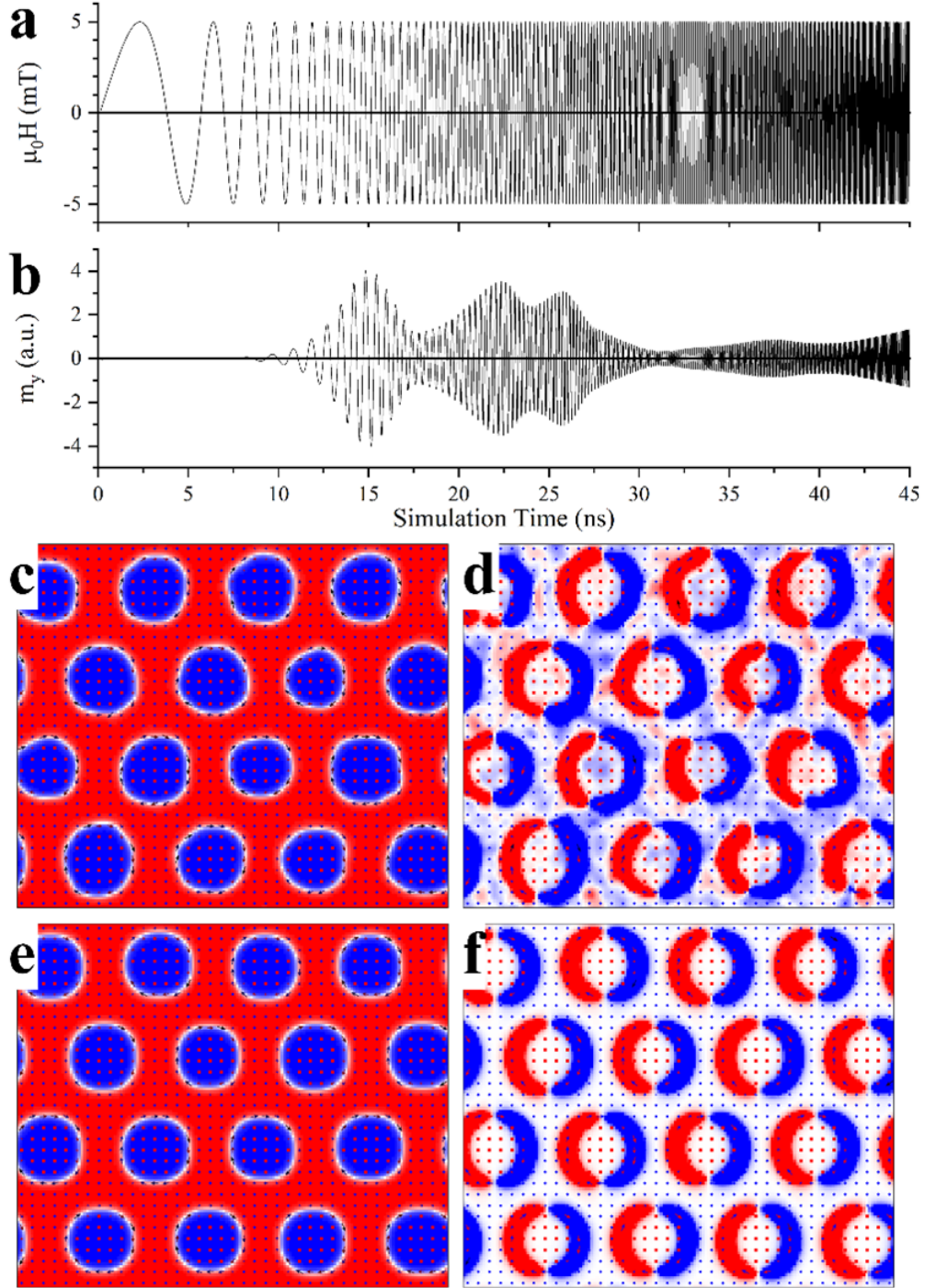


Figure 4.5: (a) Applied in-plane excitation field, applied parallel to the x-axis, and (b) net magnetization along the y-axis. Resonance occurs at 1.6 GHz, 3.6 GHz and 4.8 GHz. Micromagnetic results showing the magnetization along (c) the out-of-plane z-direction and (d) in-plane y-direction, for an excitation frequency of 1.6 GHz; magnetization along the (e) z- and (f) y-axes, driven off-resonance at 1 GHz.

In the z-contrasted images the skyrmions at resonance are not circular and have a distribution of sizes when compared to the off-resonance images, which show circular structures with a regular size. Both images show the skyrmions in an ordered hexagonal lattice, which is consistent with the SANS results. Z-contrasted video of the dynamics confirm that the skyrmions gyrate, but also that there are additional modes which propagate along the boundary of the textures. The irregular shape and edge modes may indicate that the driving frequency is slightly off resonance or may be the result of spin-wave mediated interactions between skyrmions. Video of the off-resonance condition shows minimal motion from the skyrmions. Notably, the spin waves are not visible in the z-contrasted video or images.

In the y-contrasted images captured during resonance the boundaries of the skyrmion lattice are clearly visible, but there is also a turbulent background resulting from the spin waves. Video of this state shows that the spin waves move very rapidly across the entire simulation space. Notably, the size of the magnetic regions are approximately regular – there are very few red, blue or white regions which are exceedingly small or large. The radius of the regions can be measured from the images and is found to be $\approx 220 \pm 30 \text{ \AA}$, which is in reasonable agreement with the SANS results, which identified the radius of the ‘building blocks’ as 150-200 $\text{\AA}$. Again, images taken in the off-resonance condition do not show any appreciable magnetic contrast beyond the static skyrmion lattice.

4.5 Discussion

Previous works have demonstrated that the periodic spatial modulation of the magnetization which results from the skyrmion lattice represents a natural two-dimensional magnonic crystal [Kruglyak et al. \(2010\)](#); [Ma et al. \(2015\)](#); [Nikitov et al. \(2001\)](#). Accordingly, the collective ordering enforces a specific magnon spectrum [Roldán-Molina et al. \(2015\)](#) which, in this case, is driven by the peaked spin wave emissions [Satywali et al. \(2021\)](#); [Chen et al. \(2021\)](#); [Srivastava et al. \(2021\)](#) from the gyrating skyrmions. The resulting magnetic configuration, revealed by SANS includes the original skyrmion lattice as well as a magnetic fractal structure. This fractal structure, being both periodic and self-similar, can be well

described as a (spatial) quasicrystal. However, this structure has a discrete translational symmetry in time. That is, the structure regularly repeats itself in time, locked to the driving frequency of the excitation field. Recent works have shown that driven spin waves in magnetic thin films can result in space-time crystals [Träger et al. \(2021\)](#); [Wilczek \(2012\)](#); [Khemani et al. \(2019\)](#). A similar space-time crystal (crystalline in time, quasicrystalline in space) may exist here, but would necessarily require resolving the nanoscale phase of the spin waves, which is beyond the scope of the current work.

These in situ SANS results provide several insights into the dynamics of hybrid magnetic skyrmions: (1) the skyrmion lattice is preserved and does not change significantly passing through resonance, suggestive of their robust topological protection, (2) the skyrmion dynamics eject spin waves with an associated length scale of ≈ 300 Å- 400 Å which are well represented with a mass-fractal model, (3) fitting with the mass fractal model suggests that the spin waves at resonance extend outside of the individual skyrmions to form an extended structure analogous to a fractal network, (4) the observed mass fractal model supports the simulation and theory predictions presented by Satywali et al. These last three observations provide new insights into the complex magnetic landscape which is generated during skyrmion dynamics and suggests the emergence of a macroscopic magnetic superstructure. While the correlation of the FMR and SANS results and subsequent high-quality fit offer compelling evidence that these results are correlated, considerations of alternative models are provided in the Supplemental Material.

This result presents a unique insight into magnetic dynamics and spin waves which may be applied beyond skyrmions to other magnetic systems. Specifically, the qualities of phonons and spin waves (magnons) are conventionally measured using inelastic neutron scattering techniques such as triple axis spectroscopy (TAS) and time-of-flight (ToF) spectroscopy. In these techniques, the energy loss and change in momentum of the scattered neutron is measured relative to the structural (elastic) scattering peaks. However, the momentum transfer presented in these techniques is different from the momentum transfer vector discussed in SANS diffraction measurements which were performed here; this distinction is understood in scattering jargon as little-q (momentum gain or loss relative to an elastic scattering event) versus big-Q (momentum transfer vector, change in the momentum

direction but not magnitude). The momentum transfer in TAS and ToF does not explicitly encode the real space structure of the phonon or spin wave, only its direction, energy and total momentum. These other qualities can be derived from the dispersion curve, but a direct relationship, such as $d=2\pi/q$ where d is the real space periodicity, which correlates the real-space structure to the diffraction peak location in elastic measurements, does not exist. The SANS measurements performed here, measured at unusually low- q from the spin waves at the elastic position and does encode their real space structure, which is exceptionally rare.

4.6 Conclusion

We have successfully observed the gyration mode in magnetic skyrmions using SANS as an in-situ probe. The gyration modes for the skyrmion lattice, including resonant and off-resonant states, can be excited by an in-plane gigahertz fields and an out-of-plane magnetic field. Throughout the measurements, including on- and off-resonance, the excitation field has a negligible effect on the structure of skyrmion lattice. However, during resonant gyration, a strong enhancement of background intensity in the form of a low q decay function, increasing the total scattering intensity by more than four-fold, is reported. This function can be fit with a Lorentzian squared function, corresponding to a collection of disordered magnetic clusters, but is better fit using a mass fractal model. The mass fractal model, which represents systems constructed of monodispersed elements coordinated into a fractal network, is attributed to the collection of spin waves ejected during gyration. Notably, the lack of significant changes in the scattering from the skyrmion lattice and precipitation of an ordered spin wave network are surprising results. These measurements provide surprising new insights into an emergent long-range structure within the skyrmion lattice during dynamic gyration and suggests that SANS could become a relevant technique to investigate high-frequency magnetization dynamics.

Chapter 5

Summary

This dissertation focuses on magnetic soft-hard bilayer and skyrmions. We have systematically designed sample geometry and material properties in a variety of soft-hard bilayer thin film samples. Using these samples, we have studied the interfacial magnetic configurations as probed using PNR. The skyrmions discussed in this dissertation are formed in Fe/Gd multi-layer thin films. We have successfully used SANS to probe the in-situ dynamic modes of these skyrmions. The SANS results are well represented by a mass-fractal model which suggests the formation of spin wave network for on-resonant skyrmions. These experiments results are also supported by micromagnetic simulations.

In Chapter 1, I introduce the relationships between electrons, magnetic moments and angular momentum. I then discuss the underlying energies associated with magnetic behavior. Key among these, the exchange interaction between electrons contribute to magnetic ordering, which includes ferromagnetism and antiferromagnetism. Next, the main magnetic interactions are summarized. These interactions compete with each other, which results the magnetic domain and domain wall structures. Finally, I give a brief introduction of two specific domain wall structures.

In Chapter 2, the main characterization methods I used to study these magnetic textures are summarized, which includes elastic neutron scattering, vibrating sample magnetometry (VSM) and ferromagnetic resonance (FMR). For elastic neutron scattering, I use polarized neutron reflectometry (PNR) and small angle neutron scattering (SANS) to study magnetic structures. The PNR measurements can reveal the magnetic depth profile of the thin film

sample, while the SANS measurements can probe the in-plane magnetic ordering. To support neutron scattering experiments, the VSM and FMR measurements are also applied, where VSM can provide net volume magnetization of the sample and FMR captures the magnetic dynamics. Besides these characterization methods, micromagnetic simulations are also used to help understand the experimental results. These simulations are performed using the object oriented micromagnetic framework (OOMMF) simulation platform.

In Chapter 3, we use polarized neutron reflectometry (PNR) to experimentally probe the interfacial magnetic configurations in the hard/soft bilayer thin films: $L1_0$ -FePt/ $A1$ -FePt, [Co/Pd]/CoPd, [Co/Pt]/FeNi, and $L1_0$ -FePt/Fe, all of which have a perpendicular magnetic anisotropy in the hard layer. These films were designed with different soft and hard layer thicknesses and saturation magnetization. The influences of an in-plane magnetic field and temperature are also studied using a $L1_0$ -FePt/ $A1$ -FePt bilayer sample. In addition, micromagnetic simulations are also applied to understand how these factors influence on magnetic configuration. Key among these results, for thin soft layer thickness, the hard and soft layers are rigidly coupled in the out-of-plane direction and then undergo a transition to relax in-plane. This transition can be delayed to larger soft layer thickness by decreasing the magnetization of the soft layer.

In Chapter 4, we use small angle neutron scattering (SANS) to capture the dynamics in hybrid skyrmions and investigate the spin wave structure. Performing simultaneous ferromagnetic resonance and SANS, the diffraction pattern shows a large increase in low-angle scattering which is present only in the resonance condition. This scattering pattern is best fit using a mass fractal model, which suggests the spin waves form a long-range fractal network. The fractal structure is constructed of fundamental units with a size that encodes the spin wave emissions and are constrained by the skyrmion lattice. These results offer critical insights into the nanoscale dynamics of skyrmions, identify a new dynamic spin wave fractal structure, and present SANS as a unique tool to probe high-speed dynamics.

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Appendix A

Supplemental Materials For Chapter 3

A.1 [Co/Pd]-Multilayers/CoPd Data

PNR measurements are also performed on [Co/Pd]-multilayers/CoPd with $t_{soft} = 2$ nm and 5 nm. The samples are first saturated with 500 mT magnetic field along film normal direction, and then reduce to 0. An in-plane guide field 1 mT was applied while measuring. The reflectivity of R^{++} and R^{--} are largely overlapped with each other for $t_{soft} = 2$ (Figure A.1a and A.1b), which indicates there is not net in plane magnetization. This is then proved by converged magnetic depth profiles that no net MSLD can be probed. This non-in-plane magnetic behavior is also probed in $t_{soft} = 5$ nm (Figure A.1c and A.1d).

A.2 [Co/Pt]-Multilayers/FeNi Data

With the same experimental set up on the above, PNR measurements and the model fitting on [Co/Pt]/FeNi with $t_{soft} = 0, 1$ and 3 nm are shown in Figure A.2 (a, b), (c,d) and (e,f), respectively. For each t_{soft} , R^{++} and R^{--} are largely converged with each other and the corresponding magnetic depth profile presents no MSLD. These results indicate that the magnetization in the soft layer is along the perpendicular anisotropy orientation.

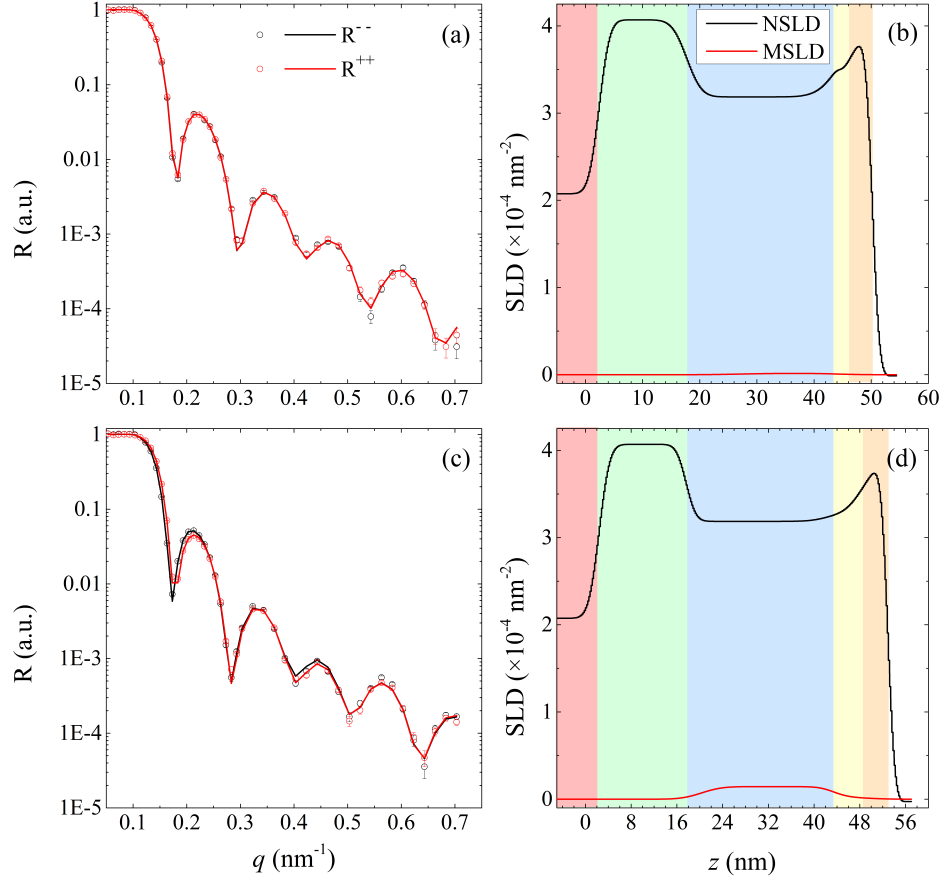


Figure A.1: PNR measurements on [Co/Pd]/CoPd (2 nm) (a) reflectivity (b) converged magnetic depth profile, and same measurements on [Co/Pd]/CoPd (5 nm) (c) reflectivity (d) converged magnetic depth profile, where pink, green, blue and yellow regions are Si, Pd seed layer, [Co/Pd] multilayers, CoPd soft layer and Pd capping layer, respectively.

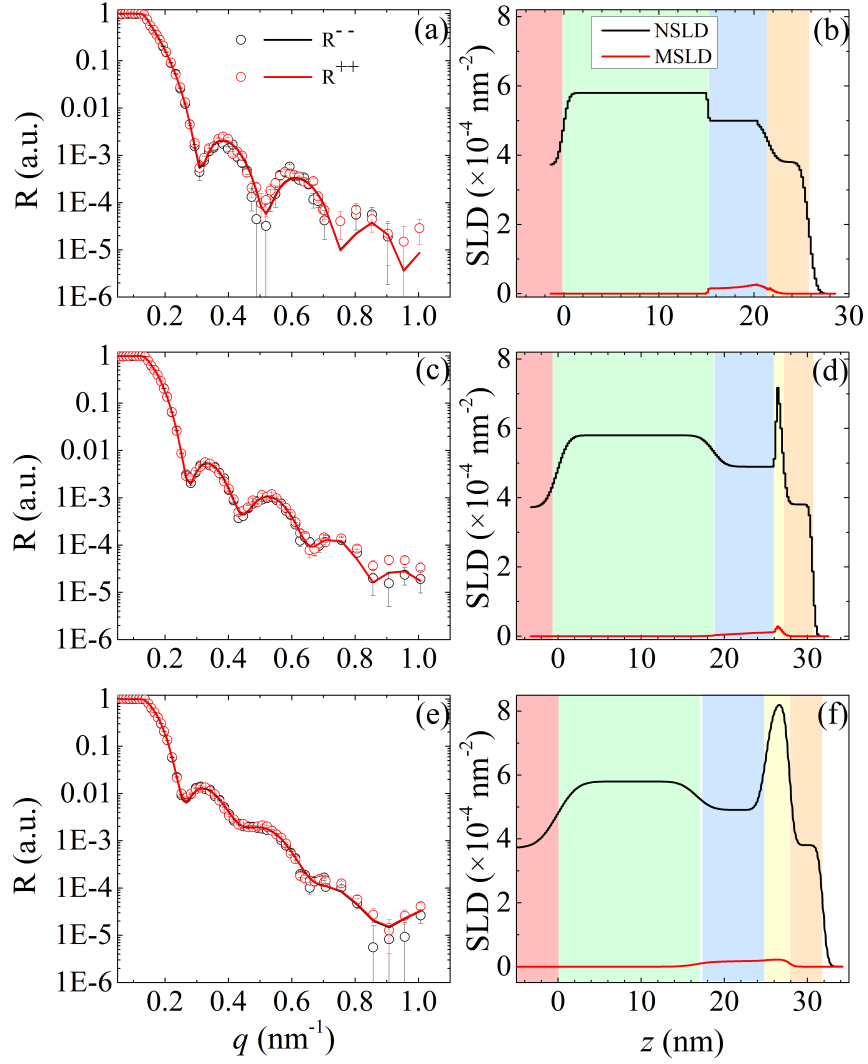


Figure A.2: PNR measurement on [Co/Pt]/FeNi. Reflectivity and correspond magnetic depth profiles: (a) (b) $t_{soft} = 0$, (c) (d) $t_{soft} = 1$ and (e) (f) $t_{soft} = 3$ nm. The magnetic depth profiles: SiO₂ (pink), Pd buffer layer (green), Co/Pt multilayers (blue), FeNi (yellow) and Pd capping layer (orange).

Appendix B

Supplemental Materials For Chapter 4

B.1 Experimental Setup

Figure [B.1](#) shows a schematic of the experimental set up. The ordered skyrmion lattice was prepared following a field sequence described in the main text. The sample was then placed in a two-turn solenoid. The film normal direction is parallel to the neutron beam and static field, while the film in-plane direction is aligned with the solenoid axis. The scattered neutrons are collected by a 2D detector, where the beam stop corresponds to the reciprocal space coordinate axis origin and q_x and q_y are reciprocal space vectors in the plan of the detector. The radii (q) on the detector is a scalar which equals to $\sqrt{(q_x)^2 + (q_y)^2}$, while the azimuthal angle (χ) is the angle in the plane of the detector relative to the positive vertical direction ($+q_y$), measured in the clockwise direction.

B.2 Fit Parameters

Supplementary Tables [B.1](#) and [B.2](#) contain the converged parameters for the plots shown in main text Figures [4.4b](#) and [4.5b](#).

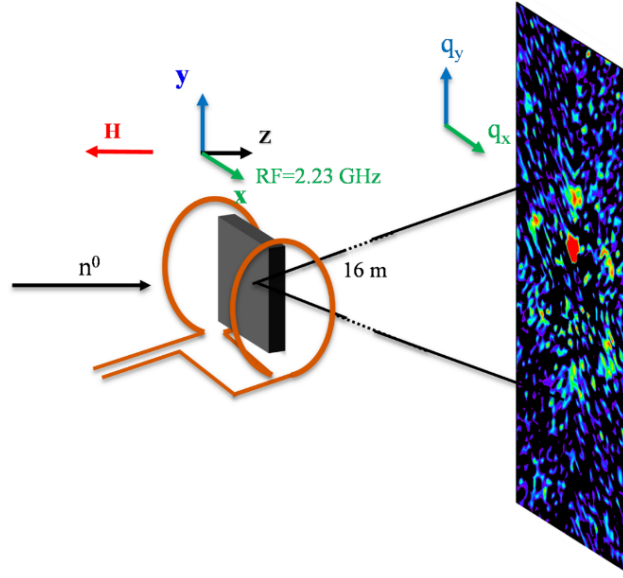


Figure B.1: The schematic of experimental set up. The x , y and z directions demonstrate for Cartesian coordinate system in the real space while q_x and q_y correspond to the SANS detector (reciprocal) space. H , RF , and n^0 here are the applied static magnetic field, in-plane dynamic field and neutron beam directions, respectively.

Table B.1: Converged simulation fitting parameters of (Gaussian + Lorentzian square) model. The chi-G and chi-L correspond to the chi-square of a Gaussian model and a Lorentzian square model, respectively. The d-parameters represent the standard deviation from the fits.

$H(mT)$	$A1$	$\mu (\text{\AA}^{-1})$	σ	$A2$	$\xi (\text{\AA})$	chi-G	chi-L
100	54	0.00291	0.00041			23	
130	44	0.00289	0.00039			19.4	
140	45	0.00284	0.00045			7.6	
150	38	0.00291	0.00036	0.000111	313	5.6	12.6
160	41	0.00281	0.00038	0.000142	294	4.4	13.3
170	40	0.00278	0.00033	0.00015	287	11.7	20.9
180	31	0.00286	0.00031	0.000197	301	15	29.8
190	33	0.00285	0.00039	0.000107	267	3.4	18.5
$dA1$	$d\mu$	$d\sigma$	$dA2$	$d\xi$			
5	0.00004	0.00004					
4	0.00005	0.00004					
3	0.00003	0.00003					
3	0.00003	0.00003	0.000006	15			
2	0.00003	0.00003	0.000005	10			
4	0.00004	0.00004	0.000007	12			
5	0.00006	0.00006	0.000006	8			
2	0.00003	0.00003	0.00001	23			

Table B.2: Converged simulation fitting parameters of (Gaussian + Mass-fractal) model. The chi-G and chi-M correspond to the chi-square of a Gaussian model and a Mass-fractal model, respectively.

H (mT)	AI	μ ($\text{\AA}^{-1}$)	σ	R ($\text{\AA}^{-1}$)	D	ξ ($\text{\AA}$)	a	chi-G	chi-M
100	54	0.00291	0.00041					23	
130	44	0.00289	0.00039					19.4	
140	45	0.00284	0.00045					7.6	
150	41	0.00291	0.00038	157	2.16	828	0.00013	4.5	9.7
160	43	0.00281	0.00042	150	2.43	445	0.00005	3.4	12.2
170	43	0.00279	0.00037	203	2.08	634	0.00036	9.9	16.3
180	42	0.00285	0.00039	246	1.76	1305	0.00349	12.4	13.7
190	35	0.00284	0.00042	100	2.18	500	0.00009	3.6	16.5
dAI	$d\mu$	$d\sigma$	dR	dD	$d\xi$	da			
5	0.00004	0.00004							
4	0.00005	0.00004							
3	0.00003	0.00003							
2	0.00003	0.00002	19	0.08	112	0.00006			
2	0.00002	0.00002	20	0.09	33	0.00003			
4	0.00004	0.00004	10	0.07	56	0.00014			
4	0.00005	0.00005	4	0.34	126	0.00062			
2	0.00003	0.00003	46	0.18	99	0.00009			

B.3 Alternative Models

Form Factor Scattering One alternative model proposes that, under dynamic excitation, the ordering of the hexagonal skyrmion lattice is reduced. As this occurs, the diffraction pattern which is dominated by the structure factor is reduced and becomes reflective of the skyrmion form factor. As this occurs, the intensity of the hexagonally coordinated diffraction peaks should decrease, corresponding to the reduced ordering. Also, the width of the diffraction peak in both q and the azimuthal direction should increase, corresponding to the reduced regularity in the lattice spacing and alignment. This transition would occur abruptly at the resonant condition. In the post-resonance condition the skyrmions may remain disordered or locally re-order; the diffraction patterns would continue to reflect the form factor, or generate a ring, respectively. Following the parameters for the Gaussian fit in the main text Figure 4.4b, these qualities are not observed, suggesting this is not form factor scattering. To further investigate the contribution of form factor scattering, the 2D neutron diffraction pattern for both static and dynamic states was calculated using the simulated structure in the main text Figure 4.3c and 4.3d Donahue (1999) and an in-house python code. Gilbert et al. (2015) The form factors, calculated as the sum of all scattering cross sections, are shown in Figure B.2a and B.2b, respectively. The I vs q plot, Fig. B.2c, shows that both of the calculated patterns have a distinct minima near $q = 0.0068 \text{ \AA}^{-1}$ which should be observable in main text Figure 4.4a and 4.5a, in the measurements taken at resonance. The absence of this minima is consistent with the claim that this is not form-factor scattering.

B.3.1 Diffuse Scattering from Random Spin Waves

Correlation between the intensity of the low- q feature and the resonant gyration suggests spin waves are likely responsible. Following the standard dispersion curve, one would expect the spin waves to possess a continuum of wavevectors (and corresponding wavelengths). The resulting spin wave structure may then be approximated as a random assemblage of magnetic fluctuations. A micromagnetic simulation was performed with this disorder encoded into the magnetic state by programming a random fluctuation into the regions between the skyrmions.

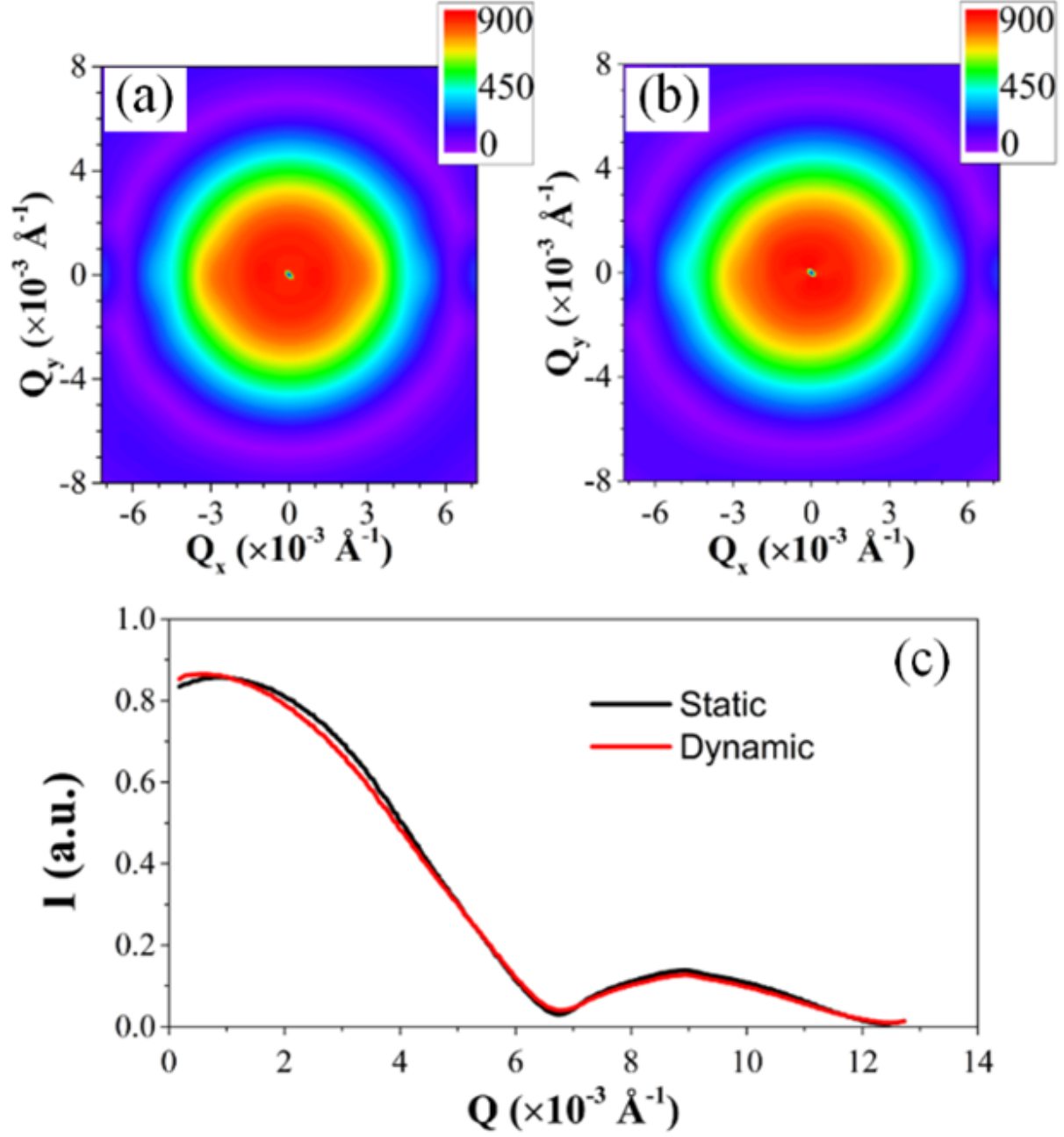


Figure B.2: Calculated form factor from a magnetic skyrmion (a) at-rest and (b) distorted during dynamic gyration. (c) Extracted I vs. q for the form factor.

Figure B.3a and B.3b show the micromagnetic structure of the hybrid skyrmion lattice for static and dynamic states, where the red and blue regions correspond to magnetization along $+z$ and $z-$ direction, respectively. Again, using the in-house python code to calculate the scattering pattern from these real space micromagnetic structures, shown in Figure B.3c and B.3d, respectively. A six-fold diffraction pattern is shown for both states, which is the Fourier transform of the hexagonal skyrmion lattice. In addition, a periodic scattering pattern along q_y axis at $q_x = 0$ was also generated, which is an artifact caused by the finite size of the simulation volume. The I vs q data were extracted from the $2D$ data, excluding the artifact regions by eliminating the data within the q_x range ($0.0015 \text{ \AA}^{-1}$, $0.0015 \text{ \AA}^{-1}$); since the hypothetical spin-waves have no in-plane structure, all of the skyrmion peaks should encode the same information, and excluding the $q_x = 0$ region does not result in reduced insight. The $1D$ data for each state was then modeled by a Gaussian distribution, as shown in Figure B.3e in which blue and red solid lines correspond to static and dynamic states, respectively. This simulation shows that the baseline scattering intensity of the dynamic state is almost same as static state, which indicates the diffuse scattering from local disorder is not significant in the low- q region. Notably, the scattering intensity from the skyrmion lattice is significantly reduced, with the decrease of peak amplitude and width, in the presence of the disordered spin waves. This suggests that a random assemblage of spin waves is likely not responsible for the experimental scattering pattern.

B.3.2 Spin Wave Excitations with a Specific Correlation Length

A Lorentzian-squared function of the form $(\frac{A_2}{(1/\xi)^2 + q^2})^2$ is used to fit this structure, where A_2 is the Lorentzian function amplitude and ξ is the correlation length. This model is the Fourier transform of a spatial correlation function which is proportional to the $\exp(-r/\xi)$, where r is the distance between coherent magnetic structures in real space and ξ is the magnetic correlation length that approximates the size of the structures [Schroeder et al. \(2020\)](#); [Sachan et al. \(2008\)](#). This model has been used previously to describe spin excitations, e.g. spin waves [Grigoriev et al. \(2019\)](#).

Using the combined Gaussian + Lorentzian model, each of the datasets ($\mu_0 H = 100 \text{ mT}$ to 190 mT) were fit shown in Figure B.4a and the resulting parameters are shown in Figure

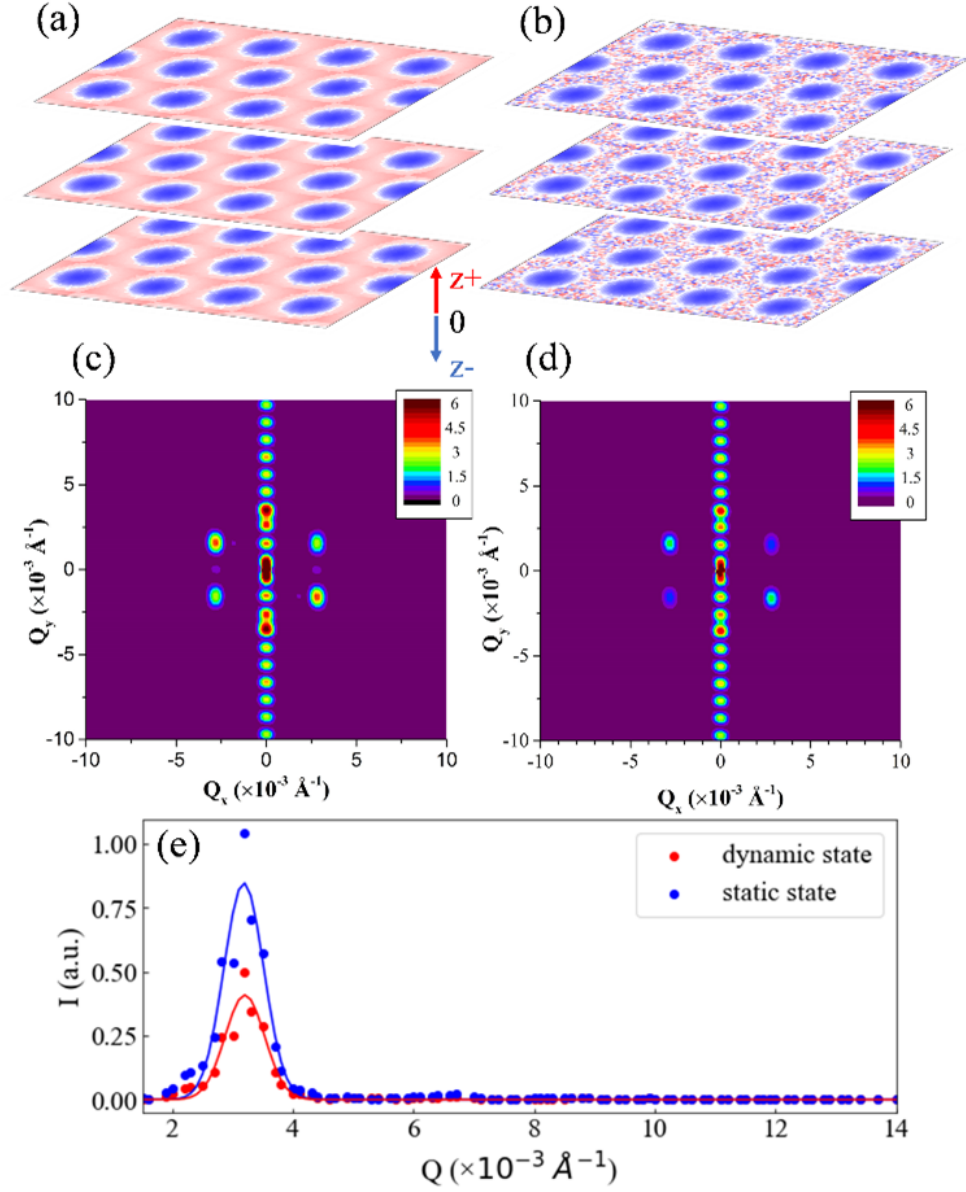


Figure B.3: Micromagnetic simulations of the Fe/Gd hybrid skyrmion lattice in (a) the static state and (b) dynamic state, with modeled spin waves, where the blue and red pixels correspond to magnetization at $z-$ and $z+$ directions, respectively, and the white regions have the magnetization equal to 0 along the z axis. Calculated 2D scattering pattern (SANS) of a hybrid skyrmion lattice in the (c) static state and (d) dynamic state. (e) Calculated I versus q plots from the calculated SANS patterns.

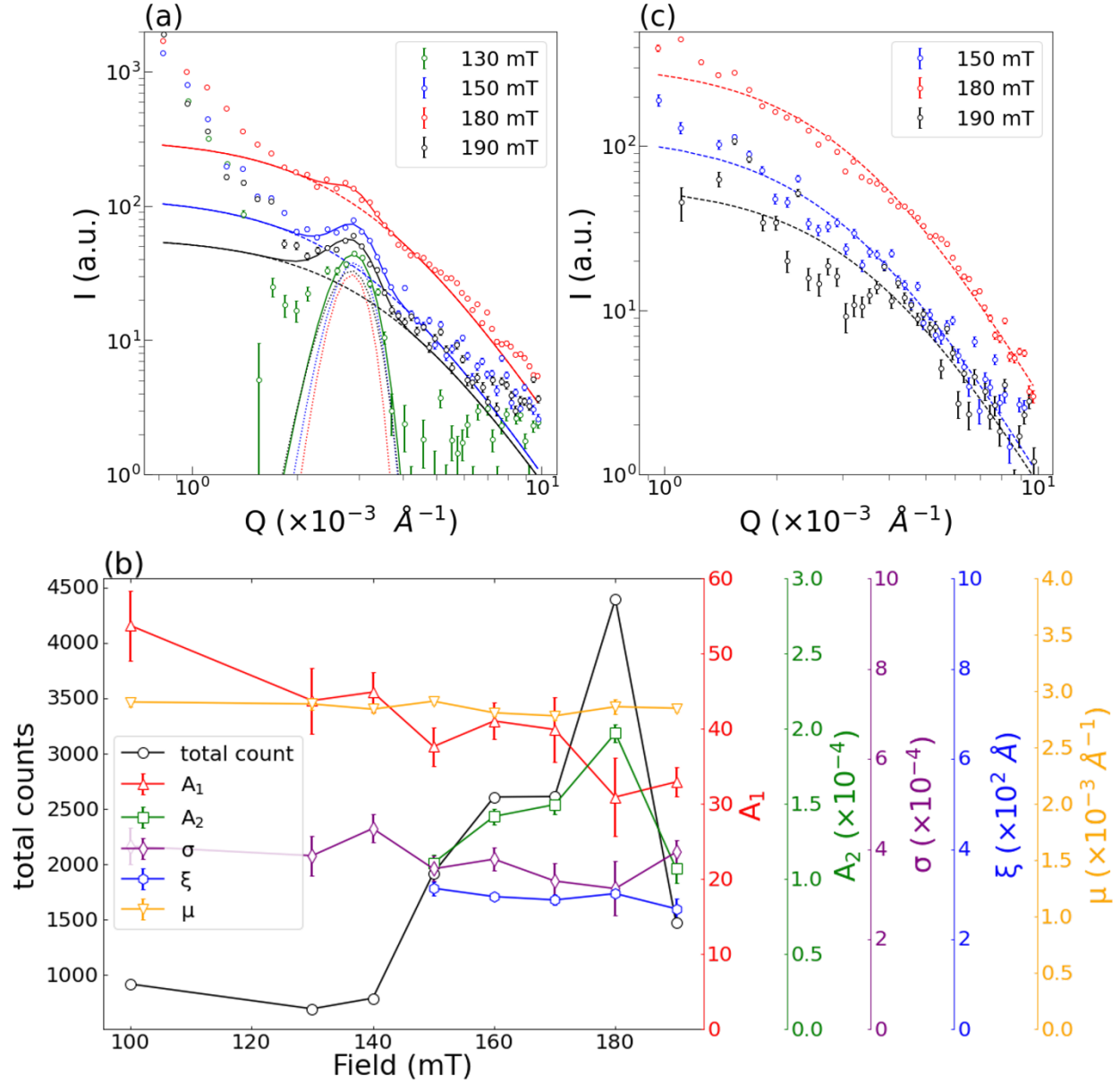


Figure B.4: (a) I vs q plots for the off-resonant (red), pre-resonance (blue), on-resonance (black) and post-resonance (green) states. The data are represented by points and the fitted models are shown as solid lines; the Gaussian component of the model is shown as a dotted line and the Lorentzian squared fit as a dashed line. (b) The total neutrons intensity between $q=(0.0018 \text{ \AA}^{-1}, 0.01 \text{ \AA}^{-1})$ and the corresponding fitting parameters: Gaussian distribution amplitude (A_1), width (w) and central peak position (μ), and the Lorentzian squared function amplitude (A_2) and the correlation length (ξ). (c) I vs q plots for pre-resonance (blue), on-resonance (black) and post-resonance (green) states using the 130 mT data as a dynamic-independent background, then fitted with the Lorentzian squared + Gaussian model.

B.4b. Fitted models, shown as dashed lines in Figure B.4a, capture the diffraction peak from the hexagonally-ordered skyrmion lattice and high- q data ($0.002 \text{ \AA}^{-1} < q$) well, but consistently fall short of the experimental data at low- q . The converged parameters show that the Gaussian intensity continuously decreases with increasing magnetic field, which has been observed previously in static measurements Desautels et al. (2019) and likely indicates that the skyrmions are shrinking in response to the external magnetic field. This dependence does not recover at 190 mT, indicating it is independent of the resonant dynamics. The other Gaussian parameters, σ and μ , change relatively little over the measured range, with $\mu = 0.00285 \text{ \AA}^{-1} \pm 0.00005 \text{ \AA}^{-1}$ corresponding to a lattice constant $\approx 2200 \text{ \AA}$. This suggests that the quality of the skyrmion lattice ordering and lattice spacing remains approximately constant during the resonant gyration.

Considering the Lorentzian-squared fit, the amplitude, A_2 is approximately zero for $\mu_0 H < 150 \text{ mT}$, then increases to a maximum at 180 mT before reducing again at 190 mT. This suggests that the low- q feature is coupled to the resonant dynamics. The magnetic correlation length as determined by these fits remain approximately constant at $\xi \approx 300 \text{ \AA}$. This magnetic correlation can be attributed to spin waves within the material. As will be discussed below, previously published theoretical work predicts that the spin wave distribution is maximal at approximately the radius of the magnetic skyrmions. Finally, in addition to the fits, the total integrated scattering intensity at $q > 0.0018 \text{ \AA}^{-1}$ is also shown in Figure B.4b and increases by more than 400% at $\mu_0 H = 180 \text{ mT}$, approximately corresponding to the resonance condition. This intensity drops rapidly at fields larger or smaller than 180 mT presumably since the resonant condition is no longer satisfied.

Since the magnetic correlation length and skyrmion lattice diffraction peak is approximately constant, the datasets can be subtracted from each other and can still be fit with the Lorentzian-squared model. This subtraction removes magnetic features which are independent of the resonance. Using the dataset measured at 130 mT as a new magnetic background, this subtraction was performed and I vs q data calculated, as shown in Figure B.4c. As expected, the diffraction peak is strongly suppressed, confirming that the ordered skyrmion lattice changes very little during resonance. Next, the data are fit with the same Lorentzian-squared function described above, achieving an improved fit, however, still fails

to fit the ultra-low q region. Therefore, the spin waves ejected during the dynamic resonance may not completely explain the extra scattering phenomena probed in SANS pattern.

B.4 NIST Data

Dynamic SANS measurements were also performed on Ta(50 Å)/[Fe(3.4 Å)/Gd(4.1 Å)]₁₂₀/Ta(50 Å) thin films at NIST on the vSANS instrument. Ten thin film samples were stacked in the sample holder and then prepared in the ordered skyrmion lattice state. The 2D SANS diffraction pattern measured without a static or excitation field, are shown in Figure B.5(a). The SANS pattern displays the characteristic six-fold structure of an ordered skyrmion lattice. Using the radius of the six-fold pattern ($q=0.003 \text{ Å}^{-1}$), the skyrmion center-to-center spacing is calculated to be 210 nm. These values are consistent with our previous work and the measurements at GPSANS. These results have no complementary FMR results and so are presented as a stand-alone dataset in the supplement. Next, a magnetic field of $\mu_0 H = 150 \text{ mT}$ was applied along the film normal direction, which is also parallel to the neutron flux, and the frequency generator was turned on, resulting in an in-plane excitation field. Increasing the in-plane excitation frequency, f , from 1.2 GHz to 1.4 GHz is shown in Figure B.5(b-d); note that all three figures are plotted on the same intensity scale. At a $f = 1.2 \text{ GHz}$, the pattern has not changed significantly from the zero-field measurement, continuing to show the same six-fold pattern as the static case. Increasing the f to 1.3 GHz, the six-fold pattern is still easily identified, but the baseline, e.g. apparent background, increases significantly across the entire spectrum. Further increasing f to 1.4 GHz, the signal returns to the six-fold pattern with a low background. These frequencies were chosen based off of FMR measurements performed on similar samples, however there were no FMR measurements performed on these precise samples or the resonant coil. As a result, it is only proposed that the SANS response identifies resonance, and that the diffraction patterns of resonant and the off-resonant states were distinctively different. For this reason, these results are included in the supplement.

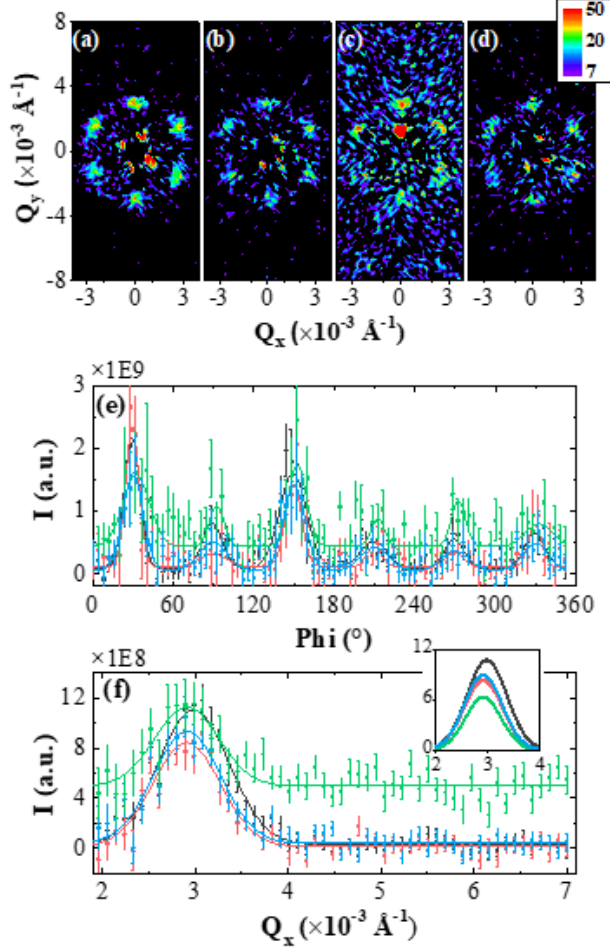


Figure B.5: 2D SANS pattern on vSANS of magnetic skyrmion lattice under different magnetic fields and excitation frequencies: (a) Static state ($H = 0$, $f = 0$), (b) below-resonance state ($\mu_0 H = 150$ mT, $f = 1.2$ GHz), (c) on-resonance state ($\mu_0 H = 150$ mT, $f = 1.3$ GHz) and (d) above-resonance state ($\mu_0 H = 150$ mT, $f = 1.4$ GHz). All the images are plotted in the same intensity scale. (e) Integrated I vs ϕ projection calculated at ($q = 0.0029 \text{ \AA}^{-1}$, $\Delta q = 0.0005 \text{ \AA}^{-1}$) and (f) I vs q , for the static (black), below-resonance (red), on-resonance (green) and post-resonance (blue). Inset in (f) shows the diffraction peak with the constant baseline (background) intensity subtracted.

One-dimensional projections of the data can be prepared by integrating all radii within a range at fixed azimuthal angles, generating a plot of intensity versus ϕ . Figure B.5e, shows the six peaks from the 2D plot, separated by 60° , respectively. In each of the four states the six peaks retain their spacing, indicating that the skyrmion lattice remains largely intact under dynamic excitation. The most notable feature is that the peaks are uniformly offset from the baseline for the on-resonance case ($f = 1.3$ GHz), meaning that the excitation has no in-plane symmetry breaking. Figure B.5f shows the I vs q plot, where a single peak at $q = 0.00298 \text{ \AA}^{-1}$ is seen for the static case and $q = 0.00290 \text{ \AA}^{-1}$ for the measurements with a static magnetic field. This shift in the lattice size was observed in our previous work and is an effect of the static magnetic field indicating an expansion of the lattice approaching positive saturation. The lattice constant at $\mu_0 H = 150$ mT can be calculated to be 216 nm, compared to 210 nm at remanence. As observed in the 2D data, the baseline is offset when the resonance condition is met, but by comparison to the main text this offset extends over a much longer q -range.

Measurements were also performed by fixing the excitation frequency at 1.3 GHz, 1.5 GHz, and 1.7 GHz and sweeping the applied magnetic field between 110 mT and 190 mT. Applying different frequencies deviates strongly from the resonant condition of the two-turn solenoid, introducing additional factors to the energy transfer and corresponding dynamics. It has been suggested that this has the appearance of detector noise; one could consider the resonant antenna broadcasting RF signals that inadvertently trigger the sensitive SANS detectors, for example. However, measurements this second set of measurements confirms this is not the case. Since the field, which is generated in a yoked magnet approximately >10 m from the detector, it is not expected to affect the detector electronics. Another consideration might be that we are observing background scattering from other nearby instruments. However, these measurements were not taken sequentially – meaning we measured with increasing frequency through resonance, then decreased the frequency and re-measured in-between values, capturing resonance again. It would be highly improbable that, at each instance that the field/frequency condition is met on the vSANS beamline, that an adjacent instrument would engage this strong scattering. Since the typical measurement for these experiments takes 45 minutes per state, this background would need to be active for

prolonged periods of time, again coincident with the field and frequency conditions. These considerations support that the signal is authentically from the sample

The I vs q data from the field-dependent measurements was fitted using a Gaussian function with an additional constant baseline intensity: $A \times \exp(-\frac{(x-\mu)^2}{2w^2}) + Y_0$, where A , w , μ and Y_0 are peak amplitude, width, and mean of the Gaussian and a flat baseline intensity, respectively. The parameter Y_0 is shown in Figure B.6, and shows a constant, low value until a precise field/frequency condition is met. This condition is in agreement with the frequency dependent measurements shown in Figure B.5 Parameters A and w from are shown in Figure B.7, but had only a weak dependence on the applied static field, similar to the observations in the main text.

These vSANS results, together with the GPSANS results, reveal the dynamic mode of hybrid skyrmion. However, there are two main differences between these measurements: (1) the frequency of the excitation field and (2) the background intensity. Differences in the apparent resonance are expected to be a consequence of inter-sample variation. Difference on the order of a fraction of an angstrom strongly effect the magnetic structure in these systems, as shown previously. Differences in how the signal manifests in the vSANS and GPSANS data are also distinctly different, with the former being q -independent and the latter being q -dependent. The q -dependent results (GPSANS data) have already been discussed in the main text. A globally q -independent scattering pattern is unrealistic, suggesting that the measured data perhaps were limited in q and did not capture the decay function. To support this point, the middle front panels in the vSANS instrument, which were not configured during the measurements but did record high- q data all the same, did not show any enhanced scattering on resonance.

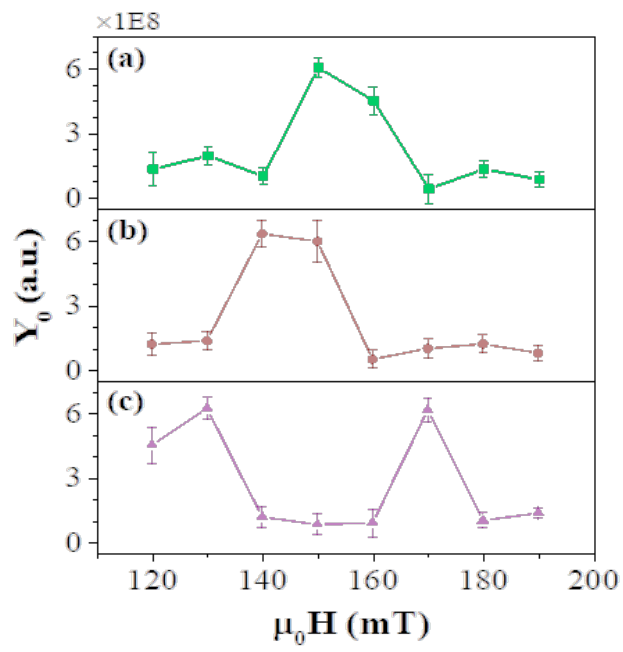


Figure B.6: The baseline intensity Y_0 as measured on vSANS for (a) 1.3 GHz, (b) 1.5 GHz and (c) 1.7 GHz as a function of magnetic field.

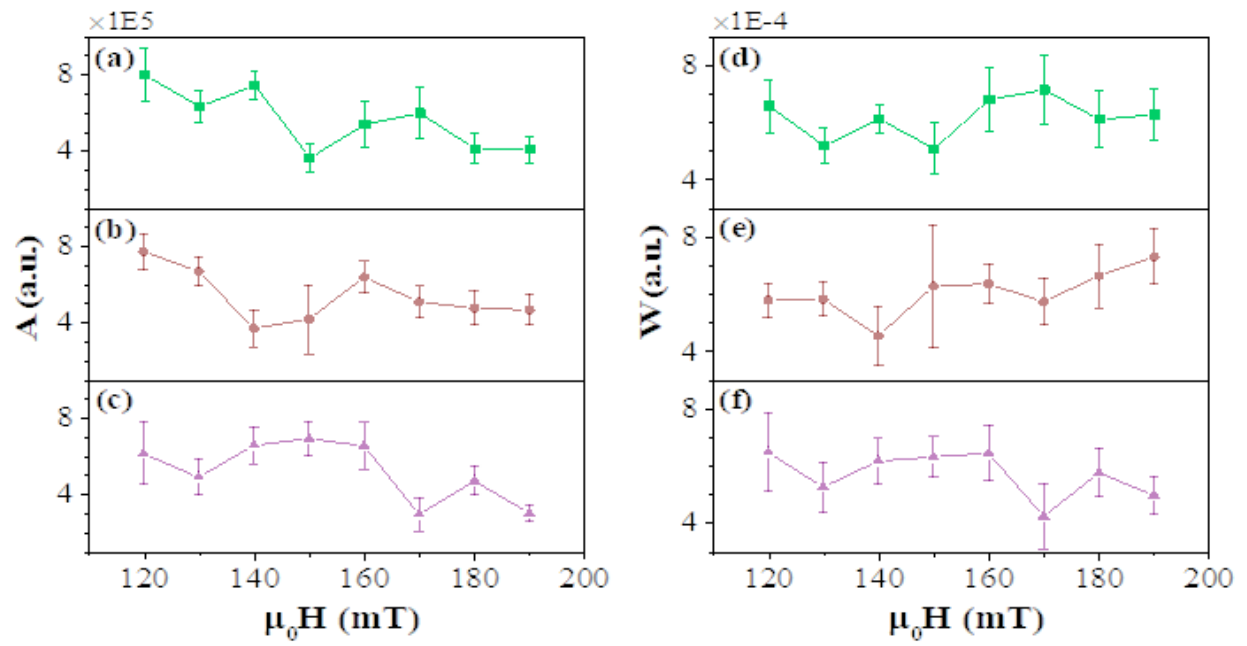


Figure B.7: Fitted parameters for the I vs magnetic field H vSANS data, showing (a-c) peak amplitude (A) and (d-f) peak width (w), for (top) 1.3 GHz, (middle) 1.5 GHz and (bottom) 1.7 GHz.

Vita

Nan Tang was born on October 28th, 1991, in Heilongjiang, China. He graduated from Sichuan University and received a B.S. in Material Chemistry in 2014. In the September of the same year, he started graduate school in Sichuan University and received a Master in Material Engineering in 2017. In 2018, He came to U.S. to pursue a Ph.D in Material Science Engineering at the University of Tennessee, Knoxville. He has worked as a research assistant and a teaching assistant during the five years in the group of Dr. Dustin Gilbert. And he is expected to receive his Ph.D in May of 2023.