Interface and Topology in Magnetism

by

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Committee in charge:

Professor Zi Q Qiu, Chair
Professor Frances Hellman
Professor Jeffrey Bokor

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Abstract

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This dissertation explores interface interactions, topology of spin structures, and the use of interface interactions in manipulating the topology of spin structures. First, I will present our work on interface interactions in magnetic thin film heterostructures. A majority of interesting phenomena in magnetic heterostructures are caused primarily by the interface interactions. In exchange bias system CoO/MgO/Fe/Ag(001), the role of frozen and rotatable antiferromagnetic spins in exchange bias system was investigated using X-ray Magnetic Linear Dichroism (XMLD). In the Py/FeMn/Ni/Cu(001) system, the indirect coupling through an antiferromagnetic spacer layer was studied through anisotropy measurements using Rotating-field Magneto-Optical Kerr Effect (ROTMOKE).

Second, I will present our work on artificial topological spin structures. Traditionally, research on nanostructured magnetic materials has focused on the study of uniformly magnetized thin films. With the increasing capability to prepare and manipulate non-uniform spin configurations in recent years, various topological spin structure have been realized. In magnetic thin films, magnetic vortex states can be stabilized by reducing the lateral dimension of the magnetic thin film to the nanoscale due to the increased shape anisotropy. By combining this shape anisotropy and interlayer exchange coupling at the interface, we showed that more complex topological spin structures such as spin skyrmions and hedgehog spin merons can be stabilized and manipulated. By utilizing uniaxial strain in a ferroelectric-ferromagnetic system Co/Cu/PMN-PT(011), we demonstrated a method to manipulate the circulation of a magnetic vortex state using strain.
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Chapter 1

Introduction

The broader theme of this dissertation is on spintronics, where we study and explore the use of electron spin as another degree of freedom (in addition to electron charge) in designing future electronic devices. Spin is a fundamental property of the electron which carries angular momentum and is largely responsible for magnetism in solids. Research on spintronics and magnetism are primarily concerned with the manipulation, storage, and transfer of information using electron spin. My research can be said to be focused on the manipulation aspect of spintronics research. In particular, I researched the different ways to manipulate spin states of various magnetic thin film heterostructures through interface interactions and more recently to use this interface interactions to manipulate the topology of a spin texture in magnetic nanodisks.

![Figure 1.1: Spin is a fundamental property of an electron](image)

In the past few decades, magnetic thin film heterostructures with various structures have been studied extensively, with stacks of different ferromagnetic layers (FM), various non-magnetic spacer materials, as well as antiferromagnetic layers (AFM) showing many interesting phenomena. The most important invention among them is the Giant Magnetoresistance (GMR) effect. Fig 1.2 shows a sketch of a GMR device. The device is composed of two ferromagnetic layers separated by a non-magnetic conductive layer. The working principle of this device is based on the Anisotropic Magnetoresistance (AMR), where the resistance across the trilayer stack is lower when the two ferromagnetic layers are aligned.
compared to when the two layers are anti-aligned. If one of the ferromagnetic layer is pinned, and the other is free, the GMR device forms a very effective sensor of magnetic field. The use of a GMR sensor (or variations of it) in the read head of magnetic recording media has fueled the explosive growth in our ability to store more and more data into a small area, with the recent technology reaching 1 Tb/in² in 2016. More recently, the GMR device has been used as a memory cell in the design of a Magnetic Random Access Memory (MRAM), which has the potential of becoming a ‘universal memory’ since it offers the access speed of traditional RAM with non-volatility of magnetic storage devices. The discovery of GMR in 1988 by Peter Grünberg and Albert Fert is so important that it has been considered by many to be the birth of the field of spintronics and also as a harbinger of the information age. For their work on GMR, Peter Grünberg and Albert Fert shared the 2007 Nobel prize in physics.

![Figure 1.2: When the two ferromagnets in the FM/non-magnetic conductor/FM trilayer is aligned, the resistance across the device is lower than when the two ferromagnets are anti-aligned](image)

**1.1 Magnetic Storage Industry Roadmap**

The major driving force of nanostructured magnetic materials research and the field of spintronics in general is the demand of creative new technologies from the magnetic storage industry. In today’s state of the art hard-disk drives (early 2016), information is stored in weakly exchange coupled magnetic grains (about 8 nm in diameter) with large anisotropy. A single bit of data is usually stored in around 20-30 of such grains that are magnetized in the same direction, which is either out-of-plane or into the plane in a perpendicular magnetic recording media. Higher density can be achieved by using smaller grain size. However, as the size of the grain reach into the nanoscale, magnetization can randomly flip direction under the influence of temperature, a phenomenon known as superparamagnetism. This superparamagnetism represents a hard limit to the ability of magnetic grains to store information. The mean time between two flips is called the Néel relaxation time $\tau_N$ and is
given by the Néel-Arrhenius equation:

$$\tau_N = \tau_0 \exp\left(\frac{KV}{k_BT}\right)$$

(1.1)

where $\tau_0$ is the intrinsic time which depends on the material parameters (magnetostriction constant, Young modulus, etc), with typical values of $\tau_0 \sim 10^{-10}$ to $10^{-9}$ s. $K$ is the magnetic anisotropy of the grain and $V$ its volume. $k_B$ is the Boltzmann constant and $T$ is the temperature of the magnetic material. For the magnetic grains to be able to store data reliably (to be used for long-term data storage), the Néel relaxation time need to be of the order of $> 10$ years, which means the ratio $\frac{KV}{k_BT}$ needs to be greater than about 60 at room temperature. If the volume of the magnetic grain is reduced, it requires the magnetic anisotropy to be increased in order to keep the ratio greater than 60. This gives rise to a challenge in the ability of the write system to produce enough magnetic field to overcome the coercivity and to flip the magnetic bits. The Heat-Assisted Magnetic recording (HAMR) are being developed in order to overcome this problem. In a HAMR media, heat is applied using a small laser to temporarily reduce the coercivity while writing takes place in part of the disk. This method allows writing on a smaller scale than before, increasing the amount of data that can be held on a standard disk platter. The latest report on HAMR recording demonstrated an areal density of 1.402 Tb/in$^2$ using a plasmonic near field transducer and high anisotropy granular FePt media \[1\]. It was reported that a HAMR drive can push the areal storage density to 5 Tb/in$^2$. Beyond that, according to the latest Advanced Storage Technology Consortium (ASTC) roadmap (shown in figure \[1.3\]), there is a consensus in the industry that, in order to continue packing more and more data into a small area, some form of bit patterned media (BPM) with pre-patterned magnetic grains combined with heat-assisted technology are necessary in the future to continue working around the fast-approaching superparamagnetic limit in conventional media. Thus understanding the behavior of ferromagnetic materials in the patterned media, especially when the lateral dimension of the ferromagnetic thin film reaches the nanoscale, will be important for future technological applications.

### 1.2 Interface and Topology

This dissertation is structured as follows: I will first give an overview of the theoretical and experimental aspects of the works done in this dissertation. In chapter \[2\] I will outline the basic physics involved in magnetism. I will start with a review of several simple models of magnetism to highlight the quantum-mechanical origin of ferromagnetism. I will then segue into the phenomenological models that are more commonly used to describe the behavior of ferromagnetic thin films heterostructures and the treatment of interfaces in those models. I will then move from the traditionally single-spin treatment of ferromagnetism in thin films, and look at non-uniform spin textures. In chapter \[3\] I will describe the experimental techniques that are used throughout this dissertation. This will include the sample preparation
CHAPTER 1. INTRODUCTION

Figure 1.3: Left: Technology roadmap from the Advanced Storage Technology Consortium (ASTC). Right: The move from conventional media (many random grains per bit) to some form of patterned media (single pre-patterned grain per bit) is necessary in order to continue working around the superparamagnetic limit. Figure adapted from [2].

method using Molecular Beam Epitaxy (MBE) in an Ultrahigh Vacuum (UHV) chamber, magneto-optical measurements, as well as X-ray spectroscopy and microscopy technique.

Next I will present several research topics concerning magnetism at the interfaces of heterostructures and topology of spin textures. Interfaces between different magnetic layers in magnetic thin film heterostructures play an important role in the processes of spin-dependent electron transport phenomenon. One such phenomena is the exchange bias in a FM/AFM bilayer systems which has enabled the commercialization of the GMR based sensor. The exchange interaction at the interface between the FM and AFM layer in such a bilayer system is responsible for pinning the the interfacial spins of the softer ferromagnetic layer, which added an energy term that leads to the shift in the switching field known as exchange bias. The direction of this exchange bias can be set by cooling through the Néel temperature ($T_N$) of the AFM layer within an applied field, while the ferromagnetic layer stays below its Curie temperature ($T_C$) at all times (i.e. $T_C > T_N$). In chapter 4 I present an investigation of exchange bias in a FM/insulating spacer/AFM system. By changing the insulating spacer thickness, the coupling between the FM and AFM layer can be tuned. We discovered that the details of the spin structure in the AFM layer (i.e. whether AFM spins are frozen or rotatable) plays a role in the exchange bias of this trilayer system. In chapter 5 we demonstrated that the antiferromagnetic spin structure can be modified by an adjacent ferromagnetic layer. In particular, an adjacent ferromagnetic layer with in-plane magnetization will enhance the in-plane component of the FeMn 3Q antiferromagnetic spin structure, while an adjacent ferromagnetic layer with out-of-plane magnetization does not enhance or even reduce this in-plane component.

Next, I present our research on topological phenomenon that may arise in non-uniform
spin texture. We prepared patterned ferromagnetic thin films by evaporation through a shadow mask (also known as stencil lithography). This method allows us to study patterned ferromagnetic samples while preserving the quality of the interfaces in our magnetic heterostructures since the whole sample structure can be grown without the sample ever leaving the ultrahigh vacuum environment. In chapter 6, we demonstrated a topological effects on artificial magnetic skyrmions. By imprinting magnetic vortices on a ferromagnetic film with out-of-plane anisotropy, we engineered spin textures with skyrmion numbers $N = +1$ and $N = 0$. We showed that if the process of changing from multi-domain state to single-domain state involves a change in skyrmion number (i.e. changing the topology), it will take additional energy to do so. In chapter 7, we showed that the hedgehog-type spin merons (half-skyrmions) can be stabilized in an asymmetric trilayer Ni/Fe/Co on Cu(001). Furthermore, the helicity of the spin merons can be tuned in the whole $0 - 2\pi$ range by fine-tuning Fe thickness with sub-monolayer precision. In chapter 8, we investigated the influence of strain on a magnetic vortex state in an artificial multiferroic structure FM film/FE substrate. By inserting a metallic spacer layer between the ferromagnetic film and the ferroelectric substrate, the magnitude of strain that is transferred to the ferromagnetic film from the ferroelectric response under an applied electric field can be tuned. We demonstrated switching of the magnetic vortex state circulation when the right magnitude of strain pulses are applied to the magnetic vortex.
Chapter 2

Basic Theory of Ferromagnetism

The Bohr - van Leeuwen theorem states that when statistical mechanics and classical mechanics are applied consistently, the thermal average of the magnetization is always zero. This hints at the fact that magnetism in solids must have a quantum mechanical origin and that classical physics cannot account for the origin of diamagnetism, paramagnetism, or ferromagnetism[3].

In this chapter I will first explore the simplest theory of magnetism in the context of Maxwell’s equation (section 2.1) and show that classical electromagnetism cannot explain ferromagnetic ordering in solids. In section 2.2 I will discuss the quantum mechanical origin of ferromagnetic ordering in some detail. Afterwards, I will move away from the \textit{ab initio} treatment of ferromagnetism, and discuss phenomenological models relevant to ferromagnetic thin films In section 2.3 I will mostly describe micromagnetic models where only uniformly magnetized films are considered (the macrospin limit). Finally in section 2.4 I will introduce the concept of topology and how it can be relevant in magnetism when non-uniform spin textures are considered.

2.1 Simple Theories of Magnetism

A magnetized body is characterized by the dipole moment $\mathbf{m} = M \mathbf{V}$ where $\mathbf{M}$ is the average magnetization (magnetization per unit volume) and $V$ is the volume of the magnet. Classically the dipole moment is probed by putting the magnet into an external field $\mathbf{H}$ which leads to an interaction energy of the form:

$$E = -\mu_0 \mathbf{m} \cdot \mathbf{H}$$

(2.1)

Here $\mu_0 = 4\pi \times 10^{-7} \text{ JA}^{-2} \text{m}^{-1}$ is the magnetic permeability. Maxwell’s equation yields $\mathbf{H}$ and the related flux density $\mathbf{B} = \mu_0(\mathbf{H} + \mathbf{M})$ from the magnetization. The magnetization is related to the field by the magnetic susceptibility as $M = \chi H$, or the more precise definition $\chi = dM/dH$ measured at $H = 0$. This relation however does not explain the origin of magnetism in solids. In general $\chi$ is not a constant and may depend on magnetization
For example, at the phase transition point from paramagnetic to ferromagnetic ordering (at Curie temperature $T_C$), the susceptibility diverges.

### 2.1.1 Origin of Magnetic Moment

If we are to consider the atomic origin, then magnetism in solids nearly exclusively originates from the electrons. Saturating the magnetization in this context means that all available magnetic moments are aligned parallel to the magnetic field. However not all electrons in a solid contributes to the magnetization. For example, $^{26}\text{Fe}$ has 26 electrons but it only has 2.15 T room temperature magnetization (Each electron contributes one Bohr magneton moment $\mu_B = 9.274 \times 10^{-24} \text{J/T}$, leading to a magnetization of 1 T per Bohr magneton). This small fraction of contributing electrons is caused by electron pairing, since the electron’s magnetic moment is cancelled by the opposite moment of the second electron in the pair. Thus, only atoms with partially filled shells (with unpaired spins) can have a net magnetic moment. Furthermore, because of Hund’s rule\[4\], the first few electrons in a shell tend to have the same spin, thereby increasing the total dipole moment. Using iron as an example, the electron configuration of $^{26}\text{Fe}$ is $[\text{Ar}]3d^64s^2$, with 0 to 4 unpaired spins depending on the details of its crystal field. The magnetic moment of a bulk Fe is measured to be 2.2$\mu_B$ which is in good agreement. However, the fact that magnetic moments of pure elements are not usually integer multiples of $\mu_B$ (Cobalt: 1.7$\mu_B$, Nickel: 0.6$\mu_B$, Gd: 6.8$\mu_B$, etc) suggests that this is not the whole story. Orbital moment, which is quenched by the crystal field, also gives a small contribution to the total magnetic moment. In fact, to calculate the value of magnetic moment in itinerant magnets, we need to consider the band structure of the solid (for example, using Density Functional Theory (DFT) with spin-dependent pseudopotentials, or other approximations to the band theory of solids).

### 2.1.2 Paramagnetic Spins

Thus far we have discussed the origin of magnetization in solids, however the bigger question to ask is what causes spontaneous ordering of these magnetic moments in a ferromagnet? Let’s first consider the ‘paramagnetic gas’ model with non-interacting moments. Each atom carries one spin with zero orbital moment. For a spin $\sigma$ and a field in the z-direction $\mathbf{H} = H\hat{z}$, the Hamiltonian is given by:

$$\hat{\mathcal{H}} = -\mu_0\mu_B \mathbf{H} \cdot \mathbf{\sigma}$$  \hspace{1cm} (2.2)

where $\sigma$ is the vector Pauli matrix $\mathbf{\sigma} = \sigma_x\hat{x} + \sigma_y\hat{y} + \sigma_z\hat{z}$:

$$\sigma_x = \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}, \sigma_y = \begin{bmatrix} 0 & -i \\ i & 0 \end{bmatrix}, \sigma_z = \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix}$$  \hspace{1cm} (2.3)

The energy eigenvalues of this Hamiltonian is $E_{\pm} = \pm \mu_0\mu_B H$, which is also known as Zeeman splitting. The Zeeman energy favors spin alignment parallel to the external magnetic field.
Given these energy eigenvalues, the probability of realizing the $\uparrow$ and $\downarrow$ states is given by:

$$P(\pm) = \frac{1}{Z} \exp(\pm \frac{\mu_0 \mu_B H}{k_B T})$$

(2.4)

where $Z = \sum_j \exp(-\frac{E_j}{k_B T}) = \exp(\frac{\mu_0 \mu_B H}{k_B T}) + \exp(-\frac{\mu_0 \mu_B H}{k_B T})$ is the partition function. The average moment at a temperature $T$ is then:

$$\langle m \rangle = P_+(\mu_B) + P_-(\mu_B)$$

$$= \frac{\mu_B}{Z} (\exp(\frac{\mu_0 \mu_B H}{k_B T}) + \exp(-\frac{\mu_0 \mu_B H}{k_B T}))$$

(2.5)

$$\langle m \rangle = \mu_B \tanh(\frac{\mu_0 \mu_B H}{k_B T})$$

Equation 2.5 shows that an external magnetic field creates a temperature dependent spin polarization. At $T = 0$, $\langle m \rangle = \pm \mu_B$ for $H_z = \pm H$ we have full spin polarization. On the other limit at $T \gg 0$ we can approximate $\tanh x \approx x$ for small arguments and obtain $\langle m \rangle = \chi H$ where $\chi = \frac{\mu_0 \mu_B^2}{k_B T V}$ is called the Curie susceptibility of the system. $V$ is the volume per spin, which makes $\chi$ a dimensionless number.

Experimentally, the Curie temperature of elemental iron (that is the temperature at which spontaneous ordering of the ferromagnetic order parameter occurs) is about 1043 K. In contrast, the average moment per spin within the paramagnetic gas model (assuming a typical laboratory scale magnetic field of 1 Tesla) is strongly reduced above the temperature of about 1 K. Furthermore, in some materials the phase transition is marked by a sharp transition at the Curie temperature, while this model predicts a smooth temperature dependence. Thus the paramagnetic gas model cannot explain ferromagnetism. This can actually be seen more directly from the smallness of the magnetic interaction energy. The temperature equivalent of the Bohr magneton $\mu_B/k_B = 0.672 K/T$ means that spin alignment due to external fields is very effectively overcome by thermal excitations. The same is true for magnetostatic interaction field. What, then, is responsible for ferromagnetism in solids?

### 2.1.3 Curie-Weiss Law

Interaction between spins of electrons is largely responsible for ferromagnetism in solid. Spin-spin interaction is an exclusively quantum-mechanical effect with no classical analogue. Phenomenologically, a self-consistent approach can be formulated by substituting the spin-spin interaction with a built-in magnetic field known as the Weiss field which tend to line up the magnetic moments. This is also known as the mean-field theory of ferromagnetism. The total magnetic field experienced by each magnetic moment is given by $H + \lambda M$, where $\lambda$ is the Weiss molecular field constant. Substituting the field $H$ in equation 2.5 with the internal Weiss field $H = \lambda \langle m \rangle$, we obtain a self-consistent equation for zero external field applied:

$$\frac{\langle m \rangle}{\mu_B} = \tanh(\frac{\mu_0 \mu_B^2}{k_B T} \lambda \langle m \rangle)$$

(2.6)
It’s difficult to obtain an analytical solution to this self-consistent equation, but we can use a graphical means by plotting the left and right hand side of this equation on the same plot. Figure 2.1 shows the plot of the left and right hand side of equation 2.6 for two different temperature values. The positions where the plot of \( \tanh(\frac{\mu_0 \mu^2 B}{k_B T} \lambda \langle m \rangle) \) intersects the \( \langle m \rangle \) plot (blue line) are the solutions to the self-consistent equation. From figure 2.1, we observe that when the temperature is higher than some transition temperature (known as Curie-Weiss temperature) \( T > T_C \), there’s only the trivial solution \( \langle m \rangle = 0 \). When the temperature is lower than the transition temperature \( T < T_C \), the two plots intersects at three positions: a trivial solution at \( \langle m \rangle = 0 \), and two spin-polarized states \( \langle m \rangle \approx \pm 1 \). These two non-trivial, spin-polarized solutions are the distinguishing characteristic of a ferromagnetic ordering, where the spontaneous symmetry breaking of the spin configuration occur even at zero external field. The Curie susceptibility can also be modified within the Weiss mean field theory, leading to \( \chi = \frac{\mu_0 \mu^2}{k_B (T-T_C)^V} \).

Figure 2.1: Plot of the left and right hand side of the self consistent equation 2.6

While the Weiss theory of ferromagnetism can successfully describe ferromagnetism phenomenologically, we do not have a microscopic picture of the origin of this molecular field constant \( \lambda \). In fact the required internal field \( \lambda M \) is so large that it’s physically unlikely to occur. In the next section we are going to look into the quantum mechanical picture of spin-spin interaction.
2.2 Models of Exchange

The only force that can overcome thermal excitation and establish ferromagnetic order is none other than the Coulomb force. The Coulomb interaction establishes an exchange interaction between two indistinguishable particles (electrons) that is responsible for establishing long range ferromagnetic ordering. This interaction is purely quantum-mechanical in origin with no classical analogue.

2.2.1 Atomic Origin of Exchange

For well separated atoms, the wavefunction can be written as linear combinations of the Hydrogen atom wavefunctions $\phi(r)$. Consider only two atoms located at $R_i = 0$ and $R_i = R$, the Schrödinger’s equation for a one-electron wavefunction is given by:

$$E \psi(r) = -\frac{\hbar^2}{2m} \nabla^2 \psi(r) + V_0(r) \psi(r) = \mathcal{H}(r) \psi(r) \quad (2.7)$$

where $V_0(r) \sim \frac{Z}{r}$ is the attractive (Coulomb) potential of the nucleus, and $\mathcal{H}$ is the one-electron Hamiltonian for the two-atoms system. This leads to a one-electron eigenstates known as the Wannier functions:

$$\psi_s(r) = \frac{1}{\sqrt{2}} (\phi_L + \phi_R) \quad \psi_a(r) = \frac{1}{\sqrt{2}} (\phi_L - \phi_R)$$

with Hydrogen atom wavefunctions: $\phi_L(r) = \phi(r)$ and $\phi_R(r) = \phi(|r - R|)$. For two electrons, the Schrödinger’s equation becomes a bit more complicated:

$$E \Psi(r, r') = \mathcal{H}(r) \Psi(r, r') + \mathcal{H}(r') \Psi(r, r') + V_C(r, r') \Psi(r, r') \quad (2.8)$$

where $\Psi(r, r')$ is the two-electron wavefunction, and $V_C(r, r')$ is the Coulomb interaction between the two electrons. This two-electron wavefunction can be written in terms of the Wannier functions:

$$\Psi(r, r') = c_1 \phi_L(r) \phi_L(r') + c_2 \phi_L(r) \phi_R(r') + c_3 \phi_R(r) \phi_L(r') + c_4 \phi_R(r) \phi_R(r') \quad (2.9)$$

or in bra-ket notation:

$$|\Psi\rangle = c_1 |LL\rangle + c_2 |LR\rangle + c_3 |RL\rangle + c_4 |RR\rangle \quad (2.10)$$

Using the four wavefunctions $|LL\rangle$, $|LR\rangle$, $|RL\rangle$, and $|RR\rangle$ as our basis, straightforward evaluation of the energy matrix elements yields:

$$\mathcal{H} = 2E_0 + \begin{bmatrix} U & t & t & J_D \\ t & 0 & J_D & t \\ t & J_D & 0 & t \\ J_D & t & t & U \end{bmatrix} \quad (2.11)$$

Here

$$t = \int dV \phi^*(r)V_0(|r - R|)\phi(|r - R|) \quad (2.12)$$
Table 2.1: Eigenfunctions and eigenfunctions of the Hamiltonian described by equation 2.11

<table>
<thead>
<tr>
<th>Eigenfunction</th>
<th>Eigenenergy</th>
</tr>
</thead>
<tbody>
<tr>
<td>$</td>
<td>1\rangle = \frac{1}{\sqrt{2}}(</td>
</tr>
<tr>
<td>$</td>
<td>2\rangle = \frac{1}{\sqrt{2}}(</td>
</tr>
<tr>
<td>$</td>
<td>3\rangle = \frac{\sin \chi}{\sqrt{2}}(</td>
</tr>
<tr>
<td>$</td>
<td>4\rangle = \frac{\cos \chi}{\sqrt{2}}(</td>
</tr>
</tbody>
</table>

is the hopping integral; which represents the delocalized nature of electron wavefunction;

$$U = \iint dV dV' \phi^*_L(r) \phi^*_L(r') V_C(r, r') \phi_L(r) \phi_L(r')$$

(2.13)
is the Coulomb integral; which describes a strong repulsion between two electrons in an atom (typically $\sim$ a few eV); and

$$J_D = \iint dV dV' \phi^*_L(r) \phi^*_R(r') V_C(r, r') \phi_R(r) \phi_L(r')$$

(2.14)
is called the direct exchange. Unlike $U$, there is no classical equivalent of $J_D$. That is to say that $J_D$ is a purely quantum mechanical phenomena, with typically a few 0.01 eV order of magnitude. The eigenenergies and eigenfunctions are summarized in table 2.1 (note $\tan \chi = -4t/U$):

The total wavefunctions (including spin) of a two electron system need to be antisymmetric since electrons are fermions ($S = 1/2$). Hence, either the spatial part or the spin part of the wavefunction need to be antisymmetric, while the other is symmetric. From table 2.1 only $|1\rangle$ is antisymmetric and hence corresponds to a ferromagnetic spin configuration. The rest correspond to antiferromagnetic spin configurations. The exchange constant that is often quoted as responsible for ferromagnetism in solid is determined by the competition between the lowest-lying ferromagnetic state $E_1$ and the lowest-lying antiferromagnetic state $E_3$ [5].

Hence the exchange constant $J = \frac{1}{2}(E_1 - E_3)$ is given by:

$$J = J_D + \frac{U}{4} - \sqrt{t^2 + \frac{U^2}{16}}$$

(2.15)

This equation tells us that the exchange constant $J$ is not solely determined by the exchange integral $J_D$. In practice, taking the exchange interaction to be equal to exchange integral amount to a rather poor approximation. In various statistical models, the electronic quantities such as hopping and Coulomb interaction, are usually mapped onto spin variables, and are included as small perturbations.

The Heisenberg model can be realized from equation 2.11 by putting $U \to \infty$. In this limit, the energy eigenvalues are $E = E_0 \pm J_D$, corresponding to $J = J_D$. The Heisenberg model is most widely written by considering individual electrons as spins, and the
Heisenberg Hamiltonian is usually written as:

\[ \hat{H} = -\sum_{i,j} J_{ij} \mathbf{s}_i \cdot \mathbf{s}_j - g\mu_0 \mu_B \sum_i \mathbf{H}_i \cdot \mathbf{s}_i \]  (2.16)

where the summation includes all spins and \( \mathbf{H}_i \) is the local magnetic field acting on the \( i \)-th spin. If the spins are taken as continuous variable, we have the classical Heisenberg model. In the quantum mechanical case, the spins \( \mathbf{s}_i \) are replaced with the quantum spins \( \mathbf{\sigma}_i \), which for \( S = \frac{1}{2} \) are given by the vector Pauli matrix (see equation 2.3). The limiting cases of quantum Heisenberg model, namely the Ising model and XXZ model, are well studied and have been used to demonstrate symmetry breaking (ferromagnetic ordering) and phase transition in Heisenberg model.

By taking the limit \( U \to \infty \), we eliminate states with two electrons on one site, hence Heisenberg model works best for well separated atoms. Experimentally, however, we know that most well known ferromagnets such as the 3d transition metals Fe, Co, Ni, are actually metallic. Heisenberg model does not account for the existence of itinerant ferromagnets. In the appendix at the end of this chapter (section 2.5.1), I include a section on Hubbard’s model that can be used to explain how Coulomb interaction may be responsible for ferromagnetism in metals.

I will end this section by stating that while it is possible to study ferromagnetism by evaluating the spin-orbit interactions of many body systems from first principle, it is instructive (and easier) to use phenomenological expressions (power series expansions that take into account crystal symmetry and sample geometry) and take the coefficient from experiments. Case in point, the Mermin-Wagner-Berezinskii theorem states that the continuous symmetry cannot be spontaneously broken at finite temperature in systems with sufficiently short-range interactions in dimensions \( d \leq 2 \). For example, in the isotropic (classical) Heisenberg model in 2-dimension (also known as the XY-model), the number of spin waves (magnons) diverges since spin fluctuations can be excited with infinitely small energy which destroys any long range order. Yet in experiments, there exist ferromagnetic ultrathin films which can be approximated as a two-dimensional system. While this is partly because the ferromagnetic thin film is not a true two-dimensional system, it’s also because of the anisotropy present in the system. In the anisotropic case, magnetic moments cannot be rotated to any direction with infinitely small energy. This additional anisotropy energy breaks the continuous symmetry and is responsible for the occurrence of ferromagnetism in quasi-two-dimensional systems at finite temperature. This also highlights the importance of anisotropy in thin film ferromagnetism.
2.3 Ferromagnetism in Ultrathin Film

Micromagnetics is a field of physics dealing with the prediction of magnetic behaviors at sub-micrometer length scales. A micromagnetic model aims to explain the behavior of a magnetic system when subjected to an external stimuli, such as an external magnetic field, or some rf frequency excitation. Examples of problems in micromagnetics include predicting the hysteresis of a magnetic material from the material’s atomic or intrinsic parameters and calculating the resonance behavior of a magnetic thin film in response to an external rf field. In this section, we will start by consider a phenomenological model where each magnetic layer in a heterostructure as considered as a single spin.

2.3.1 Stoner-Wohlfarth Model

One of the simplest micromagnetic models is called the Stoner-Wohlfarth model (also known as uniform rotation, macrospin or coherent rotation model). The basic assumption of the Stoner-Wohlfarth model is that the magnetization is constant throughout the magnet (single domain), and the magnetization is represented by a single spin vector. Most of the magnetic interactions manifest themselves as some kind of anisotropy to the energy-scape of the single spin. The most important magnetic anisotropies in thin films are:

1. Shape anisotropy. Due to dipolar interaction (also known as demagnetization energy), the magnetization is affected by the macroscopic shape of the solid.

2. Magnetocrystalline anisotropy. The magnetization is oriented along specific crystalline axes.

3. Surface and Interface anisotropy. Surfaces and interfaces often exhibit different magnetic properties compared to the bulk due to their asymmetric environment. The interlayer exchange coupling can be thought of as another interface anisotropy term.

Shape anisotropy.

The origin of the shape anisotropy is the magnetic field (H-field) generated by the magnetization (M) in a magnetic body. The finite size of a magnetic sample causes poles on the surfaces of the magnetic body, which leads to a stray field outside of the sample. This stray field results in a demagnetizing field inside the sample. The energy of a sample in its own stray field is given by the demagnetizing energy:

\[ E_{\text{demag}} = -\frac{1}{2} \int \mu_0 \mathbf{M} \cdot \mathbf{H}_{\text{demag}} dV \]  

(2.17)

with \( \mathbf{H}_{\text{demag}} \) being the demagnetizing field inside the sample. For a uniform magnetization (i.e. single-spin approximation), the demagnetizing field can be written as:

\[ \mathbf{H}_{\text{demag}} = -\mathbf{N} \cdot \mathbf{M} \]  

(2.18)
with $\mathbf{N}$ being the demagnetizing tensor. The calculation of the demagnetizing tensor is complicated for a general shape, with the most efficient method involve calculating the convolution between the stray field of a point dipole with the shape of the magnetic body. Most micromagnetic simulation package divides the magnetic body into rectangular shape cells, and the demagnetizing field of each cell are calculated using Newell’s demagnetizing tensor $[7]$. The calculation is much simpler for symmetric objects. For example, in the case of a spherical magnetic body ($a = b = c$, the tensor $\mathbf{N}$ amounts to:

$$
\mathbf{N} = \begin{bmatrix}
\frac{1}{3} & 0 & 0 \\
0 & \frac{1}{3} & 0 \\
0 & 0 & \frac{1}{3}
\end{bmatrix}
$$

and the demagnetization energy density to:

$$
E_{demag}/V = \frac{1}{2} \mu_0 M^2 \frac{1}{3}(\alpha^2 + \beta^2 + \gamma^2) = \frac{1}{6} \mu_0 M^2
$$

Here we’ve taken $\mathbf{M} = M(\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta) = M(\alpha, \beta, \gamma)$ for the arbitrary magnetization direction, with $\theta$ being the angle from the $z$-axis, and $\phi$ the angle of the magnetization projection to the $xy$ plane from the $x$-axis. Hence, we found an isotropic behavior for the demagnetization energy since all directions are energetically equivalent for a spherical magnetic body. For an infinitely long cylinder ($a = b, c = \infty$):

$$
\mathbf{N} = \begin{bmatrix}
\frac{1}{2} & 0 & 0 \\
0 & 1/2 & 0 \\
0 & 0 & 0
\end{bmatrix}
$$

and the demagnetization energy density in this case is:

$$
E_{demag}/V = \frac{1}{2} \mu_0 M^2 \frac{1}{2}(\sin^2 \theta \cos^2 \phi + \sin^2 \theta \cos^2 \phi)
= \frac{1}{4} \mu_0 M^2 \cos^2 \theta
$$

For an infinitely expanded and very thin plate ($a = b = \infty$):

$$
\mathbf{N} = \begin{bmatrix}
0 & 0 & 0 \\
0 & 0 & 0 \\
0 & 0 & 1
\end{bmatrix}
$$

the demagnetization energy density is:

$$
E_{demag}/V = \frac{1}{2} \mu_0 M^2 \cos^2 \theta = K_{shape}^V \cos^2 \theta
$$

This result is important for thin magnetic film and multilayers. Equation (2.24) shows that the demagnetization energy can be written as a magnetic anisotropy energy. The energy is
minimized when $\theta = \pi/2$, that is, when the magnetization is within the film plane. In most thin magnetic film and multilayers, the demagnetization energy (shape anisotropy) dominates the total energy and hence it’s sometimes implicitly assumed that the magnetization is always within the film plane.

**Magnetocrystalline anisotropy.**

The magnetocrystalline anisotropy is caused by the spin orbit interaction of the electrons, which makes the spins prefer to align along well-defined crystallographic axes. Therefore, there are directions in space which a magnetic material is easier to magnetize in than in other ones (i.e. easy magnetization axes). The magnetocrystalline energy is usually small compared to the exchange energy, but the direction of magnetization is only determined by the anisotropy because the exchange interaction only tries to align the magnetic moments parallel, no matter in which direction.

In magnetic thin film, this term is usually smaller compared to the shape anisotropy. Hence the magnetocrystalline anisotropy behavior is usually observed on top of the shape anisotropy. For example, in face-centered-cubic (fcc) single crystal cobalt (Co) thin film on copper Cu(001), the primary behavior is set by the shape anisotropy, which causes Co magnetization to stay in-plane. The magnetocrystalline anisotropy shows up on top of this shape anisotropy, such that an in-plane four-fold anisotropy is established as an in-plane projection of the cubic anisotropy to the (001) plane. The four-fold anisotropy can be written as:

$$E/V = K_4 \sin^2(\theta_M) \cos^2(\theta_M) \tag{2.25}$$

where $\theta_M$ is the in-plane magnetization angle measured from the $+x$ direction. If $K_4 > 0$, equation 2.25 is minimized for $\theta_M = 0, \pi/2, \pi$, and $3\pi/2$. In the Co single crystal example stated earlier, these four angles should correspond to the four in-plane [110] easy-axis directions for Co magnetization.

**Surface and interface anisotropy.**

Due to the broken symmetry at interfaces, each effective anisotropy constant $K^{eff}$ is divided into two parts, one describing the volume and one the surface contribution:

$$K^{eff} = K^V + \frac{K^{S1} + K^{S2}}{d} \tag{2.26}$$

with $K^V$ being the volume dependent magnetocrystalline anisotropy constant and the two $K^S$ the surface dependent magnetocrystalline anisotropy constant. There are two $K^S$ term to account for the upper and lower surface of a film. The surface dependent term has inverse dependence on the thickness $d$ of the system, thus it’s only important for thin films.

An example of the surface induced anisotropy occurs in a vicinal substrate, a substrate whose surface normal deviates slightly from a major crystallographic axis. Epitaxial magnetic thin films grown on a vicinal substrate would have dangling bonds at the interface
which results in a magnetic uniaxial anisotropy along the steps of the vicinal substrate. The relative strength of this induced uniaxial anisotropy would be inversely proportional to the thickness of the magnetic thin film, since the interface interaction is the same no matter what the thickness of the thin film is. Another example of a surface anisotropy happens in interlayer-exchange-coupled magnetic heterostructures. To one of the ferromagnetic (FM) layer in a FM/spacer/FM trilayer, the interlayer exchange coupling with the other FM layer can be considered as adding a magnetic field to the layer. For example, if the interlayer coupling is written as $J \mathbf{m}_1 \cdot \mathbf{m}_2$, then we can write $H_1 = J \mathbf{m}_1$ as an equivalent magnetic field that is felt by $\mathbf{m}_2$. We can also think of this equivalent field as a unidirectional anisotropy on $\mathbf{m}_2$. In section 2.3.2 we will discuss this interlayer exchange coupling in more detail.

With most of the important magnetic interactions represented as anisotropy terms, we can write a macrospin simulation to calculate the hysteresis loop of a magnetic thin film. In figure 2.2 I show an example result of such a simulation for the hysteresis loop of a magnetic thin film with both four-fold anisotropy and uniaxial anisotropy. The simulation reproduces the experimental result quite well (see figure 3.8). The code for this simulation is included in section 2.5.2.

![Hysteresis Loop](image)

Figure 2.2: Simulated hysteresis loop from the macrospin model. The double-loop is a characteristic of a ferromagnet with both uniaxial and four-fold anisotropy

### 2.3.2 Interlayer Exchange Coupling

The exchange interaction described previously in section 2.2 occurs for very small separation $d_0$ of the order of a few Å, due to the overlap of electronic wave function. Magnetic interlayer
exchange coupling (IEC) describes the situation that the exchange interaction is mediated by
the interlayer material. Experimental data collected by different techniques and on various
multilayer systems revealed that there are three basic alignments of adjacent layer magneti-
zations: parallel, antiparallel, and at an angle of $90^\circ$ [8, 9]. These basic configurations are
phenomenologically described as the interlayer exchange coupling energy density per unit
area as follows:

$$E_{IEC}/A = -J_L \mathbf{\hat{m}}_1 \cdot \mathbf{\hat{m}}_2 - J_Q (\mathbf{\hat{m}}_1 \cdot \mathbf{\hat{m}}_2)^2$$

Here $J_L$ is called the bilinear coupling since the energy per area is linear in the directions
of both magnetizations $\mathbf{\hat{m}}$, with $J_L > 0$ ($J_L < 0$) favoring parallel (antiparallel) alignment
respectively. $J_Q$ is called the biquadratic coupling, with $J_Q > 0$ ($J_Q < 0$) favoring parallel
(orthogonal) alignment between the two ferromagnetic layers. $\theta$ here is the angle between
the two magnetizations.

The bilinear coupling term is relatively well understood. The RKKY (Ruderman Kittel
Kasuya Yoshida) interaction, together with the fact that in the transition metals spin mag-
netism prevail, can be taken as justification for this bilinear term. Systematic experiments
on various interlayer thicknesses reveal an oscillatory behavior of the magnitude of
$J_L$, with a typical periodicity of several Å. Consider a ferromagnet/non-magnetic metal/ferromag-
net (FM/NM/FM) trilayer system. The oscillatory behavior in this type of trilayer can be
roughly explained considering the itinerant nature of electrons in transition metal ferromag-
nets, which gives rise to an exchange-split band structure and spin-dependent reflectivities
at the NM/FM interfaces. The majority (minority) spins are weakly (strongly) reflected at
the interfaces since there is a good match between the majority band of the ferromagnet and
the interlayer (e.g. noble metal). Electrons in these majority spin states can freely move
from one material to the other. On the other hand, the minority spin electrons do not match
equally well, which gives rise to quantum well states (QWS) for the minority electrons. This
is illustrated in figure 2.3.

The description is similar to electrons in a one-dimensional potential well, where the QWS
form when the momentum component perpendicular to the layers $k_\perp$ fulfill the condition:

$$2k_\perp d_0 + \phi_C + \phi_B = 2\pi n; \quad n \in \mathbb{Z}$$

Here $d_0$ is the thickness of the spacer layer, $\phi_B$ and $\phi_C$ are the phase gains of the electron
wavefunction upon reflection at the two boundaries of the box, and $n$ is the number of
half-wavelengths confined inside the QW. For fixed $n$, equation 2.28 predicts decreasing
QW energies with increasing $d_0$, which is opposite from experimental observations [10].

Also, the oscillation periodicity at the Fermi level is $\pi/k_F$ from equation 2.28 instead of the
experimental observation of $\pi/|k_{BZ} - k_F|$ ($k_{BZ} = \pi/a$ is the Brillouin-zone vector). This
difference can be resolved by taking into account the discreteness of a thin film. If we take the spacer thickness as integer multiples ($m$) of the atomic spacing $a$, or $d_0 = ma$, then
equation 2.28 can be rewritten in terms of a new quantum number \( \nu \):

\[
2m\pi - 2k_\perp d_0 - \phi_C - \phi_B = 2\pi(m - n) \\
2k^e d_0 - \phi_C - \phi_B = 2\pi\nu
\]

(2.29)

where \( k^e = k_{BZ} - k_\perp \) and \( \nu = m - n \). The oscillation periodicity of the magnetic interlayer coupling from equation 2.29 is given by \( \pi/|k_{BZ} - k_F| \) at the Fermi level, as observed experimentally. This QW equation can be understood as a single one-dimensional potential well that is modulated by the (atomic) discreteness of the spacer layer. A more detailed theoretical treatment using realistic electronic band structures by considering the Fermi surface of the spacer material, where the oscillatory coupling is related to the critical spanning vector \( Q \) in reciprocal space that points perpendicular to the interface and connects two sheets of the Fermi surface.

The biquadratic coupling term on the other hand is much less understood. It is believed that the biquadratic term originate from extrinsic effects such as interface roughness or magnetic impurities in the interlayer. Fluctuation mechanism based on monolayer-thick roughness with some regions favoring parallel coupling and the rest favoring anti-parallel coupling has been proposed [11], leading to a non-analytic phenomenological formula:

\[
W = C_+ \{\theta\}^2 + C_- \{\theta - \pi\}^2
\]

(2.30)

for the mean energy of the interlayer coupling. The coupling coefficients \( C_\pm \) are positive definite, and they measure the mean contribution of the portions of the spacer favoring \( m_1 = \pm m_2 \) respectively. Nonetheless, the behavior of this equation is similar to that of equation 2.27, aside from asymptotic behavior toward saturation of the magnetization. In this dissertation, we’ll only consider interlayer coupling of the form used in equation 2.27.
2.4 Topology in Magnetism

Topology is the idea that inequivalent objects cannot be continuously transformed into each other. The most commonly used example is the equivalence between a coffee cup and donut (left side of figure 2.4). The coffee cup and donut are topologically equivalent because they can be transformed to each other without having to make a cut or puncture a hole on the surface of the object (animated[12]). Likewise, the coffee cup with incomplete handle is topologically equivalent to a sphere, and the trophy cup is equivalent to a double-torus. In these cases, we can assign a topological number (topological charge, or genus) to each group which relates to the Euler characteristic of the surfaces. We can simply think of the genus to be the number of handles on the two-dimensional surface.

![Figure 2.4: Example of objects that are topologically equivalent. In this figure, the genus, or topological number of each group corresponds to the Euler characteristic of the group.](image)

In magnetism, the questions of topology become important when considering non-uniform spin textures. Traditionally, study of magnetism in thin film focuses on the uniformly magnetized spins. In recent years, there has been an increase in the capability to prepare non-uniform spin textures through patterning, or by mixed-crystal engineering to include non-collinear exchange interaction. Some of these spin configurations are known as 'topological defects' as their stability can be traced to topological arguments[12]. Two magnetiza-
tion configurations are said to be topologically (or homotopically) equivalent if it is possible to deform them continuously into each other without having to surpass an infinite energy barrier or resort to the discreteness of the lattice.

To illustrate the concept topology in magnetism, let’s look at the following simple example. Consider a chain of spins in one dimension (1D). Each of the spins are allowed to rotate only in the $yz$-plane. A uniaxial anisotropy set the preferred spin direction in either $+z$ or $-z$ direction. Consider the arrangement shown in figure 2.5(a) and (b). At first glance, the single domain configuration in figure 2.5(a) (first configuration) is completely different from the three domains in figure 2.5(b) (second configuration). However, depending on the details of the domain walls in the second configuration, it may or may not be topologically equivalent to the first configuration. Figure 2.5(c) and (d) show two possible configurations of the domain walls. We can assign a topological charge (winding number) to each of these domain walls which correspond to sense of rotation of the spins in going from left to right. For figure 2.5(c), going from left to right, the first domain wall correspond to a clockwise rotation of the spins amounting to $+\pi/2$ rotation, and hence we assign a number $N = +1/2$ to this domain wall. The second domain wall correspond to a counter-clockwise rotation amounting to $-\pi/2$ rotation, and hence we assign a winding number of $N = -1/2$. The total winding number of this configuration is $N = 0$ which means this third configuration

\[ N = 0 \]

\[ \begin{align*} 
(a) & \quad N = 0 \\
(b) & \quad \begin{array}{c}
\hat{y} \\
\hat{z} \\
\hat{x}
\end{array} \\
(c) & \quad N = +\frac{1}{2} \quad N = +\frac{1}{2} \\
(d) & \quad N = -\frac{1}{2} \quad N = +\frac{1}{2}
\end{align*} \]

Figure 2.5: An example on how the concept of topology can be applied in magnetism.

of the domain walls. We can assign a topological charge (winding number) to each of these domain walls which correspond to sense of rotation of the spins in going from left to right. For figure 2.5(c), going from left to right, the first domain wall correspond to a clockwise rotation of the spins amounting to $+\pi/2$ rotation, and hence we assign a number $N = +1/2$ to this domain wall. The second domain wall correspond to a counter-clockwise rotation amounting to $-\pi/2$ rotation, and hence we assign a winding number of $N = -1/2$. The total winding number of this configuration is $N = 0$ which means this third configuration
is topologically equivalent to the single domain configuration. When we assign the numbers to figure 2.5(d), we obtained \( N = +1/2 \) for the first domain wall, and the same \( N = +1/2 \) for the second domain wall. The total topological charge (winding number) is then \( N = +1 \), which makes it inequivalent to the single domain configuration.

Now let’s imagine applying an increasing external magnetic field in the +\( z \) direction. This will cause the domain with spins pointing in the +\( z \) direction (red color) to expand as the spins would like to align with the external field. As a result, the two domain walls in figure 2.5(c) and (d) will approach each other as the spins flip from +\( z \) to −\( z \) direction. When the external field is strong enough, the two domain walls will collide and eventually all spins will point along the external magnetic field. For (c) configuration, this process happen smoothly, since the spins in the green domain only need to rotate through the −\( y \) direction (a counterclockwise rotation) in order to merge with the red domains. For (d) configuration, this process cannot happen as smoothly. As the two domain walls collide, it will form a metastable 2\( \pi \) domain wall. This 2\( \pi \) domain wall is a topological object that require a higher magnetic field to annihilate when compared to the (c) configuration. This process may involve rotating the spin to the (forbidden) ±\( x \)-direction, or by exploiting the discreteness of the spin chain, and thus breaking/changing the topology of the one dimensional spin chain from \( N = +1 \) to \( N = 0 \).

In the case of two dimensional ferromagnetic thin film, the topological charge is defined differently from the one dimensional case. Before assigning topological charge to different spin configurations, the allowed spin orientation (i.e. the spin-space) need to be defined. If the spins are confined to in-plane directions, in the same manner as the XY-model, then a winding number can be used to define the topology around a topological ’defect’ (see figure 2.6). Within the classical Heisenberg model (more details in section 2.5.3), the spin is a vector of fixed length that can point anywhere on a two-sphere \( S^2 \) (i.e. a spin-space two-sphere). For a spin configuration in an infinite two-dimensional plane to have a finite energy all spins at \( r \to \infty \) must point in the same direction. The topological charge of such configuration is given in the integral representation as:

\[
q = \frac{1}{4\pi} \int_{\mathbb{R}^2} \phi \cdot (\partial_1 \phi \times \partial_2 \phi) dx dy
\]

where \( \phi(x, y) \) is a vector field on the infinite two-dimensional plane with each point belonging to the spin-space two-sphere \( S^2 \), and \( q \) is called the Skyrmion number. This skyrmion number can be visualized with a two-step mapping process: first is the (conformal) mapping from an infinite two-dimensional plane onto a two-sphere in real-space. This is done through a stereographic projection as shown in figure 2.7. In this projection, all points at \( r \to \infty \) are mapped to a single point in the north-pole of the real-space two-sphere. The spins on these real-space two-sphere are then mapped onto the spin-space two-sphere, keeping the nearest-neighbor arrangements during the mapping. The skyrmion number (equation 2.31) counts the number of times the spin texture wraps around this spin-space two-sphere during the mapping process. A skyrmion (\( q = +1 \)) is defined to be a spin configuration that, when mapped to the spin-space two-sphere, wraps around the sphere exactly once.
Figure 2.6: Illustration of the vortex and antivortex 'topological defects' with different winding number as topological 'charge'.

Figure 2.7: A two dimensional magnetic skyrmion can be projected onto a sphere. A single skyrmion wrap around the two-sphere exactly once.

In real experiments, the spin lattice can’t be represented well by an infinite plane since experimental systems are necessarily finite. Unlike the pure isotropic Heisenberg model discussed earlier, dipolar interactions should be added to the Hamiltonian. This means that the single domain state may no longer be the lowest energy state possible (not a global minima). A rigorous mathematical formalism in these cases are lacking, but we can still make use of equation 2.31 as a measure of topological charge, since these additional interaction terms serve to create a smooth deformation to the spin texture from the pure Heisenberg model case. A well known example is the magnetic vortex state that occur when the lateral
dimension of the magnetic thin film is reduced to the nanoscale. In order to minimize the demagnetization energy cost, the spins form a ‘flux-closure’ domain where the magnetization circles around a core. In figure 2.8 we show an example of magnetic vortex state that occurs for 1 μm diameter Co disks grown on Cu(001) substrate. If we use equation 2.31 to evaluate the skyrmion number of a magnetic vortex state, we can simplify the equation into:

$$ q = \frac{1}{2} wp = \pm \frac{1}{2} $$

(2.32)

where \( w \) is the winding number of the spins circling around the core, and \( p \) is the orientation of the spins at the core of the vortex state (polarity). Because of the shape anisotropy of the magnetic nanodisk, only configurations with \( w = +1 \) is allowed, whereas \( p \) can be either \( \pm 1 \). Furthermore, to minimize the demagnetization energy, the helicity (\( \gamma \), defined to be the whirling angle between the spins and a radial unit vector) of these spin configurations are limited to \( \gamma = \pm \pi/2 \), or clockwise (\( c = -1 \)) and counterclockwise (\( c = +1 \)) circulation around the core respectively. In figure 2.9 we enumerate the four possible magnetic vortex state that can be stabilized in a magnetic nanodisk: Note that while the skyrmion number (equation 2.32) is independent of the circulation of the vortex, an energy barrier exist between the two configurations with \( c = +1 \) and \( c = -1 \) due to the shape anisotropy. For the two polarities, the energy barrier is determined by the exchange stiffness of the spins at the vortex core.

A significant amount of today's research in thin film magnetism are dedicated to the control of the magnetic vortex state with different means of excitation. In particular, the switching from one of the four possible states to another can be achieved using magnetic field pulses [13], or spin current excitations [14] at appropriate conditions. These are motivated
Figure 2.9: Enumeration of the four possible vortex states in a magnetic nanodisk.

No doubt, the study of topology in magnetism is still in the early phases and there are plenty of discoveries to be made.

by the possible application in a logic or storage devices. Another driving force is the search for skyrmion-materials with expanded phase diagram in order to realize a robust skyrmion lattice at room temperature, which may be used in low-powered electronic/spintronics devices.
2.5 Appendix

2.5.1 Hubbard Model

Hubbard model is one of the simplest model to describe interacting particles in a lattice. It only has two terms in the Hamiltonian: the hopping term $t$ and the on-site repulsion $U$. This model retains the non-local characteristic of the electron wave-function through the hopping term while including a short-range electron-electron interaction through the on-site repulsion $U$. Hubbard model is a useful starting point in studying ferromagnetism in itinerant electron system. Heisenberg exchange has been identified as the origin of ferromagnetism in a system where the atoms are well-separated, which often means the materials are insulating with well-localized wave-function. This is a severe restriction (overcorrelated limit) since interatomic hopping lowers the energy and gives rise to some ionicity on the lattice sites. Hubbard model addresses this problem by allowing electron hopping and capturing the essential Coulomb interaction in the Hamiltonian.

Within the language of second-quantization, the simplest Hubbard model Hamiltonian can be written as:

$$H = -t \sum_{<i,j>, \sigma} c^\dagger_{j,\sigma} c_{i,\sigma} + U \sum_i n_{i,\uparrow} n_{i,\downarrow} \quad (2.33)$$

We can interpret this Hamiltonian as follows: allowing electrons to ‘hop’ between neighboring sites (allowing electrons to de-localize) lowered its energy, as long as no two electrons are in the same site, in which case the energy is penalized due to electron-electron repulsion. The concept of second-quantization is perhaps the most familiar when introduced for the harmonic oscillator Hamiltonian as the ladder operators.

$$\hat{\mathcal{H}} = \frac{1}{2m} \hat{p}^2 + \frac{1}{2} m \omega^2 \hat{x}^2 = \hbar \omega (\hat{a}^\dagger \hat{a} + \frac{1}{2}) \quad (2.34)$$

$\hat{a}^\dagger$ and $\hat{a}$ are the raising and lowering operators respectively. These operators are more commonly referred to as the creation and annihilation operators of quasiparticles and are defined in terms of the position and momentum operators:

$$\hat{a} = \sqrt{\frac{m \omega}{2 \hbar}} \hat{x} + i \sqrt{\frac{1}{2m \omega \hbar}} \hat{p} \quad (2.35)$$

$$\hat{a}^\dagger = \sqrt{\frac{m \omega}{2 \hbar}} \hat{x} - i \sqrt{\frac{1}{2m \omega \hbar}} \hat{p} \quad (2.36)$$

Similarly, the operators $c^\dagger_{i,\sigma}$ and $c_{i,\sigma}$ in equation 2.33 are also creation and annihilation operators, defined in much the same way as it was for harmonic oscillators. Operator $c^\dagger_{j,\sigma}$ creates an electron of spin $\sigma$ at site $j$, and operator $c_{i,\sigma}$ annihilates an electron of spin $\sigma$ at site $i$. The operators defined as $\hat{n}_{i,\uparrow} = \hat{a}^\dagger_{i,\uparrow} \hat{a}_{i,\uparrow}$ is the number operator that will return the eigenvalue of 1 if site $i$ is occupied by a spin $\uparrow$ electron and will return 0 otherwise.
Figure 2.10: Illustration of the second order perturbation in half-filled Mott insulator. The energy correction from the addition of small hopping constant prefers an antiferromagnetic ground state.

It is interesting to address the type of magnetic order that arises in the ground state of a half-filled Mott Insulator (large $U$ limit). It can be shown that if the hopping is treated as a small perturbation to the unperturbed one-site Hamiltonian, a staggered (antiferromagnetic) order of the electron spins is favored. This arises since a second-order perturbation, which in this case is an electron hopping twice (from site $i$ to $j$ then back to site $i$), always reduces the ground state energy while the first order perturbation is not allowed since a single hopping will change the ground state. Mathematically this goes as follows: At half filling the ground states of the one-site ($U = 0$) limit are highly degenerate since there is only one electron per site and different spin-configuration does not change the ground state energy $E_0$. If the hopping term is now included as a small perturbation, defining $|G\rangle$ as the true ground state of the perturbed Hamiltonian with energy $E_G$, then the energy correction can be written as:

$$E_G = E_0 + E^{(1)} + E^{(2)} \tag{2.37}$$

with the first and second order correction given as:

$$E^{(1)} = \left\langle G \bigg| -t \sum_{<i,j>\sigma} c^\dagger_{j\sigma} c_{i\sigma} \bigg| G \right\rangle \tag{2.38}$$

$$E^{(2)} = \sum_m \frac{\left\langle G \bigg| -t \sum_{<i,j>\sigma} c^\dagger_{j\sigma} c_{i\sigma} \bigg| m \right\rangle \left\langle m \bigg| -t \sum_{<i,j>\sigma} c^\dagger_{j\sigma} c_{i\sigma} \bigg| G \right\rangle}{E_m - E_G} \tag{2.39}$$

The first order energy correction is zero since $|G\rangle$ is perpendicular to all $c^\dagger_{j\sigma} c_{i\sigma} |G\rangle$. The maximum correction coming from the second-order correction occurs when $|G\rangle$ has a staggered (antiferromagnetic) order, which means that every electron on site $i$ has four neighbours of opposite spins (due to Pauli exclusion principle). This also means that there are four
intermediate states $|m\rangle$ for each site $i$ with energy $U - t$ in which the electron at site $i$ hops onto one of its neighbour, gaining energy $t$ but costing the Coulomb repulsion energy $U$ as illustrated in the right figure of figure 2.10.

We conclude that adding a small hopping as perturbation to the one-site ($U = 0$) Hamiltonian lifted the degeneracy of the ground state. In this case the new ground state is the antiferromagnetic ground state with (corrected) energy of

$$E_G = -\frac{4t^2}{U}$$

where we’ve ignored $t$ in the denominator since $U >> t$. Examples of materials that can be described adequately using this model are the insulating antiferromagnets CoO, NiO, and MnO.

2.5.1.1 Ferromagnetism in Hubbard Model

It is easy to see that the addition of the on-site repulsion term in the Hubbard Hamiltonian gives rise to some ferromagnetic characteristic. If we write the magnetization as $m = n\uparrow - n\downarrow$, and given a fixed total number of electrons $n = n\uparrow + n\downarrow$, the second term will give us:

$$Un\uparrow n\downarrow = -U\left(\frac{m + n}{2}\right)\left(\frac{m - n}{2}\right) = \frac{U}{4}(n^2 - m^2)$$

This means that Coulomb interaction (on-site repulsion) favors the formation of magnetic moment, though this is not the only consideration in Hubbard model and we must also consider the average hopping energy.

Stoner criterion for ferromagnetism.

Stoner criterion is a very simple picture of ferromagnetism based on the competition between the kinetic energy cost of making the up and down spin electron number different and the associated potential energy gain. Consider a system with density of states $N(E)$ and both up and downspin electrons filling the energy levels up to the same maximum called the ‘Fermi Level’ $E_F$. The density of up and down electrons are both equal to $n$. Computing the change in energy which results from a reduction in the density of down spin electrons by $\delta n$ and at the same time an increase of the number of up spin electrons by $\delta n$. The potential energy (of electron-electron interaction given by Hubbard $U$) changes by,

$$\delta P = U(n + \delta n)(n - \delta n) - Un^2 = -U(\delta n)^2$$

This shift of $\delta n$ electrons costs some energy since they now occupy energy levels above the original $E_F$. We can write $\delta n$ in terms of the density of states in energy as $\delta n = N(E_F)\delta E$ where $N(E_F)$ is the density of states at the Fermi energy. The net result is the shift of $\delta n$
Figure 2.11: If the density of states and/or the Coulomb interaction is strong enough then it is favorable to have ferromagnetic order. This is known as Stoner criterion and is given by $UN(E_F) > 1$ for ferromagnetic ordering to be favorable.

Electrons up in energy by an amount $\delta E$, resulting in total a change in the kinetic energy of:

$$\delta K = +\delta n\delta E = +\frac{1}{N(E_F)}(\delta n)^2$$

Putting this together with the change in potential energy, we’ll get:

$$\delta E = \delta P + \delta K = (-U + \frac{1}{N(E_F)})(\delta n)^2 = (-UN(E_F) + 1)\frac{(\delta n)^2}{N(E_F)}$$

From here we obtained Stoner criterion: if $UN(E_F) > 1$ the total energy change $\delta E < 0$, so it’s favorable to have a different up and down electron densities and hence ferromagnetic order is favored. **Ferromagnetic ordering is favored by large electron interaction.**
2.5.2 Python Code for Macrospin Simulation

```python
# file macrospin.py
__author__ = 'Ali Sucipto Tan'

import numpy as np
from scipy.misc import derivative

class MacroSpin(object):
    # Simulates hysteresis loops given the energy-scape of a magnetic system
    
    def __init__(self, energy):
        # In the macrospin model, the magnetization is a vector of fixed length that is characterized only by
        # the angle.
        self.angle = 0
        self.hx = None
        self.energy = energy

    def set_field_sweep(self, hx):
        self.hx = hx
        return self.hx

    def find_minima(self, h, step=1e-3, precision=1e-9, random_factor=0.01):
        # For current energy-scape, find magnetization angle that minimizes total energy from starting position angle_init
        angle_old = self.angle
        self.angle = self.angle + \n        (np.random.ranf() - 0.5)*random_factor
        energy_scape = self.energy(h)

        while abs(self.angle - angle_old) > precision:
            angle_old = self.angle
            self.angle = angle_old - \n            step*derivative(energy_scape, self.angle, dx=step)
            print '{}'.format(self.angle),

        return self.angle
```
def calc_mx(self):
    mx = np.array([])
    for h in self.hx:
        self.angle = self.find_minima(h)
        mx = np.append(mx, np.cos(self.angle))
        #print 'Current field value is %r' % round(h, 2)
    return mx

An example simulating the hysteresis loop of a thin film with both four-fold anisotropy and uniaxial anisotropy:

from macrospin import *
import numpy as np
import matplotlib.pyplot as plt

hx = np.append(np.delete(np.linspace(6, -6, 101), -1),
                np.linspace(-6, 6, 101))

def energy(H):
    return lambda angle: -2*(np.sin(angle))**2
                        + (np.sin(angle)*np.cos(angle))**2
                        - H*np.cos(angle)

sim = MacroSpin(energy)
sim.set_field_sweep(hx)

mx = sim.calc_mx()

fig = plt.figure(figsize = (10, 6))
ax = fig.add_subplot(111)
pl1 = ax.plot(hx, mx, 'rs-', label='Hysteresis')
ax.set_ylim(-1.1, 1.1)
# Shrink current axis by 20%
box = ax.get_position()
ax.set_position([box.x0, box.y0, box.width * 0.8, box.height])

# Put a legend to the right of the current axis
ax.legend(loc='center left', bbox_to_anchor=(1, 0.5))
plt.show()
2.5.3 Topological Spin Texture from Nonlinear Sigma Model

The Heisenberg ferromagnetic model can be mapped into the nonlinear O(3) sigma model by considering the spin vectors on a lattice as a classical field configuration. In particular, by taking the lattice as a (2+1)-dimensional Minkowski spacetime $\mathbb{R}^{2+1}$ and confining the spin vectors into a unit sphere $S^2$, the dynamics of the field is governed by the Lagrangian action density:

$$L = \frac{1}{2} \partial_{\mu} \phi \partial^{\mu} \phi$$  \hspace{1cm} (2.45)

This leads to a total energy \[15\] given by

$$E(\phi) = \frac{1}{2} \int_{\mathbb{R}^2} \left\{ (\partial_1 \phi)^2 + (\partial_2 \phi)^2 \right\} dx$$  \hspace{1cm} (2.46)

If we require finite energy from this integral, it is necessary for $\phi$ to be a constant unit vector at infinity. This means that $\phi$ must be a continuous map from $S^2$ to $S^2$, and it represents a homotopy class in the homotopy group

$$\pi_2(S^2) = \mathbb{Z}$$  \hspace{1cm} (2.47)

This model admits a lower energy bound $E(\phi) \geq 4\pi|q|$ for each integer $q$ specifying the homotopy class \[16\]. In this case, the true ground state of this model is given by a uniform state $\phi = (0,0,\pm 1)$ where $q = 0$. $q$ can be seen intuitively as the number of times the unit sphere $S^2$ of a total surface area $4\pi$ is being covered under a stereographic projection of $\phi$ from $\mathbb{R}^2$. In literature this number is often called the skyrmion number, and the integral representation of $q$ is given by:

$$q = \frac{1}{4\pi} \int_{\mathbb{R}^2} \phi \cdot (\partial_1 \phi \times \partial_2 \phi) dxdy$$  \hspace{1cm} (2.48)

Note that for a perfectly smooth configurations (spatially continuous spin texture), the total skyrmion number is independent of time. However, since the original microscopic model is defined on a lattice, processes that change $q$ by some integer amount are allowed \[17\]. The continuum approximation of the spins as classical field is justified as long as we analyze phenomena with characteristic wavelength larger than the lattice constant. The topological barrier provides a protection from small disturbances which makes a topological phase of a spin texture a good, robust candidate for use in topological quantum computing.

In real systems, various other interactions can be added to the Hamiltonian, which will modify the action defined by equation \[(2.45)\]. These additional interactions, if well defined, will provide a smooth deformation to the ground state of each homotopy class. This way it is possible to stabilize spin textures with $q \neq 0$ as a ground state. The most well known example is the Dzyaloshinskii-Moriya (DM) interactions:

$$H_{DMI} = D \int_{\mathbb{R}^2} \phi \cdot (\nabla \times \phi) dx$$  \hspace{1cm} (2.49)
which breaks chiral symmetry and favors the formation of a skyrmion lattice. Other examples are skyrmions stabilized by dipole-dipole interactions [18] or a combination of interlayer exchange coupling and shape anisotropy [19].
Chapter 3

Experimental Techniques

In this chapter, I will describe the experimental techniques that are used throughout this dissertation. In section 3.1 I present an overview of the molecular beam epitaxy (MBE) technique. This section includes the requirements for achieving ultrahigh vacuum (UHV) condition necessary for MBE, description of our sample preparation process used to achieve epitaxial growth, and a description of a low-cost *in situ* stencil lithography used to create patterned ferromagnetic film samples. Section 3.2 describes the Magneto-Optical Kerr Effect (MOKE), which is our technique of choice to measure the magnetization of a ferromagnetic thin film sample. An extension of this technique using a rotating magnetic field (known as Rotating-field MOKE, or ROTMOKE) is also included. Finally, in section 3.3 characterization techniques using X-rays generated by a synchrotron light source will be briefly described.

3.1 Molecular Beam Epitaxy

The ultra-high Vacuum is defined to be the vacuum regime characterized by pressures lower than $10^{-9}$ Torr. Most of surface science work is done in such an environment so that the sample surface can be cleaned and maintained in the cleaned state for the duration of time needed for the experiment[20]. In MBE, especially if ultrathin film thickness is desired, it is necessary to maintain the UHV condition throughout the sample growth process.

Recall that the density of the gas molecules in the chamber can be approximated by the ideal gas law:

$$PV = nRT = Nk_BT$$  \hspace{1cm} (3.1)

where $R = k_BN_A$ and $N = nN_A$ while $k_B = 1.38 \times 10^{-23} \text{JK}^{-1}$ is the Boltzmann constant and $N_A = 6.02 \times 10^{23}$ is the Avogadro’s number. Assuming the velocities of the gas molecules follow Maxwell’s distribution, it can be derived that the mean free path of the gas molecules
Table 3.1: Parameters affected by pressure

<table>
<thead>
<tr>
<th>Degree of Vacuum</th>
<th>Pressure (Torr)</th>
<th>Gas Density (molecules. m^{-3})</th>
<th>Mean Free Path (m)</th>
<th>Time/ML (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atmospheric</td>
<td>760</td>
<td>$2 \times 10^{25}$</td>
<td>$7 \times 10^{-8}$</td>
<td>$10^{-9}$</td>
</tr>
<tr>
<td>Low</td>
<td>1</td>
<td>$3 \times 10^{22}$</td>
<td>$5 \times 10^{-5}$</td>
<td>$10^{-6}$</td>
</tr>
<tr>
<td>Medium</td>
<td>$10^{-3}$</td>
<td>$3 \times 10^{19}$</td>
<td>$5 \times 10^{-2}$</td>
<td>$10^{-3}$</td>
</tr>
<tr>
<td>High</td>
<td>$10^{-6}$</td>
<td>$3 \times 10^{16}$</td>
<td>50</td>
<td>1</td>
</tr>
<tr>
<td>Ultrahigh</td>
<td>$10^{-10}$</td>
<td>$3 \times 10^{12}$</td>
<td>$5 \times 10^5$</td>
<td>$10^4$</td>
</tr>
</tbody>
</table>

at a given temperature and pressure is given by:

$$l = \frac{k_B T}{\sqrt{2 \pi d^2 p}}$$  \hspace{1cm} (3.2)

Here $T$ is the temperature in $K$, $p$ is the pressure in Pascal, and $d$ is the diameter of the gas particles in meters. The mean free path of a gas molecule is inversely proportional to the pressure. At at ambient pressure (1 atm = $1.013 \times 10^5$ Pa = 760 Torr) the mean free path is 68 nm, whereas at ultra-high vacuum pressure of $10^{-8} - 10^{-12}$ Torr, the mean free path reaches into thousands of kilometers.

One of the crucial factors determining how long a surface can be maintained clean is the incident molecular flux, i.e. how long it takes for one monolayer (1 ML) of adsorbed gas to cover the surface. This is the number of gas molecules impacting on the surface from the gas phase, and is given by the Hertz-Knudsen formula:

$$\frac{1}{A} \frac{dN}{dt} \equiv \phi = \frac{\alpha p}{\sqrt{2 \pi m k_B T}}$$  \hspace{1cm} (3.3)

where $0 \leq \alpha \leq 1$ is called the sticking coefficient of the gas molecules onto the surface, or the probability that molecules hitting the surface will stay on the surface. Table 3.1 lists the time it takes assuming $\alpha = 1$ for a cleaned surface to be completely covered by 1 ML of adsorbed gas. During materials deposition, the molecular flux determined the rate at which the evaporation should take place in order for the deposited film to be free from impurities. For example, in molecular beam epitaxy, the typical growth/evaporation rate of materials is $\sim 0.5$ Å/min, or $\sim 0.25$ ML/min for epitaxy on copper Cu(001) substrate. If this is attempted at high vacuum ($\sim 10^{-6}$ Torr), where molecular flux is about 1 ML/s, most of the film grown will not be pure, but rather composed of adsorbed gases that stick to the susbtrate. On the other hand, at ultrahigh vacuum, the molecular flux is much smaller, with 1 ML coverage taking the time on the order of hours. For the same reasoning care must be taken such that the MBE chamber remains in ultrahigh vacuum throughout the entire heterostructure growth process.
3.1.1 Achieving Ultra-High Vacuum (UHV) Condition

In our labs, the UHV environment is achieved by pumping a stainless steel chamber in three stages. The first stage is usually handled by a mechanical or diaphragm pump. These can pump out gases with high volumetric flow rate but are usually unable to hold high vacuum condition on the inlet side. A typical pumping station would combine a mechanical pump or a diaphragm pump with a small turbomolecular pump to achieve high volumetric flow rate as well as a reasonably high vacuum. A pumping station could reliably provide mid-vacuum down to $10^{-5}$ Torr pressure. The second stage is pumping by a larger turbomolecular pump, which can hold even lower pressure and achieve high vacuum ($10^{-7}$ Torr). A backing pump (or roughing pump) is required in the operation of a turbomolecular pump. This is because the for the pumped gas to escape from the rotating angled blades of the turbine (usually several thousands of rpm), the mean free path of the gas at the outlet must be larger than the gap between the moving blades. As the typical size of the gap is on the order of 1 mm, the exhaust pressure should be less than $10^{-2}$ Torr, where the mean free path is about 5 mm. The last stage of pumping is by an ion capture pump, which is capable of reaching pressures as low as $10^{-11}$ Torr under ideal conditions. An ion pump works as a Penning trap [21], where a swirling cloud of electrons produced by an electric discharge ionizes gas within the vacuum chamber. The ion pump employs a strong electrical potential (a few kilovolts), which allows the electron to accelerate into and be captured by a solid, reactive metals such as tantalum or titanium. The bombarding captured ions effectively sputters the cathode, exposing fresh titanium which acts to evacuate the gas by physical or chemical adsorption, resulting in a net pumping action. Another type of pump called the Titanium Sublimation Pump (TSP) can be used to help the ion pump. In a TSP pump, titanium filament is put inside the vacuum chamber near the ion pump, and is heated up with large current (40-50 A) for a few minutes. The evaporated titanium will be adsorbed into the inside walls of the chamber and the ion pump cathode, which will adsorb ions more readily.

To measure the pressure inside a vacuum chamber, two kinds of gauges are most often used. The first one is the Pirani gauge (thermo gauge) which can measure pressure down to $10^{-3}$ Torr. Since the gauge only covers low vacuum range, it is often used to check the vacuum between a mechanical pump and a turbo pump. The Pirani gauge head is based around a heated wire placed in a vacuum system, the electrical resistance of the wire is proportional to its temperature. At atmospheric pressure, gas molecules collide with the wire and remove heat energy from it (effectively cooling the wire). As gas molecules are removed (i.e. the system is pumped down) there are less molecules and therefore less collisions. Fewer collisions mean that less heat is removed from the wire and so it heats up. As it heats up, its electrical resistance increases. A simple circuit utilizing the wire detects the change in resistance and once calibrated can directly correlate the relationship between pressure and resistance. In this way you can use a calibrated meter to indicate pressure. The pressure of ultrahigh vacuum chamber can be measured by the hot-filament ionization gauge (ion gauge for short). The ion gauge consists of three distinct parts, the filament, the grid, and the collector. The filament is used for the production of electrons
by thermionic emission. The grid has positive voltage which pulls the electrons from the filament. Electrons circulate around the grid passing through the fine structure many times until eventually they collide with the grid. Gas molecules inside the grid may collide with circulating electrons causing a fraction of them to be ionized. The collector inside the grid has negative voltage and attracts these positively charged ions. The number of ions collected by the collector is directly proportional to the number of molecules inside the vacuum system allowing a measure of molecular gas density and hence pressure in the chamber.

If we start from atmospheric pressure of 760 Torr (i.e. with the inside of the vacuum chamber exposed to air), and we pump down the chamber through the three stages of pumping described earlier, chances are we will not be able to reach vacuum pressure of lower than $10^{-8}$ Torr. At extremely low pressures, more gas molecules are adsorbed on the walls than are floating in the chamber. Water vapor is a significant source of outgassing because a thin layer of water vapor rapidly adsorbs to everything whenever the chamber is exposed to air. The UHV pressure range is only achievable by baking the UHV chamber to a temperature above 100° C for several hours to remove water vapor and other trace gases that adsorb on the inner surfaces of the chamber.

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**Figure 3.1**: Left: a photo of the ultrahigh-vacuum chamber used in this dissertation. Right: a schematic of materials deposition process for molecular beam epitaxy.
3.1.2 Substrate Preparation

Metallic single crystals copper (Cu) and silver (Ag) are typically used in our laboratory. Although Cu(001) and Ag(001) are commercially available, in most cases more steps of polishing are required before using them as substrates for ultrathin films. The first stage of polishing is the mechanical planarization. The substrate is mounted on an epoxy and ground into the desired shape using various grit sand-paper. If necessary, the flat substrate can be shaped into a stepped or curved surface during this process. The substrates are prepared by a series of mechanical polishing on a lapping machine. After planarization, the substrates are put through a series further of chemical-mechanical planarization using diamond, alumina $\text{Al}_2\text{O}_3$ (for Ag substrate), or silica (for Cu substrate) slurry with increasingly finer grain size in conjunction with a polishing pad on a lapping machine. The typical steps are mechanical polishing with 6 $\mu$m diamond suspension, followed by mechanical polishing with 1 $\mu$m micron diamond suspension. The final polishing for Ag substrate uses the 0.05 $\mu$m polycrystalline alumina suspension. Cu substrate on the other hand uses the 0.02 $\mu$m silica suspension. A period of cleaning in distilled water with ultrasonic cleaner (sonicator) is usually done in-between polishing steps.

After the final step of mechanical polishing, the surface of the substrates should have a mirror-like finish. The substrates is then taken out from the epoxy by dissolving the epoxy in acetone. The final polishing step can be repeated if necessary by finger-holding the substrates carefully on the lapping machine at suitable speed. At this stage, although the substrates have mirror-like surfaces, they may not be very clean or flat because small particles of Cu or Ag may fill the scratches or defects to make the surfaces look clean. Chemical polishing (for Ag) or electrochemical polishing (for Cu)) may be used to remove these small particles from the substrates and further reduce the surface roughness. Before doing the electrochemical polishing, the substrate must be cleaned with acetone in an ultrasonic cleaner. The following steps are used for the electrochemical polishing of Cu substrates:

1. Mix 40 ml phosphoric acid (85%) to 9 ml distilled water and then add 5 ml sulfuric acid (98%) to the mixture.

2. Set the power supply to output constant voltage of 1.8V and turn it off. Put a Cu cathode into solution and connect the anode to the cleaned and dried substrate and put it into the solution.

3. Turn on the power supply and apply 1.8V of constant voltage between the cathode and the crystal. Make sure to apply positive to the crystal and ground to the solution.

4. As soon as the voltage is applied, bubbles should form around the crystal and the cathode. After $\sim$20 seconds when the current drops and stabilizes, take the crystal out of the solution and immediately rinse with distilled water followed by sonication. Afterwards rinse and sonicate further with acetone.
After the electrochemical polishing the Cu substrate should look clean with very few defects. Sometimes after this process the surface may appear cloudy, which may be caused by large imperfections on the surface due to mechanical polishing. In this case, the mechanical polishing and electrochemical polishing should be repeated to obtain a flat and clean Cu substrate (for example, see figure 3.2).

Figure 3.2: A copper Cu(001) substrate in between polishing steps. In this photo, the copper substrate had been polished with 1 µm diamond paste slurry on the lapping machine

For Ag substrate, the chemical polishing is done using a solution made up of saturated chromic acid and 3 ml of hydrochloric acid (HCl). The chemical polishing is performed by swabbing the silver surface with a cotton-tipped applicator saturated with the solution and rinsing the sample intermittently in running water. The Ag substrate is then rinsed with distilled water followed by cleaning in acetone in an ultrasonic bath. After this procedure, the Ag substrate should be optically flat with few defects. Glass/ceramic substrates such as Magnesium Oxide single crystals, SiN₃ membrane and ferroelectric PMN-PT are also used in this work. These substrates are usually already polished well when purchased. In this case, the substrates are cleaned with acetone bath in the ultrasonic cleaner and dried before loading the sample into the vacuum chamber. To mount the substrates into sample holders for use in UHV chamber, small pieces of tantalum (Ta) foils are spot-welded on the sample holder to hold the substrate in place. The all-metal construction of the sample holder ensures very little outgassing when the substrate is put into the UHV environment compared to using less-friendly adhesives or epoxy such as silver paste or carbon tape. It also ensures that the substrate will be firmly affixed to the sample holder at high temperature (> 600°C) or low temperature (~ −150°C). The substrates are loaded into the vacuum chamber through
a load-lock system, which is an intermediate chamber between the UHV main chamber with the atmosphere. The load-lock facilitates transferring samples in and out of the vacuum chamber without directly exposing the main chamber to atmosphere.

After the substrate is loaded into the main UHV chamber, the final sample treatment process can be done in situ. For metallic single crystal substrates such as Cu and Ag, the surface should be treated with cycles of sputtering and annealing. The sputtering of the Cu/Ag substrates is achieved with high kinetic energy Ar\(^+\) (Argon) ions with 1 ∼ 5 keV energy. The Ar\(^+\) ions bombarded the surface of the substrate and removed the contaminant rich top layers. Ar is chosen for sputtering because as an inert noble gas, Ar does not interact chemically with other material. This chemical inertness ensures that the Ar\(^+\) ions themselves do not become a contaminant either by staying on the surface of the substrate during sputtering or by being absorbed on the inside surface of the chamber. By the same reasoning, the Ar gas cannot be effectively removed by the ion pump as Ar atoms are only held at the cathode of ion pump by a weak van der Waals force, preventing the surface refreshing process of ion pump. Hence, to prolong the life of the ion pumps, the ion pump should be isolated from the main chamber housing the substrate and the sputtering gun. This is usually achieved by a gate valve between the ion pump and the main chamber.

This sputtering process, while very effective in removing the top layers off from the substrate, also makes the surface rough. In order to smoothen the surface after sputtering, the substrate is subsequently annealed. The substrate annealing was done by heating up the substrate to ∼ 600°C and slowly cooled down over > 60 minutes. At elevated temperatures, surface atoms and adatoms have higher mobility, which allows them to relieve the mechanical stress and fill in defects. Slow cooling gives the atoms enough time to settle in position where they have the lowest energy and arrange themselves into the appropriate crystal structure. Several cycles of sputtering and annealing are necessary before one can obtain a clean and well-ordered surface. Note that for some other substrates such as MgO or PMN-PT, the only preparation step taken before sample growth is annealing to promote outgassing of impurities.

3.1.3 Materials Deposition: Thermal and Electron-Beam Evaporation

The clean and ordered substrate can now act as seed crystals for other materials. Deposition of a crystalline overlayer on a crystalline substrate is commonly known as epitaxy (from Greek epi, meaning 'above' and taxis, meaning 'an ordered manner'). Epitaxial growth can be achieved when the atoms being deposited and the substrate has a similar lattice constant. Table 3.2 lists the lattice constants of commonly used materials in this work. For example, from this table we can see that the lattice constants of the face-centered-cubic (f.c.c.) phase of iron (Fe), cobalt (Co), and nickel (Ni) are within a few percent to the lattice constant of copper (i.e. -0.5% for Fe, 1.9% for Co, and 2.5% for Ni). The same is true for alloys made of Fe and Ni (permalloy, Py). This is the reason why, although the f.c.c. phase of
### Table 3.2: Common materials parameter for epitaxy considerations

<table>
<thead>
<tr>
<th>Atoms/Molecules</th>
<th>Lattice constant (Å)</th>
<th>Crystal structure</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cr</td>
<td>2.91</td>
<td>b.c.c.</td>
</tr>
<tr>
<td>Mn</td>
<td>8.9125</td>
<td>b.c.c.</td>
</tr>
<tr>
<td>Fe</td>
<td>2.8665</td>
<td>b.c.c.</td>
</tr>
<tr>
<td>γ-Fe (&gt; 1000 K) [22]</td>
<td>3.6320</td>
<td>f.c.c.</td>
</tr>
<tr>
<td>Co</td>
<td>2.5071</td>
<td>h.c.p</td>
</tr>
<tr>
<td>Co (&gt; 720 K) [23]</td>
<td>3.548</td>
<td>f.c.c.</td>
</tr>
<tr>
<td>Ni</td>
<td>3.524</td>
<td>f.c.c.</td>
</tr>
<tr>
<td>Cu</td>
<td>3.6149</td>
<td>f.c.c.</td>
</tr>
<tr>
<td>Py (Ni$<em>{80}$Fe$</em>{20}$)</td>
<td>3.55</td>
<td>f.c.c.</td>
</tr>
<tr>
<td>Ag</td>
<td>4.0853</td>
<td>f.c.c.</td>
</tr>
<tr>
<td>MgO</td>
<td>4.211</td>
<td>periclase (rock salt)</td>
</tr>
<tr>
<td>CoO</td>
<td>4.2615</td>
<td>periclase (rock salt)</td>
</tr>
<tr>
<td>NiO</td>
<td>4.1767</td>
<td>periclase (rock salt)</td>
</tr>
</tbody>
</table>

Fe and Co only occur naturally at high temperatures, they can be stabilized by epitaxial growth on a single-crystal Cu(001) substrate. Furthermore, the large density of states at the Fermi level of 3d-block transition metals allows for rich magnetic properties to emerge from ultrathin epitaxial films Fe, Co, Ni grown on Cu(001). In the same manner, CoO, NiO, Cr, and body centered cubic (b.c.c.) Fe can be grown epitaxially on single crystal Ag(001) substrate or MgO(001) substrate. In the case of Cr and b.c.c. Fe, the lattice parameter in the [100] direction of Cr (2.91 Å) and Fe (2.8665 Å) coincide with the lattice parameter in the [110] direction of Ag (4.0853/√2 = 2.889 Å) and MgO (4.211/√2 = 2.978 Å). These allow epitaxial growth of ferromagnetic Fe and antiferromagnetic Cr, CoO, and NiO on Ag(001) and MgO(001) substrates.

In molecular beam epitaxy (MBE), a source material is heated to produce an evaporated beam of particles. In our laboratories, this is achieved by one of two means: thermal evaporation, or electron-beam evaporation (see figure 3.3). For thermal evaporation, the materials for evaporation are put inside an alumina (aluminum oxide) crucible with tungsten wires wrapped around it. The tungsten wire is heated by flowing current around 10-20 Amperes (with voltages 10-20 Volts, leading to 100-400 Watt of dissipative heat), depending on the sublimation temperature of the materials to be evaporated. The crucible is typically covered with an external shielding made of tantalum foil with an aperture. This shielding is used to focus the radiative heating onto the crucible, and also to prevent excessive outgassing from the vacuum chamber near the evaporators. In a typical operation of a thermal evaporator, a full outgassing of the thermal evaporator is required before depositing materials on the substrate. This is done by applying current below the evaporation current but with enough dissipative heat to outgas impurities present in the materials. A general rule of thumb I usually use is that the evaporator can be called clean if during evaporation, the pressure is...
lower than twice the base pressure of the chamber. For example, if the base pressure of the chamber is $2 \times 10^{-10}$ Torr, then during deposition, the pressure should be less than $4 \times 10^{-10}$ Torr.

![Figure 3.3: Left: A thermal evaporator with copper inside an aluminium oxide crucible. Right: A four-pocket electron-beam evaporator from Mantis Deposition Ltd.](image)

Electron beam evaporation is another way to produce evaporated beam of particles. In an e-beam evaporator, a hot cathode or thermionic cathode is heated to make it emit electrons due to thermionic emission. The thermally emitted electrons are accelerated by using high voltage and the kinetic energy of electrons are transferred as thermal energy of the materials to be evaporated. The cathode used in our electron-beam evaporation is usually the Lanthanum Hexaboride ($\text{LaB}_6$) coated filaments or thoriated tungsten filament. The advantage of evaporation using electron beam is that it allows evaporation of materials with high sublimation/melting point. It also produces comparatively smaller radiative heating which allows the substrate to be kept at constant temperature throughout sample growth process.

The thickness of the deposited thin film is calibrated by measuring the rate of evaporation using quartz crystal microbalance (QCM). The mass variations per unit area is measured by measuring the change in frequency of a quartz crystal resonator. The change in the oscillation frequency is correlated with the mass deposited on it by the Sauerbrey equation:

$$\Delta f = -\frac{2f_0^2}{A\sqrt{\rho_q\mu_q}} \Delta m$$

where $f_0$ is the resonant frequency in Hz, $\Delta f$ the frequency change, $\Delta m$ the mass change, $A$ is the piezoelectrically active area, $\rho_q = 2.648$ g/cm$^3$ is the density of quartz, and $\mu_q = 2.947 \times 10^{11}$ g cm$^{-1}$s$^{-2}$ is the shear modulus of quartz. If the materials being deposited is very different from quartz, with a large mismatch in the shear modulus, then the Z-match method must be used to determine the change in mass. The formula for the Z-match method
Table 3.3: Growth parameters for Quartz Crystal Deposition Monitor

<table>
<thead>
<tr>
<th>Material</th>
<th>Density (g/cm³)</th>
<th>Z-ratio</th>
<th>Material</th>
<th>Density (g/cm³)</th>
<th>Z-ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cr</td>
<td>7.20</td>
<td>0.305</td>
<td>Ni</td>
<td>8.91</td>
<td>0.331</td>
</tr>
<tr>
<td>Mn</td>
<td>7.20</td>
<td>0.377</td>
<td>Co</td>
<td>8.90</td>
<td>0.343</td>
</tr>
<tr>
<td>Co</td>
<td>8.90</td>
<td>0.343</td>
<td>Ag</td>
<td>10.50</td>
<td>0.529</td>
</tr>
<tr>
<td>Fe</td>
<td>7.86</td>
<td>0.349</td>
<td>Au</td>
<td>19.30</td>
<td>0.381</td>
</tr>
<tr>
<td>CoO</td>
<td>6.44</td>
<td>0.412</td>
<td>NiO</td>
<td>7.45</td>
<td>1.000</td>
</tr>
<tr>
<td>Py (Ni₈₀Fe₂₀)</td>
<td>8.90</td>
<td>1.000</td>
<td>MgO</td>
<td>3.580</td>
<td>0.411</td>
</tr>
</tbody>
</table>

is given by:

$$\frac{\Delta m}{A} = \frac{N_2 \rho_2}{\pi Z f_L} \tan^{-1}[Z \tan(\pi \frac{f_U - f_L}{f_U})]$$  \hspace{1cm} (3.5)$$

Here $\Delta f = f_U - f_L$, where $f_U$ is the resonant frequency of unloaded quartz crystal and $f_L$ is the resonant frequency of loaded quartz crystal. In practice, the correction due to this acoustic impedance mismatch is hard-coded into the quartz crystal deposition monitor controller, and an accurate thickness can be calibrated for different materials using the densities and Z-ratios of commonly used materials in table 3.3.

The quartz crystal is usually mounted on a stage that can be moved inside the UHV chamber. The evaporation rate is measured with the quartz crystal put in the position where the substrate would be during the actual materials deposition. After the evaporation rate from either thermal evaporator or e-beam evaporator is stable, the quartz crystal can be put away. A shutter is used to block the molecular beam while the substrate is being put in the right position. The thickness of the thin film being deposited can then be controlled precisely by the deposition time after the shutter is opened and closed. From experience, this method can give us thin film thicknesses that are accurate to within 2 Å, or about 1 ML thickness. For ultrathin film with sub-monolayer accuracy, a wedged shape thin film must be grown in order to ensure the right film thickness is deposited somewhere on the substrate. The slope of the wedged sample is typically a few monolayers over 3-5 mm distance, or of the order of 1 ML/mm. With such a shallow slope, locally the thin film is effectively flat, and can be treated as continuous film.

The advantage of a wedged sample is two-fold. The first one is that with a wedged sample structure, we will always obtain the right thin film thickness even if the calibration of evaporation rate is not perfect, or changes over time. The second is that by growing a wedge, the effect of thickness variations on the physical properties that we are going to measure can be studied without having to worry about the influence of the variations in substrate or sample growth conditions.

In our group’s research over the past few years, we are interested in patterned magnetic nanostructures. We developed a method to deposit magnetic nanodisks with about 1 µm diameter using an in situ stencil lithography technique (see figure 3.5).
CHAPTER 3. EXPERIMENTAL TECHNIQUES

Figure 3.4: A wedge structure for deposition of materials with various thicknesses are made by moving the substrate behind a shutter blade during deposition

3.1.4 Low Energy Electron Diffraction

Low energy electron diffraction is used to determine the structural properties of the surface of crystalline materials. A collimated electron beam of low energy electrons are directed to the surface of the single crystalline substrate/sample. The energy of these low energy electrons correspond to a de Broglie wavelength given by the relation:

$$E = \frac{p^2}{2m_e} = \frac{h^2}{2m_e\lambda^2} = \frac{150.4 eV}{\lambda^2}$$

(3.6)

where $h$ is the Planck’s constant, $m_e$ the mass of electron, and $\lambda$ is the electron’s wavelength in Å. Since the lattice constant of a typical single crystalline sample on the order of 1 Å, then electron matter wave with similar wavelength should be used, which lead to a typical electron energy of 20-200 eV. The elastically back-scattered electrons form diffraction patterns on a fluorescent screen according to the Bragg’s diffraction condition:

$$2d\sin\theta = n\lambda$$

(3.7)

Generally, the structural information given by a LEED pattern results from the position and the intensity of the diffraction spots as well as from the spot profiles. In particular, the surface unit cell of the reciprocal lattice and the corresponding real space unit cell follow from the positions of the LEED spots. For example, for a fcc single crystal copper with (001) face, the first diffraction spot appears in the [110] direction, the position of the nearest neighbour of a Cu atom. From the spot profiles the quality and degree of long range order at the surface can also be deduced. In practice, the LEED patterns are used as an indicator of the quality of our epitaxially grown single crystalline sample. It’s also used to confirm the crystal orientation of our substrates.
Figure 3.5: (a) Sketch of the *in situ* stencil lithography developed to produce patterned structures. (b) The mask used as stencil, with 1 µm diameter holes and 4 µm edge to edge hole distance. (c) Optical microscope photo of Co disks grown using the stencil lithography method. (d) A PEEM contrast image of Co disks grown using the shadow mask technique.
Figure 3.6: Left: Sketch of a low energy electron diffraction (LEED) experiment. Right: Example of LEED patterns of a trilayer Ni/Fe/Co sample epitaxially grown on single crystal Cu(001)
3.2 Magneto-Optical Kerr Effect (MOKE)

Magneto-Optical Kerr Effect is a powerful tool with monolayer sensitivity used to study surface magnetization of a magnetic thin film. When linearly polarized light goes through or is reflected from a magnetized medium, the difference in the refractive index experienced by the left and right circularly polarized components result in the rotation of the linearly polarized light. This effect is known as Faraday rotation, or Kerr rotation in the case of reflected light. Phenomenologically, the dielectric property of a medium is characterized by a $3 \times 3$ tensor $\epsilon_{ij}$ with $i, j = 1, 2, 3$. This dielectric tensor can be decomposed into symmetric and antisymmetric parts. The normal modes of the symmetric tensor are linearly polarized light along the three principal axis. Therefore, the symmetric part of the dielectric tensor does not give rise to the Faraday effect\cite{24}. The antisymmetric part of $\epsilon_{ij}$ is responsible for Faraday effect (and Kerr effect). The antisymmetric part of $\epsilon_{ij}$ can be written as:

$$\bar{\epsilon} = \epsilon \begin{bmatrix} 1 & iQ_z & -iQ_y \\ iQ_z & 1 & iQ_x \\ iQ_y & -iQ_x & 1 \end{bmatrix}$$

The two normal modes are the left-circular polarized light ($E_y = iE_x$) with $\epsilon_L = \epsilon(1 - Q \cdot \hat{k})$ and right-circular polarized light ($E_y = iE_x$) with $\epsilon_R = \epsilon(1 + Q \cdot \hat{k})$ where $Q = (Q_x, Q_y, Q_z)$ is known as the Voigt vector and $\hat{k}$ is the unit vector along the light propagation direction. The complex Faraday rotation of the polarization plane after traveling a distance $L$ within a magnetized medium is:

$$\theta = \frac{\pi L}{\lambda} (n_L - n_R) = -\frac{\pi L n}{\lambda} Q \cdot \hat{k}$$

where $n$ is the refractive index. The real part of equation\cite{3.9} gives rise to Faraday rotation, and the imaginary part gives the Faraday ellipticity. It is instructive to note that the external magnetic field has a stronger effect on the polarization plane of light than an external electric field. Since electric field is unchanged under time reversal symmetry, and magnetic field undergoes a change of sign, Onsager’s relation gives $\epsilon_{ij}(E, H) = \epsilon_{ij}(E, -H)$, where $E$ and $H$ are the external electric and magnetic fields, respectively. Expanding $\epsilon_{ij}$ up to terms that are linear in the electric and magnetic field, we can conclude that the antisymmetric part of $\epsilon_{ij}$ comes from the magnetic field.

Classically, the different dielectric properties of right and left circularly polarized light can be understood by analyzing electron motion in response to an electromagnetic wave. Without magnetic field present, a circular-polarized electric field will drive the electron into circular motion with the same radius for both right and left circularly polarized light, and thus there is no Faraday rotation. With magnetic field applied in the propagation direction of the electromagnetic wave, there is an additional Lorentz force acting on each electron. The direction of this Lorentz force is opposite for right or left circular motions and thus expands or shrinks the resultant radii of the circular electron orbit. This difference yields the difference.
in the corresponding dielectric constants, and we conclude it is the Lorentz force of the magnetic field that generates the Faraday effect. This, together with equation 3.9, shows that the Faraday rotation $\theta$ is proportional to the local magnetic field $H$ experienced by the electron motion, and thus to the magnetization $M$. Quantum mechanical description of the Faraday rotation uses the spin-orbit interaction to couple electron spin with its motion, and thus the spin to electromagnetic wave.

![Figure 3.7: A Magneto-optical Kerr Effect setup in the longitudinal configuration.](image)

The basic working principle of a MOKE setup is to measure the change in polarization of light as it is reflected from a magnetized surface. A p-polarized light (linearly polarized parallel to optical plane) is delivered to the magnetic surface from a Helium-Neon laser ($\lambda = 632.8$ nm) through a polarizer (figure 3.7). We have concluded earlier that the Kerr rotation $\theta$ of the reflected light is proportional to the magnetization along the propagation direction of light $M \cdot \hat{k}$. This polarization change can be measured, for example, by using a second polarizer in a near cross-polarization configuration. If the analyzer is set in a cross polarized configuration (extinction condition), the intensity of light reaching the photodetector will be proportional to the square of the magnetization ($I \sim |E|^2 \sim |Q|^2$) instead of a linear dependence. This disadvantage can be circumvented by rotating the analyzer a small angle $\delta$ away from extinction. The intensity measured by the photodetector in this configuration becomes linearly proportional to the magnetization $[24]$

$$I(M) = I_0(1 + \frac{2\phi'(M)}{\delta})$$

(3.10)

Here $\phi'(M)$ is the Kerr rotation angle when the magnetization is $M$ and $I_0 = |E_p|^2\delta^2$ is the average background signal. A magnetic hysteresis loop can be obtained by measuring
the Kerr rotation angle while an external sweeping magnetic field is applied to the magnetic sample. Example of MOKE measurement of hysteresis loop is shown in Figure 3.8.

Figure 3.8: An example of a hysteresis loop obtained using MOKE. In this example, the MOKE optics was set to measure hysteresis loop for magnetic field applied along a four-fold anisotropy easy-axis and a uniaxial anisotropy hard-axis.

One of the most important consideration in a MOKE setup is the extinction condition, i.e. how close to zero the light intensity is when the two polarizers are set to cross-polarization configuration. Hence it is important to use a high-extinction-ratio ($< 1 : 100,000$) Glan-Taylor polarizers to obtain a pure linear polarization and hence smallest possible light intensity at extinction. Both polarizers and sometimes also quarter-wave plate need to be fine-tuned with a precision rotation mount. A typical value for voltage measured by the photodetector at extinction is $V_{\text{ext}} \sim 0.1 \text{ nV}$. The estimated sensitivity of a well-aligned MOKE setup is about $10^{-5}$ radian.

### 3.2.1 ROTMOKE

The rotating-field MOKE (ROTMOKE) is an optical technique specialized in measuring the magnetic anisotropy of magnetic thin films. In this technique, the projection of the magnetization vector along a given direction is measured using MOKE while an external magnetic field of constant magnitude drags the magnetization to rotate within the film plane. Since MOKE is able to reach monolayer sensitivity, ROTMOKE, by extension, is
also able to reach monolayer sensitivity. In most ROTMOKE apparatus, the rotating field is produced by physically rotating the electromagnet with a stepping motor while keeping the sample stage and the corresponding MOKE measurement apparatus fixed. While the noise associated with the mechanical vibration of the sample can be eliminated in such design, the mechanical rotation of a heavy electromagnet usually takes at least 10 min to finish a complete rotation of 360°. While fast fluctuations at < 1 min scale can be easily averaged out by repeating measurement, slow (or long-period) fluctuations at > 1 min scale usually results in a slow drifting of the background signal that could overwhelm the measurement signal. For example, the laser intensity drift within 1 to 10 min time-scale was comparable or greater than the Kerr rotation from monolayer thick magnetic thin films so that the MOKE signal usually drifts noticeably after one round rotation of the magnet.

Other than obtaining a laser light source with greater stability, another solution to the drifting intensity problem is to simply speed up the rotation of the magnetic field. It is usually difficult to mechanically rotate an electromagnet with a period of ∼10 s per round without causing other problem. To solve this issue, we designed and assembled a vector magnet that rotates the magnetic field without a mechanical rotation of the electromagnet. This is achieved by a four-pole electromagnet as shown in figure 3.9. The upper and lower (left and right) coils are connected in series. Two identical bipolar power supplies are used to supply current \( I_x \) and \( I_y \) to each of the coil pairs. When current is applied only to the upper and lower coils (i.e. \( I_x > 0 \) and \( I_y = 0 \)), a horizontal magnetic field is produced \( H_x > 0 \). Likewise when current is applied only to the left and right coils (i.e. \( I_x = 0 \) and \( I_y > 0 \)), a vertical magnetic field is produced \( H_y > 0 \). A rotating field is produced by a suitable superposition of the two magnetic fields given a (to first order) linear relationship between magnetic field \( H_x \) and \( H_y \) and the coil currents \( I_x \) and \( I_y \).

The typical raw data obtained from a ROTMOKE experiment consist of two arrays: an array of applied field angle \( \theta_H \), and an array of measured Kerr angle, usually the output from the photodiode in volts \( V \). The measured Kerr angle represents the component of the magnetization along the optical plane of the laser, which means that \( V \) is proportional to \( \cos(\theta_M) \). These information, together with the knowledge of the strength of the rotating field \( H_{ROT} \), can be used to retrieve the magnetic anisotropy quantitatively by calculating the torque balancing condition in a magnetic thin film\[25\]. For a magnetic thin film consisting of a uniaxial anisotropy \( K_2 \) and a 4-fold anisotropy \( K_4 \), the energy density for a single domain state is given by:

\[
\frac{E}{V} = -M_S H_{ROT} \cos(\theta_H - \theta_M) + K_2 \sin^2(\theta_M) + K_4 \sin^2(\theta_M) \cos^2(\theta_M)
\]  

(3.11)

Here \( M_S \) is the magnitude of the ferromagnetic film magnetization. The first term represents the Zeeman energy of the magnetization within the applied magnetic field \( H_{ROT} \). The equilibrium magnetization angle \( \theta_M \) is obtained by minimizing Eq. (3.11) with respect to \( \theta_M \), which leads to the torque moment \( l(\theta_M) \) as follows:

\[
\frac{d(E/V)}{d\theta_M} = -M_S H_{ROT} \sin(\theta_H - \theta_M) + 2K_2 \sin \theta_M \cos \theta_M + K_4 \sin 2\theta_M \cos 2\theta_M
\]  

(3.12)
Figure 3.9: Design of the vector magnet for ROTMOKE measurements. The vector magnet is controlled by two bipolar power supply in current source mode. One power supply controls the magnetic field in the $x$ direction $H_x$ and the other one controls magnetic field in the $y$ direction $H_y$.

Setting $\frac{d(E/V)}{d\theta_M} = 0$ and rearranging the terms lead to:

$$l(\theta_M) = H_{\text{ROT}} \sin(\theta_H - \theta_M) = -\frac{2K_2}{M_S} \frac{1}{2} 2 \sin \theta_M \cos \theta_M - \frac{2K_4}{M_S} \frac{1}{4} 2 \sin 2\theta_M \cos 2\theta_M$$

$$l(\theta_M) = -\frac{1}{2} H_2 \sin 2\theta_M - \frac{1}{4} \sin 4\theta_M$$

(3.13)

The anisotropic fields are defined as $H_2 = \frac{2K_2}{M_S}$ and $H_4 = \frac{2K_4}{M_S}$. Typically the ROTMOKE experiment is setup such that $\theta_M = 0$ coincides with the $+x$ direction. Here $H_2 > 0$ ($H_2 < 0$) means the $+x$ direction is along the easy-axis (hard-axis) of $K_2$. Likewise
$H_4 > 0$ means the easy-axis of $K_4$ are along the $x$- and $y$- axes and the hard-axes are at 45° from the $x$- and $y$- axes. In practice, the $l(\theta_M)$ curve can be extracted from the experimental data $V(\theta_H)$, which is then fitted with equation 3.13 to yield the anisotropic field constants. The Python code used to extract the magnetic anisotropy constant from the raw data is included in an appendix at the end of this chapter at section 3.4. The code extracts the torque moment $l(\theta_M)$ from the raw data $V(\theta_H)$ with a known rotating magnetic field strength $H_{ROT}$. The code folds the data using the method of Ref. [25] to remove the quadratic Voigt effect. The downside is that we would possibly lose information of unidirectional anisotropy such as exchange bias from our data. Note that the folding method is not necessary in order to extract the anisotropy information, but it will improve the fitting result. The code fits $l(\theta_M)$ using the least-square method with the equation:

$$y = A + H_1 \sin\left(\frac{x - x_1}{180}\right) + \frac{1}{2} H_2 \sin\left(\frac{2x - x_2}{180}\right) + \frac{1}{4} H_4 \sin\left(\frac{4x - x_4}{180}\right)$$

(3.14)

with fitting parameters $A, H_1, x_1, H_2, x_2, H_4, x_4$ used to account for additional background or misalignment artifacts in the data. An example of a fitted ROTMOKE data is shown in 3.10. In this example, the fitted parameters are $H_2 = -257.13$ Oe, and $H_4 = 226.0$ Oe, with the R squared value (coefficient of determination) of 0.9975.

Figure 3.10: A typical example of a ROTMOKE data fit
CHAPTER 3. EXPERIMENTAL TECHNIQUES

3.3 X-ray Spectroscopy and Microscopy

In the simplest picture of resonant X-ray absorption, a photon transfers its energy to a core electron and the electron is excited into an unoccupied electronic state. The absorption cross-sections are calculated by the time-dependent perturbation of the sample by the electromagnetic (EM) field. Formally the transition probability up to second order can be written according to Fermi’s golden rule [26]:

$$T_{if} = \frac{2\pi}{\hbar} \left| \langle f | H_{int} | i \rangle \right|^2 \sum_n \frac{\langle f | H_{int} | n \rangle \langle n | H_{int} | i \rangle}{\epsilon_i - \epsilon_n} \delta(\epsilon_i - \epsilon_f) \rho(\epsilon_f)$$  \hspace{1cm} (3.15)

Here $T_{if}$ is the transition probability per unit time from state $|i\rangle$ to a state $|f\rangle$, with the interaction Hamiltonian $H_{int}$ given by:

$$H_{int} = \frac{e}{m_e} \mathbf{p} \cdot \mathbf{A}$$  \hspace{1cm} (3.16)

The evaluation of this transition probability is complicated, but by quantizing the electromagnetic field, the matrix element relevant to the transition between two electronic states $|a\rangle$ and $|b\rangle$ have the general form $\mathcal{M} = \langle b | \mathbf{p} \cdot e^{ikr} | a \rangle$. Working with dipole approximation, we can write the polarization dependent X-ray absorption resonance intensity as:

$$I_{res} = \mathcal{A} |\langle b | \mathbf{\epsilon} \cdot \mathbf{r} | a \rangle|^2$$  \hspace{1cm} (3.17)

where $\mathbf{\epsilon}$ is the unit photon polarization vector. With the wavefunctions written as linear combination of atomic orbitals (states of the form $|n, l, m_l, s, m_s\rangle$), and by inspecting the nonvanishing matrix elements in the transition matrix, we obtain the dipole selection rules: $\Delta l = \pm 1, \Delta m_l = q = 0, \pm 1, \Delta s = 0,$ and $\Delta m_s = 0$, where $qh$ is the X-ray angular momentum with $q = 0$ for linearly polarized x-ray and $q = +1, -1$ for left and right circularly polarized x-ray respectively.

Note that the strong localization of the core shell makes X-ray absorption spectroscopy (XAS) element-specific and sensitive to the valence shell properties within the atomic volume. At the absorption thresholds of the elements the spectrum shows strong resonances arising from transitions to unfilled valence band states. Since the transitions are governed by the $\Delta l = \pm 1$ selection rule, the $d$ band transition metals are best studied using $L_{2,3}$ edges ($2p \rightarrow 3d$) and the rare earths using $M_{4,5}$ edges ($3d \rightarrow 4f$). XAS is also sensitive to the chemical environment, as shown by broadening/modification of the spectra to show the fine structure of the absorption edges. Table 3.4 show the absorption edges of some elements used in this dissertation. Element-specific magnetic hysteresis loops and magnetic domain images in this dissertation are obtained using x-ray magnetic circular dichroism (XMCD) or x-ray magnetic linear dichroism (XMLD) techniques at the X-ray magnetic spectroscopy endstation at beamline 4.0.1 and PEEM-3 endstation at beamline 11.0.1 of the Advanced Light Source at the Lawrence Berkeley National Laboratory. This third generation X-ray...
synchrotron facility provides soft X-rays with tunable energy range of 150-2000 eV with energy resolution of $E/\Delta E = 4000$ at 800 eV using a 5.0-cm period elliptical polarization undulator. X-ray Absorption Spectroscopy (XAS) are typically measured by means of total electron yield (EY) detection. Electron Yield detection is often used in the soft X-ray region where the Auger decay channel dominates over fluorescence. Experimentally, this is achieved by simply using a picoammeter for the measurement of photocurrent. For PEEM measurements, secondary electrons emitted by the decay of core hole are collected by a series of electron optics, and captured by a CCD camera.

Table 3.4: Resonant X-ray absorption edge for $2p \rightarrow 3d$ transition in some 3d transition metals

<table>
<thead>
<tr>
<th>Element</th>
<th>$L_3$ peak (eV)</th>
<th>$L_2$ peak (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe</td>
<td>706.8</td>
<td>719.9</td>
</tr>
<tr>
<td>Co</td>
<td>778.1</td>
<td>793.2</td>
</tr>
<tr>
<td>Ni</td>
<td>852.7</td>
<td>870.0</td>
</tr>
</tbody>
</table>

X-ray Magnetic Circular Dichroism (XMCD).

X-ray Magnetic Circular Dichroism (XMCD) is used to measure the size and direction of magnetic moments. The origin of this dichroism is similar to MOKE, where spin alignment and spin orbit coupling causes a polarization dependent absorption of light. In the case of soft X-ray beam, the element-specificity allows for measurement of magnetization coming from specific element in a sample. This allows layer-by-layer analysis of the magnetic heterostructures, whereas MOKE only measures the sum of all the magnetization along the optical path. XMCD intensity can be summarized in a two-step process:

1. Circularly polarized X-rays generate photoelectrons with a spin and/or orbital momentum from a localized atomic inner shell to the Fermi level in the 3d band.

2. The 3d shell serves as the ‘detector’ of the spin or orbital momentum of the photoelectron. If the 3d shell has nonzero magnetic moment, right and left circularly polarized X-rays will yield different photoelectron intensity, due to the dipole selection rule.

The XMCD intensity could be written as:

$$I_{XMCD} \propto P_{circ} \mathbf{m} \cdot \mathbf{L}_{ph} \propto P_{circ} \langle \mathbf{m} \rangle \cos \theta$$  \hspace{1cm} (3.18)

where it depends on three parameters: the degree of circular photon polarization $P_{circ}$, the expectation value of magnetic moment of the 3d shell $\langle \mathbf{m} \rangle$, and the angle $\theta$ between the magnetization and the X-ray photon momentum. The actual spin and orbital moment can be derived according to several sum rules based on the two XAS spectrums with left and right circularly polarized X-rays [26]. In practice, to obtain the XMCD value and use it as
CHAPTER 3. EXPERIMENTAL TECHNIQUES

a measure of magnetization, the XAS value at the $L_3$ edge is measured with left and right circularly polarized X-rays, yielding $I_L$ and $I_R$ respectively. The XMCD is then calculated as:

$$XMCD = \frac{I_L - I_R}{I_L + I_R} \propto \mathbf{m} \cdot \hat{k}$$

(3.19)

Other than the element-specificity, the advantage of magnetization measurement using XMCD is that the XMCD value is immune to changes in the optical plane of the X-ray as the electron yield is unchanged if the X-ray flux remains constant. This makes a systematic measurements along a wedged sample more reliable.

Figure 3.11: A typical example of XMCD spectrum (Figure adapted from [26]). The X-ray absorption spectra for right circularly polarized X-rays are different when the sample is magnetized along (blue curve) or against (red curve) X-ray direction.

X-ray Magnetic Linear Dichroism (XMLD).

X-ray Magnetic Linear Dichroism (XMLD) spectroscopy is used to determine the orientation of the antiferromagnetic axis in an antiferromagnetic single crystals CoO and NiO. The electric field vector $\mathbf{E}$ of linearly polarized x-rays acts as a search light for the number of valence holes in different directions of the atomic volume (since the electric field vector oscillates in time along an axis and the radiation may be absorbed at any time, linearly polarized x-rays are only sensitive to axial not directional properties). In most cases the anisotropy of the charge in the atomic volume is caused by an anisotropy in the bonding, i.e. by the electrostatic potential. In the presence of spin order the spin-orbit coupling leads
to preferential charge order relative to the spin direction even in cubic systems. As a consequence the charge exhibits a small anisotropy in the unit cell, i.e. it is no longer spherical but shows an ellipse-like distortion about the magnetic direction. This charge anisotropy lifted some degeneracy in the core energy levels in a manner similar to crystal field splitting, which leads to an asymmetry of the x-ray absorption spectrum.
3.4 Appendix: Python Code for ROTMOKE Data Analysis

```python
import numpy as np
import matplotlib.pyplot as plt
from scipy.optimize import leastsq

# Fold mx data to symmetrize and normalize
def norm(m):
m = m - 0.5*(np.max(m) + np.min(m))
return m/np.max(np.abs(m))

# Calculate HK2, HK4 and plot torque vs magnetization angle
def rmoke(fname, H=300, plot=True):
data = np.genfromtxt(fname, delimiter='	')
phi = data[:, 0]
mx = data[:, 1]

mx_fold = norm(0.5*(mx + np.flipud(mx)))

# Extract magnetization angle from mx data
theta = np.arccos(mx_fold)

# Put theta in the [0, 2\pi] range
theta_2 = 2*np.pi - theta
theta[37:] = theta_2[37:]

# Extract the torque information
torque = H*np.sin(np.pi*phi/180 - theta)

# Fit the data with several sine function

# First guess of the fit parameter
hk1 = 0
x1 = 0
hk2 = 10
x2 = 0
```
hk4 = 0
x4 = 0
background = 0  # a[4]

param = leastsq(func, [hk1, x1, hk2, x2, hk4, x4, background])

b = param[0]

fit = b[0]*np.sin(theta - np.pi*b[1]/180)\ 
    + b[2]*np.sin(2*(theta - np.pi*b[3]/180))/2\ 
    + b[4]*np.sin(4*(theta - np.pi*b[5]/180))/4\ 
    + b[6]

# Calculate r-squared value
mean = np.mean(torque)
ss_tot = np.sum((torque - mean)**2)
ss_res = np.sum((torque - fit)**2)
rsquared = 1 - ss_res/ss_tot

print 'HK2 = %r, HK4 = %r,
      R-squared = %r' % (b[2], b[4], rsquared)

if plot==True:
    # Plot everything
    fig = plt.figure(figsize = (10, 12))
    ax1 = fig.add_subplot(211)
    pl1 = ax1.plot(theta, torque, 'bs', label='Torque')
    pl2 = ax1.plot(theta, fit, 'r-', label='Fit')
    ax1.set_xlim(0, 2*np.pi)
    box1 = ax1.get_position()
    ax1.set_position([box1.x0, box1.y0, box1.width*0.8, box1.height])
    # Put a legend to the right of the current axis
    ax1.legend(loc='center left', bbox_to_anchor=(1, 0.5))

    ax2 = fig.add_subplot(212)
    pl3 = ax2.plot(np.pi*phi/180, norm(mx), 'bs', label='mx')
    pl4 = ax2.plot(np.pi*phi/180, np.cos(phi*np.pi/180), 'r-',

label=r'$\cos(\phi)$')

ax2.set_xlim(0, 2*np.pi)
ax2.set_ylim(-1.1, 1.1)

# Shrink current axis by 20%
box2 = ax2.get_position()
ax2.set_position([box2.x0, box2.y0, 
    box2.width*0.8, box2.height])

# Put a legend to the right of the current axis
ax2.legend(loc='center left', bbox_to_anchor=(1, 0.5))
plt.show()

return b[2], b[4], rsquared

# Load a file and fit the anisotropies
fname = 'rmoke 100 Oe L 92 400V. txt'
rmoke(fname, H=300, plot=True)
Chapter 4

Exchange Bias in CoO/MgO/Fe/Ag(001)

In the chapter, CoO/MgO/Fe/Ag(001) films were grown epitaxially and studied using Magneto-Optic Kerr Effect and X-ray Magnetic Linear Dichroism (XMLD). For thick CoO films we show that both the Fe coercivity and exchange bias exhibit the expected exponential decrease with increasing MgO thickness. For thin CoO films, however, we find that while the coercivity decreases exponentially, the exchange bias increases initially and then decreases with increasing MgO thickness. By measuring the CoO XMLD as a function of MgO thickness, we show that the unusual behavior of the exchange bias at thinner CoO is due to a transition of the CoO spins from rotatable to frozen spins as the MgO thickness increases.

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4.1 Introduction

Since the discovery of the exchange bias effect in CoO/Co particles [27], Ferromagnet(FM)/Antiferromagnet (AFM) system has attracted great interest because of its application in spintronics technology [28]. Initial understanding of the exchange bias is that as a FM/AFM system is cooled down within a magnetic field to below the Néel temperature of the AFM layer, the FM/AFM interfacial interaction aligns the AFM interfacial spins to the FM spin direction so that the reversal of the FM spins by an external magnetic field has to overcome the FM/AFM interfacial coupling, leading to an exchange bias field that equals to the interfacial coupling. Although correctly catching the role of the FM/AFM interfacial interaction, this intuitive picture gives an exchange bias several orders of magnitude greater than the experimental values.

Two different types of models were proposed to address the above difficulty. The first type assumes a random compensated/uncompensated AFM interface so that the residual interfacial coupling is significantly reduced [29]. The weakness of this model is that the derived exchange bias depends severely on the interfacial roughness and varies from sample to
sample which doesn’t fully agree with experiment. In addition, this model does not address the FM/AFM interfacial coupling for a perfectly compensated AFM surface. Koon finds that a perfectly compensated AFM surface should couple to a FM layer perpendicularly by canting the AFM spins [30]. However, Schulthess and Butler pointed out that Koons model should not lead to an exchange bias but only a uniaxial magnetic anisotropy [31].

Different from the first type model that depends heavily on the realistic interfacial roughness and defects, the second type model (referred to as Mauris model [32] based on an earlier model by Néel[33]) considers domain formation near the FM/AFM interface during the FM magnetization reversal. Mauris model shows that in the strong FM/AFM interfacial coupling limit the exchange bias should correspond to a 180-degree domain wall formation in the AFM layer. While there is experimental evidence of the AFM domain wall formation during the FM magnetization saturation process [34], this model is not valid for ultrathin AFM films that are too thin to host a domain wall. Later development in exchange bias theory is more or less model dependent, aiming to address specific AFM spin configurations or domain structures [35, 36, 37]. Although Mauri’s model does not account directly to ultrathin AFM films, the model does point out an important fact that has been ignored in many theoretical models: one cannot ignore the rotation of the AFM spins in response to the FM magnetization reversal in a FM/AFM system.

Recent development in X-ray Magnetic Linear Dichroism (XMLD) allows a direct measurement of the AFM spins in FM/AFM system. The result shows that there exist rotatable and frozen AFM spins and that these two types of AFM spins play very different roles in the induced exchange bias and magnetic anisotropies [38, 39]. Then taking the interfacial coupling as a phenomenological effective coupling that depends on the interfacial roughness, uncompensated spins, defects, AFM domains, etc., the final exchange bias should be a result of the interplay between the effective interfacial coupling and the AFM frozen spins. To avoid complexity, experimental works usually address the exchange bias by fine-tuning some physical quantities such as the AFM film thickness [40, 41] or a spacer layer thickness between the FM and AFM layers. While the former can certainly change the frozen spins in AFM layer [38] and have been widely applied to study exchange bias, the latter could be used to tune the interfacial coupling strength between the FM and AFM layers. For metallic spacer layer, the exchange bias is found to exhibit an oscillatory, non-monotonic, or an exponential decay with the spacer layer thickness [42, 43, 44, 45, 46, 47]. For insulating spacer layer, the exchange bias has only been reported so far to decay exponentially with the spacer layer thickness [48]. We show that as a result of the exponential decay of the CoO/MgO/Fe interlayer coupling with increasing MgO thickness ($d_{MgO}$), the exchange bias behave very differently for thin and thick CoO films. For thick CoO films, the exchange bias decays exponentially with increasing the MgO thickness. In the ultrathin limit of the CoO thickness ($d_{CoO}$), however, the exchange bias increases initially with increasing MgO thickness before resuming the expected exponential decay. This observation manifests the important role of the CoO frozen spins in generating the exchange bias.
4.2 Experiments

A Ag(001) single crystal substrate was prepared by mechanical polishing down to a 0.25-\(\mu\)m diamond-paste finish, followed by chemical polishing \[49\]. The Ag substrate was cleaned in an UHV system by cycles of Ar+ ion sputtering at \(\sim 2\) keV and annealing at 600\(^{\circ}\)C. A 20-monolayer (ML) Fe film was grown on top of the Ag(001) substrate at room temperature. The film was then annealed at 155\(^{\circ}\)C for about 30 minutes to improve the surface flatness. A MgO wedge (0-4 ML over 3 mm) was then grown on top of the Fe film at room temperature using electron beam evaporation from a MgO target. Then a CoO wedge (0-25 ML over 3 mm) was grown on top of the MgO wedge by deposition of Co under an oxygen pressure of \(2 \times 10^{-6}\) Torr. The MgO and CoO wedges are orthogonal to each other so that their thicknesses can be varied independently along two orthogonal directions on the sample surface. Fe film on Ag(001) has a bcc structure with the Fe[100] axis parallel to the Ag[110] axis, and CoO film on Fe(001) has a rock salt structure with the CoO[110] axis parallel to the Fe[100] axis \[38\]. The sample is finally covered by a 2 nm Ag-protection layer. The formation of the Low Energy Electron Diffraction (LEED) confirms the epitaxial growth of single crystalline CoO/MgO/Fe films on Ag(001) substrate (Fig. 4.1). The sample was measured using Magneto-Optic Kerr Effect (MOKE) and X-ray Magnetic Linear Dichroism (XMLD) at the beamline 4.0.2 of the Advanced Light Source (ALS) of the Lawrence Berkeley National Laboratory. For MOKE measurements, the sample was first cooled down to \(\sim 90\) K from room temperature within a 1000-Oe external magnetic field applied in the Fe in-plane [100] axis. MOKE was used to obtain Fe hysteresis loop of the film. For coercivity greater than 1000 Oe, which is the highest field of our MOKE setup, hysteresis loops were obtained by XMCD measurement at BL 4.0.2 \[38\]. For measurements at BL 4.0.2, the sample was first cooled down to \(\sim 80\) K from room temperature within a 4000-Oe external magnetic field applied in the Fe in-plane [100] axis.

4.3 Results and Discussion

We first present the result of CoO/MgO/Fe/Ag(001) for 20 ML CoO film \(d_{CoO} = 20\) ML. At this thickness, all CoO spins are frozen during the Fe magnetization switching. Fig. 4.2(a) shows representative Fe hysteresis loops at different MgO thicknesses. It is clearly seen that both the coercivity and the exchange bias decrease exponentially with increasing the MgO thickness, showing that the CoO/MgO/Fe interlayer coupling decreases exponentially with increasing the MgO thickness (Fig. 4.2(b)). It has been shown that the imaginary wave vector at the Fermi level in insulators gives rise to an exponential decay of the coupling between two ferromagnetic layers across an insulator \[50, 51\]. This was indeed confirmed in experiments \[52, 53\]. Then it is not surprising that the magnetic interlayer coupling in AFM/insulator/FM system should also decay exponentially with the insulating spacer layer thickness \[48\]. For our sample, the coercivity reached the corresponding bulk Fe value and the exchange bias reached zero at MgO thickness as thin as 4 ML, showing the vanish of
CHAPTER 4. EXCHANGE BIAS IN COO/MGO/FE/AG(001)

Figure 4.1: (a) schematic drawing of the CoO(0-25 ML)/MgO(0-4 ML)/Fe(20 ML)/Ag(001) double wedged sample. The wedge slope is along the Fe[100] axis. LEED patterns of (b) Ag substrate at 123 eV, (c) Fe layer at 158 eV, (d) MgO layer at 149 eV, and (e) CoO layer at 164 eV. The LEED pattern shows the formation of single crystalline growth of each layer.

the interlayer coupling at 4 ML MgO thickness. The small value of 4 ML MgO thickness indicates a high quality of our epitaxial MgO film on Fe(001) (Ref. [54]), and agrees with the interlayer coupling result in Fe/MgO/Fe sandwiches [53].

We now present the result of CoO/MgO/Fe/Ag(001) at $d_{CoO} = 5$ ML [Fig. 4.2(c) and (d)]. Since the interlayer coupling between the CoO and Fe films across the MgO spacer layer should be determined by the MgO thickness; it is easy to understand that the coercivity of this sample also decays exponentially with the MgO thickness, the same behavior as the 20 ML CoO sample. However, as the CoO/MgO/Fe interlayer coupling decreases with increasing the MgO thickness, the exchange bias increases initially to reach a maximum value at $\sim 2$ ML MgO before the exponential decay above 3 ML MgO [Fig. 4.2(d)]. This behavior is unexpected because the exchange bias usually decreases with decreasing the AFM/FM coupling strength.

To ensure the result of Fig. 4.2, we performed a systematic study on the coercivity and exchange bias as a function of the MgO thickness at different CoO thicknesses (Fig. 4.2). Regardless of the initial difference at $d_{MgO} = 0$, the coercivity in CoO/MgO/Fe system (Fig. 4.3(a)) follows the same trend of the exponential decay with increasing MgO thickness for all CoO thicknesses. The exchange bias (Fig. 4.3), on the other hand, exhibits a very
Figure 4.2: (a) Hysteresis loops of CoO(20 ML)/MgO/Fe/Ag(001) at $d_{\text{MgO}} = 0.6$ ML, 1.3 ML, 2.6 ML MgO spacer thicknesses. (b) Coercivity and exchange bias as a function of MgO spacer thickness. Both coercivity and exchange bias decreases exponentially with the MgO thickness, showing that the interlayer coupling across the MgO decreases exponentially with the MgO thickness. (c) Hysteresis loops of CoO(5 ML)/MgO/Fe/Ag(001) at $d_{\text{MgO}} = 0.6$ ML, 1.3 ML, 2.6 ML. (d) Coercivity and exchange bias as a function of the MgO thickness. While the coercivity decreases exponentially, the exchange bias increases initially with increasing MgO thickness before resuming the expected exponential decay at $d_{\text{MgO}} > 3$ ML. The result of Fig. 4.3 indicates that the coercivity depends mostly on the CoO/MgO/Fe interlayer coupling strength, but the exchange bias depends not only on the interlayer coupling but also something else especially in the thinner limit of CoO.
Figure 4.3: (a) Coercivity and (b) exchange bias as a function of MgO thickness at different CoO thicknesses. The coercivity exhibits an exponential decrease with increasing the MgO thickness for all CoO thicknesses. The exchange bias on the other hand shows an unusual behavior for thin CoO thickness, which is attributed to the rotatable-to-frozen transition of the CoO spins with decreasing the CoO/MgO/Fe interlayer coupling. The solid lines are guide to eyes.

In an AFM/FM system, the coercivity enhancement does not require frozen spins in the AFM layer but the exchange bias does. This was confirmed by our previous experiment that the CoO/Fe coercivity enhancement occurs at much thinner CoO thickness than the exchange bias. Therefore the result of Fig. 4.3 shows that the increase of the exchange bias with the MgO thickness in $d_{\text{CoO}} < 15$ ML samples must be associated to the change of the CoO spins (e.g., rotatable/frozen spins) as the MgO thickness increases. As discussed in the introduction, Mauris model addresses the rotation of the AFM spins near the interface. If the AFM spins are completely frozen due to its anisotropy energy, the FM magnetization reversal will have to overcome the AFM/FM interfacial coupling, leading to an exchange bias equal to the interfacial coupling. This is the result of the strong anisotropy limit ($\lambda \ll 1$, where $\lambda$ is the ratio of the interfacial coupling to the AFM domain wall energy) in Mauris model. In contrast, if the AFM/FM interfacial coupling is much stronger than the AFM anisotropy (for example, infinitely strong interfacial coupling), the FM magnetization reversal will force the AFM spins at the AFM/FM interface to twist and thus creating a $180^\circ$ domain wall in the AFM layer. Under this condition, the exchange bias corresponds to the domain wall energy in the AFM layer which is the result of Mauris model in the strong coupling limit ($\lambda \gg 1$).

While Mauris model correctly points out the importance of the AFM spin rotation in response to the FM magnetization reversal in the strong coupling limit, it cannot be applied to ultrathin AFM film where the AFM film thickness is too thin to host a domain wall formation. In this ultrathin limit, all AFM spins should switch to follow the FM magne-
tization reversal, leading to a zero exchange bias (or a weak exchange bias if part of AFM spins is still frozen). Noticing that the rotation of the AFM spins is due to the FM/AFM coupling, thus a weakening of the FM/AFM coupling should favor the formation of frozen spins over rotatable spins in the AFM layer. Then the following physical picture can be applied to qualitatively understand our CoO/MgO/Fe/Ag(001) result. For thick CoO films, where the CoO spins are completely frozen at \(d_{\text{MgO}} = 0\), the decrease of the CoO/MgO/Fe interlayer coupling by increasing the MgO thickness will certainly keep the CoO spins frozen. The exchange bias under this condition should correspond to the strong anisotropy limit in Mauris model \((\lambda \ll 1)\) so that the exchange bias is determined by the CoO/Fe interlayer coupling across the MgO layer. That is why the exchange bias for \(d_{\text{CoO}} > 20\) ML decreases exponentially with increasing the MgO thickness. For thin CoO films, where the CoO spins are totally or partially rotatable at \(d_{\text{MgO}} = 0\), the system is in the strong coupling limit in Mauris model \((\lambda \gg 1)\) so that the exchange bias at \(d_{\text{MgO}} = 0\) should be zero or very weak for the reason discussed above. As the CoO/Fe interlayer coupling decays exponentially to approach zero with increasing MgO thickness, the CoO/MgO/Fe system should evolve from the strong coupling limit \((\lambda \gg 1)\) at \(d_{\text{MgO}} = 0\) to the weak coupling limit \((\lambda \ll 1)\) at thicker MgO. Consequently, the CoO spins should evolve from rotatable to frozen as the CoO/MgO/Fe interlayer coupling becomes weaker and weaker. Since the exchange bias is zero or very weak in the strong coupling limit \((\lambda \gg 1)\) and equals to the AFM/FM coupling in the weak coupling limit \((\lambda \ll 1)\), the exchange bias in the CoO/MgO/Fe system in ultra-thin CoO limit should increases initially and then decreases as the MgO thickness increases, which is what we observed for \(d_{\text{CoO}} < 15\) ML in Fig. 4.3. The above physical picture implies a transition of the CoO spins from rotatable to frozen as the MgO thickness increases for \(d_{\text{CoO}} < 15\) ML.

To verify this transition, we performed XMLD measurement at \(d_{\text{CoO}} = 5\) ML where the CoO spins are totally rotatable at \(d_{\text{MgO}} = 0\) within the experimental sensitivity [38]. The CoO \(L_3\) edge absorption spectra are taken for magnetic field parallel and perpendicular to the field cooling direction (Fig. 4.4(a)). The difference between these two spectra then measures the amount of rotatable CoO spins. Detailed experimental procedure is described in Ref. [38]. Fig. 4.4(b)-(e) shows two representative spectra at different MgO thicknesses. At \(d_{\text{MgO}} = 0.13\) ML, the change of the spectra for field parallel and perpendicular to the field cooling direction corresponds to a complete rotatable CoO spins [38]. At \(d_{\text{MgO}} = 3.5\) ML, there is no difference between the spectra for field parallel and perpendicular to the field cooling direction, showing that all the CoO spins are frozen. Using the conventional definition of the XMLD, we retrieve the amount of CoO frozen spins as a function of the MgO thickness in the 5 ML CoO sample. The result shows that the CoO spins becomes frozen gradually as the MgO thickness increases and become completely frozen above 3 ML of MgO (Fig. 4.5). We would like to mention that the result of Fig. 4.5 should be an approximation because it is unclear if the rotatable CoO spins will rotate by 90 degrees or less after the magnetic field rotates by 90 degrees. A reliable theoretical model is needed from the community for the description of exchange bias for partially frozen AFM spins.

We would like to discuss other possible mechanisms that could affect the exchange bias.
Figure 4.4: (a) XMLD measurement condition. (b) and (d) are the Co L3 edge X-ray absorption spectra at $d_{MgO} = 0.13$ ML and 3.5 ML, respectively, for magnetic field applied parallel (red) and perpendicular (black) to the field-cooling direction. (c) and (e) are the differences of the spectra in (b) and (d) respectively, corresponding to the rotatable CoO spins.

First, it is shown that the exchange bias in the CoO/Fe system is not directly proportional to the CoO frozen spins.\cite{38, 39} Instead, there is a ‘thickness delay’ between the exchange bias and the CoO frozen spins. However, the increase in the CoO frozen spins should not decrease the exchange bias provided that other conditions are fixed. In our previous papers, the amount of CoO frozen spins is increased by increasing the CoO thickness and, thus,
cannot isolate the effect of CoO frozen spins from the CoO thickness. In the present paper, the amount of CoO frozen spins is increased without changing the CoO thickness, thus, providing a cleaner isolation of the CoO frozen spins from its film thickness. On the other hand, because of the lack of a direct proportionality, it is difficult to perform a quantitative analysis for Fig. 4.3(d) in terms of the result of Fig. 4.5. Second, although the MgO energy gap is stabilized at its bulk value at ~3 ML thickness in MgO/Fe(001)\cite{54}, the electronic structure of MgO could be varied below 3 ML thickness. Then, the interesting question is if the nonmonotonic behavior in $H_{ex} - d_{MgO}$ could be due to the MgO electronic variation below 3 ML, especially, a possible metallic MgO electronic structure? We think this is unlikely because the insulating behavior of MgO was actually observed even at 1 ML MgO, although the energy gap may be different from its bulk value. In addition, we observe a monotonic $H_{ex} - d_{MgO}$ relation for a thicker CoO film where it is the same MgO film that leads to the nonmonotonic $H_{ex} - d_{MgO}$ relation at a thinner CoO film. Last, it is well known that film and interfacial roughnesses could affect the exchange bias\cite{55}. Then, it is reasonable to ask if the variation in the exchange bias at the monolayer regime of MgO thickness could be due to the MgO film roughness and the MgO/CoO interfacial roughness? Note that our sample is CoO/MgO/Fe/Ag(001), and there should be the same MgO film roughness and the same MgO/Fe and CoO/MgO interfacial roughnesses for all CoO thicknesses. Therefore,
although the film roughness plays a role in determining the absolute exchange bias value, it is unlikely that it is responsible for the different $H_{ex} - d_{MgO}$ behaviors at different CoO thicknesses.

### 4.4 Summary

In summary, we investigated the exchange bias in epitaxial CoO/MgO/Fe/Ag(001) films. We find that above 20 ML CoO thickness both the Fe coercivity and the exchange bias decreases exponentially with increasing the MgO thickness, showing that the interlayer coupling between CoO and Fe films decreases exponentially with the MgO thickness. Below 17.5 ML CoO thickness, the exchange bias increases initially and then decreases with increasing the MgO thickness. This unusual behavior is explained by a transition of the CoO spins from rotatable to frozen spins as the CoO/Fe interlayer coupling decreases with increasing the MgO thickness. This rotatable-to-frozen transition of the CoO spins with increasing the MgO thickness is confirmed by XMLD measurement.
Chapter 5

Magnetic Anisotropy in Py/FeMn/Ni/Cu(001)

Py/FeMn/Cu(001) and Py/FeMn/Ni/Cu(001) films were grown and studied as a function of both the FeMn and the Ni thicknesses using Rotating Magneto-Optic Kerr Effect (ROTMOKE) and X-ray Magnetic Circular Dichroism (XMCD). For Py/FeMn/Cu(001), we find that the FeMn antiferromagnetic(AFM) order switches the sign of the Py 4-fold magnetic anisotropy but has little effect on the step-induced uniaxial magnetic anisotropy. For Py/FeMn/Ni/Cu(001), we find that out-of-plane Ni magnetization has little effect on the Py magnetic anisotropy but in-plane Ni magnetization enhances the Py magnetic anisotropy in the region just above antiferromagnetic transition thickness. The underlying mechanism could be attributed to the FeMn 3Q spin structure.

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5.1 Introduction

Antiferromagnetic(AFM)/ferromagnetic(FM) bilayer system has been studied extensively for several decades because of its fascinating property (e.g. exchange bias) and its application in magnetic devices. [27, 56] Despite the renewed interest in recent years on the AFM/FM system, an understanding of the AFM/FM interfacial interaction has remained far from complete. It is now well recognized that the original model of a complete uncompensated AFM/FM interface is overly simplified and the theoretical value of the AFM/FM interfacial coupling misses the experimental value by orders of magnitude. In order to address a realistic experimental systems, various models have been proposed with different underlying mechanisms [29, 30, 31, 32, 35, 36, 37]. The main difficulty in addressing the AFM/FM interfacial interaction is the inevitable spin frustration at the interface which depends on the details of the interfacial roughness and the AFM spin configurations.

The main theme of the investigation in experiments is to first obtain the phenomenological values of the AFM induced magnetic anisotropy in the FM layer, and then to make efforts
to reduce the complexity of the interfacial frustration. Along this direction, single crystalline FeMn/Co bilayers grown on Cu(001) have become one model system because of the epitaxial growth nature and the special FeMn spin structure. The FeMn has a non-collinear 3Q spin structure which consists of four sublattices with the spins pointing to four different [111] directions [57]. This spin structure gives rise to a complete compensated (001) surface and is ideal for the study of AFM/FM interfacial interaction in epitaxially grown thin films [58]. For realistic (001) surface with random atomic steps, the 3Q spin structure will lead to uncompensated spins only at the [100]-type steps. Then as these uncompensated moments interact with the FM layer at the AFM/FM interface (e.g., in FeMn/Co system), the interfacial coupling will result in an easy magnetization direction in the four equivalent [100] directions within the film plane. This fact is indeed confirmed in experiment by the switching of the Co easy magnetization direction from [110] to [100] axes as the FeMn establishes its AFM order [59]. Identifying the relevant atomic steps to only [100]-type steps in FeMn/Co(001) system is a significant step forward in reducing the complexity of interface interaction, considering the step-induced magnetic frustration is still inevitable in epitaxial FeMn films. Although we cannot completely eliminate this effect, we can keep this variable constant by keeping the interface identical. An example is the investigation of Co/Ni/FeMn/Cu(001) system where adding Co layer on top of Ni changes the magnetization from out-of-plane to in-plane direction without changing the Ni/FeMn interfacial roughness. It was found that the FeMn Néel temperature will be changed with the Co/Ni spin reorientation transition (SRT) [60]. The significance of this approach is that we are able to tune the spin configuration at the AFM/FM interface without changing the interfacial roughness and hence we can isolate the effect of FM spin configurations on the interface.

Regardless of the detailed spin configuration at the AFM/FM interface, the phenomenological effect of the AFM/FM interfacial interaction is to induce additional magnetic anisotropy terms to the FM layer. In fact, the AFM order induces not only the exchange bias but various types of magnetic anisotropies in the FM layer [39]. It was found recently that the AFM order of the AFM layer could even change the perpendicular magnetic anisotropy of the FM overlayer to result in a change of the FM SRT in FeMn/Ni/Cu(001) (Ref. [61]) and Fe/Mn/Cu$_3$Au(001) (Ref. [62]) systems. Then with the experimental approaches mentioned in the last paragraph, it is interesting to ask if the magnetic state in one FM layer of a FM/AFM/FM sandwich could change the AFM spin configuration such that it influences the magnetic anisotropies of the other FM layer without changing the film roughness. It was found that the exchange biases of two FM layers in FM/AFM/FM sandwiches are indeed correlated [63]. Yang and Chien claimed a spiral structure of the FeMn spins in Py/FeMn(111)/Co when oppositely biased with the spiral length $\sim$9 nm. [64] Nam et al. also reported that the exchange bias of the bottom and top CoFe layers in CoFe/FeMn(111)/CoFe trilayers are mediated by the FeMn layer in 5-10 nm range. [65] The recent work by Kim et al. [66] also proves that the magnetization reversals of Py and CoFe layers in Py/FeMn-/CoFe trilayers are coupled up to 30 nm FeMn. These results indicate that, albeit indirectly, one FM layer in the FM/AFM/FM could have an influence on the magnetic property of the other FM through modification of the spin structure of the AFM layer. All previous works
CHAPTER 5. MAGNETIC ANISOTROPY IN PY/FEMN/NI/CU(001)

mentioned were done on textured or polycrystalline FeMn films and are in the thicker regime (> 5 nm) so that spin twisting of the FeMn layer could be formed by creating domain-like spirals; which obscures the effect of the FM on the spin structure of the FeMn layer.

With this motivation, we carried out an investigation on epitaxially grown single crystalline Py/FeMn/Ni/Cu(001) system. We chose this system for the following reasons. First, Py/FeMn/Ni sandwich grows epitaxially on Cu(001) because of the lattice matching so that the advantage of FeMn 3Q spin structure mentioned above could be applied to this system. Second, Ni/Cu(001) undergoes a SRT from in-plane to out-plane direction with increasing the Ni film thickness so that the effect of the Ni spin direction on the Py/FeMn magnetic anisotropy in the Py/FeMn/Ni/Cu(001) system could be studied. Finally, the weak magnetic anisotropy of the permalloy film make it easy to identify any AFM/FM interaction-induced magnetic anisotropy in the Py film. We find that different Ni spin directions have different effects on the magnetic anisotropy of the Py film in Py/FeMn/Ni/Cu(001).

5.2 Experiment

![LEED patterns](image)

Figure 5.1: LEED patterns of (a) Cu(001) substrate at 130 eV, (b) 10 ML Ni layer at 130 eV, (d) 10 ML FeMn layer at 128 eV, and (d) 15 ML Py overlayer at 127 eV.

A 10-mm diameter Cu(001) single crystals was mechanically polished down to 0.25µm diamond paste finish, and was further electrochemically polished in a mixture of liquid of 75 % phosphoric acid, 10% water, and 10% sulfuric acid while applying a constant voltage of 1.8V for 20 seconds. The Cu(001) substrate was then cleaned in an ultrahigh vacuum
(UHV) chamber by cycles of Ar+ sputtering at $\sim 2$ keV and annealing at $\sim 600^\circ$C. All the thin films are prepared at room temperature by Molecular Beam Epitaxy (MBE) using thermal evaporation of pure metals (99.999%). A Ni wedge of 0-20 monolayers (ML) was grown on top of the Cu(001) substrate. An FeMn wedge (0-20 ML) was then grown by co-evaporating Fe and Mn with 1:1 evaporation rate ratio. The FeMn wedge direction is orthogonal to the Ni wedge so that the FeMn and Ni thicknesses can be varied independently along two orthogonal directions within the sample surface. A permalloy (Py) layer of 15 ML was then deposited over the entire surface by co-evaporating Fe and Ni at 1:4 evaporation rate ratio. The sample is finally covered by an 11 ML Cu protection layer. The formation of the Low Energy Electron Diffraction (LEED) confirms the epitaxial growth of single crystalline Py/FeMn/Ni films on Cu(001) substrate (Fig. 5.1).

Magnetic anisotropy of the sample was measured by Rotating Magneto-Optic Kerr Effect (ROTMOKE). Two pairs of electromagnets were used to supply a rotating magnetic field of constant magnitude within the film plane without the need of a physical rotation of the magnet. In this way, any slow drift due to environmental fluctuations can be eliminated with the fast rotation of the magnetic field. Detailed design and operation of our ROTMOKE setup is described in our previous paper [67]. Element-specific magnetic circular dichroism (XMCD) measurement was also measured at beamline 6.3.1 of the Advanced Light Source (ALS) of the Lawrence Berkeley National Laboratory.

5.3 Result and Discussion

5.3.1 Determination of the Py and Ni Spin Directions from MOKE and XMCD

It is well known that Py film has an in-plane magnetization due to its shape anisotropy but Ni/Cu(001) has an in-plane to perpendicular SRT with increasing Ni thickness ($d_{Ni}$) [68]. The specific SRT thickness depends on the overlayer materials (e.g. vacuum, Cu, or FeMn). The paramagnetic, in-plane, and out-of-plane Ni phase diagram has already been constructed in our previous work [61]. Then for Py/FeMn/Ni/Cu(001) sample, it is trivial to conclude that both Py and Ni films should have in-plane magnetizations when FeMn/Ni/Cu(001) has in-plane Ni magnetization. For perpendicular Ni magnetization in FeMn/Ni/Cu(001), however, it is not trivial to claim a perpendicular Ni or in-plane Py magnetizations because of the magnetic coupling between Py and Ni across the FeMn.

Fig. 5.2(a) and (b) show a series of longitudinal and polar MOKE hysteresis loops of Py(15ML)/FeMn/Ni(18ML)/Cu(001) at different FeMn thickness ($d_{FeMn}$). 18 ML Ni alone should favor a perpendicular magnetization in FeMn/Ni(18ML)/Cu(001) [61]. With increasing FeMn thickness, the hysteresis loops undergo two transitions. The first transition occurs at $d_{FeMn} \sim 5$ ML. At $d_{FeMn} < 4$ ML, the longitudinal remanence is almost a constant with the polar loop being a hard axis loop. At $d_{FeMn} \sim 5$ ML, the longitudinal Kerr signal drops and then becomes a constant at $d_{FeMn} > 6$ ML with a lower value than that at
Figure 5.2: Hysteresis loops of Py(15ML)/FeMn/Ni(18ML)/Cu(001) at different FeMn thicknesses. Loops in rows (a) and (b) are collected using Magneto-Optical Kerr Effect in (a) longitudinal and (b) polar configurations respectively. Loops in rows (c) and (d) are collected using XMCD effect at Fe L3 peak for (c) longitudinal and (d) polar configurations respectively. The XMCD loops represent the Py hysteresis loops.

$d_{FeMn} < 4$ ML, while the polar loop becomes an easy-axis loop. The second transition occurs at $d_{FeMn} \sim 10$ ML and is characterized by a coercivity ($H_C$) increase. The coercivity increase is due to the AFM order of the FeMn film which we will discuss later. Now we focus on the first transition at $d_{FeMn} \sim 5$ ML. Since the Py magnetization has an in-plane anisotropy, the higher value of remanence at $d_{FeMn} < 4$ ML than that at $d_{FeMn} > 6$ ML suggests that the Ni magnetization has an almost in-plane magnetization at $d_{FeMn} < 4$ ML due to its strong coupling with the Py magnetization. This conclusion is supported by the polar MOKE result which shows a hard axis loop at $d_{FeMn} < 4$ ML, and an easy axis loop at $d_{FeMn} > 6$ ML.

Fig. 5.3 shows the MOKE polar remanence of Py(15ML)/FeMn/Ni(18ML)/Cu(001) as a function of FeMn thickness. An in-plane to out-of-plane SRT could be clearly seen at $d_{FeMn} \sim 5$ ML. However, the in-plane orientation is not perfect because we still observe a non-zero polar remanence at $d_{FeMn} < 5$ ML, indicating an out-of-plane tilting component of the magnetization from the sample. Since MOKE measures the total magnetization of
Figure 5.3: Polar MOKE remanence and polar Py saturation filed of Py(15ML)/FeMn/Ni(18 ML)/Cu(001) as a function of FeMn thickness. Both curves shows a pseudo spin reorientation transition at $d_{FeMn} \sim 5$ ML. The Ni magnetization is forced to be more in-plane by its strong coupling to the in-plane Py at $d_{FeMn} < 5$ ML and switches to out-of-plane direction as its coupling to the Py becomes weaker at $d_{FeMn} > 5$ ML.

the film and the Kerr signals from Py and Ni have different magnitude, it is difficult to distinguish the Py and Ni magnetization directions from the MOKE measurement alone. Then we measured XMCD hysteresis loops to further distinguish the Py and the Ni magnetizations. Because Ni is present in both Py and Ni films, we measured the Fe XMCD to obtain Py hysteresis loops. Fig. 5.2(c) and (d) shows the longitudinal and polar hysteresis loops from the Fe edge XMCD. The Py longitudinal loop always exhibits an easy-axis loop and the Py polar hysteresis loop always exhibits a hard axis loop with a very low remanence, showing that the Py magnetization has an in-plane easy axis throughout the FeMn thickness range. The small but non-zero remanence of the Py polar loop (10% of the saturation value) could be either due to a misalignment of the sample relative to the x-ray incident direction (so that the polar measurement may have picked up some longitudinal component), or due to a small out-of-plane tilting of the Py magnetization. Assuming the latter case, we estimate the upper limit of the out-of-plane tilting angle of Py magnetization to be less than 6 degrees. Therefore we can conclude safely that the Py has
an in-plane magnetization in the \( \text{Py}(15\text{ML})/\text{FeMn/\text{Ni}}(18\text{ML})/\text{Cu}(001) \) in the whole FeMn thickness range. Then the polar loop at \( d_{\text{FeMn}} > 6 \text{ ML} \) in Fig. 5.2(a) must indicate an out-of-plane Ni magnetization, and that the non-zero polar MOKE remanence at \( d_{\text{FeMn}} < 4 \text{ ML} \) must be due to an out-of-plane tilting of the Ni magnetization. Because the 18 ML Ni film alone favors a perpendicular magnetic anisotropy, the interlayer coupling between Py and Ni in \( \text{Py}(15\text{ML})/\text{FeMn/\text{Ni}}(18\text{ML})/\text{Cu}(001) \) must make it easier to saturate the Py magnetization in the perpendicular direction when Ni magnetization is in-plane (indicating stronger coupling with Py) at \( d_{\text{FeMn}} < 5 \) than when Ni magnetization is out-of-plane (indicating weaker coupling) at \( d_{\text{FeMn}} > 5 \text{ ML} \). The Py polar loop saturation field from the Fe XMCD polar loop is also plotted in Fig. 5.3 to compare with the polar MOKE remanence result. Both quantities in Fig. 5.3 indicates the same fact that the Ni film in \( \text{Py}(15\text{ML})/\text{FeMn/\text{Ni}}(18\text{ML})/\text{Cu}(001) \) undergoes a pseudo SRT from out-of-plane direction at \( d_{\text{FeMn}} > 5 \text{ ML} \) to an almost in-plane direction at \( d_{\text{FeMn}} < 5 \text{ ML} \). Then for the study of the effect of perpendicular Ni on the Py magnetic anisotropy in section 5.3.3, we should focus on the \( d_{\text{FeMn}} > 5 \text{ ML} \) regime, as otherwise the Ni tilting magnetization could mislead the interpretation.

### 5.3.2 Magnetic Anisotropy of \( \text{Py/FeMn/Cu}(001) \)

The coercivity \( (H_C) \) of \( \text{Py}(15\text{ML})/\text{FeMn/Cu}(001) \) is small at \( d_{\text{FeMn}} < 10 \text{ ML} \) and then increases at \( d_{\text{FeMn}} \sim 10 \text{ ML} \) to reach a peak value at \( d_{\text{FeMn}} \sim 12 \text{ ML} \), and then become constant at \( d_{\text{FeMn}} > 15 \text{ ML} \). This phenomenon has been reported in literature and has been attributed to the AFM order induced magnetic anisotropy by the FeMn layer to the FM layer, i.e., the FeMn film at room temperature is paramagnetic at \( d_{\text{FeMn}} < 10 \text{ ML} \) and establishes the AFM order at \( d_{\text{FeMn}} > 10 \text{ ML} \). The peak behavior in \( H_C \) was also reported in other AFM/FM systems such as CoO/Fe/Ag(001). To understand the behavior quantitatively, we performed ROTMOKE measurement on \( \text{Py}(15\text{ML})/\text{FeMn/Cu}(001) \) to determine the Py magnetic anisotropy as a function of the FeMn thickness. The schematic drawing of our ROTMOKE experiment is shown in Fig. 5.4(a). The ROTMOKE optical plane is set in such a way that the Kerr signal measures the projection of the in-plane magnetization along the Py [100] axis as a 700 Oe magnetic field rotates the magnetization within the film plane. Then the difference between the field angle and the magnetization angle relative to the [100] axis reveals information on the magnetic anisotropy of the Py film. Specifically, the equilibrium condition is reached by minimizing the energy density of the Py film.

\[
E = -M_S H_{\text{ROT}} \cos(\theta_H - \theta_M) + K_2 \sin^2(\theta_M) + K_4 \sin^2(\theta_M) \cos^2(\theta_M)
\]  

(5.1)

Here \( H_{\text{ROT}} \) is the strength of the rotating magnetic field, \( K_4 \) is the 4-fold anisotropy, and \( \theta_H \) and \( \theta_M \) are the angles of the magnetic field and the Py magnetization relative to the [100] axis, respectively. We also include a uniaxial anisotropy \( K_2 \) in case of possible mechanism that breaks the 4-fold rotation symmetry of (001) film (e.g., vicinal surface). We didn’t include the exchange bias because we did not perform field cooling to
the film. Minimizing Eqn. (5.1) with respect to $\theta_M$ yields the magnetic torque equilibrium condition:

$$ l(\theta_M) \equiv H_{\text{ROT}} \sin(\theta_H - \theta_M) = \frac{1}{2} H_2 \sin(2\theta_M) + \frac{1}{4} H_4 \sin(4\theta_M) $$  \hspace{1cm} (5.2)

Here the anisotropic fields are defined as $H_2 = 2K_2/M_S$ and $H_4 = 2K_4/M_S$. In this paper, we follow the convention to use the anisotropic field values to indicate the uniaxial and 4-fold anisotropies. Because $\theta_H$ and $H_{\text{ROT}} = 700$ Oe are given in experiment and $\theta_M$ (or precisely $\cos\theta_M$) is directly measured by ROTMOKE, the magnetic torque $l(\theta_M) \equiv H_{\text{ROT}} \sin(\theta_H - \theta_M)$ can then be determined from experiment as a function of $\theta_H$. A fit of $l(\theta_M)$ using Eqn. (5.2) will yield the anisotropy constants of $H_2 = 2K_2/M_S$ and $H_4 = 2K_4/M_S$. This is the basic principle of ROTMOKE and has been widely used to determine the magnetic anisotropy of magnetic thin films [67].

Fig. 5.4(b) shows a series of $l(\theta_M)$ vs $\theta_M$ curves. We notice that $H_4$ alone wont fit our experimental curve. In order to fit the experimental curve, we have to include a small but non-zero uniaxial anisotropy term of $H_2$ in Eqn. (5.2). Then the experimental curves can be well fit by Eqn. (2) for all FeMn thicknesses, (red lines) with coefficient of determination (R$^2$ value) better than 0.95 for all data presented. The derived $H_2$ and $H_4$ from the fitting are shown in Fig. 5.5(a) together with the $H_C$. As the FeMn thickness increases, the most dramatic change is that $H_4$ switches its sign from $H_4 < 0$ at $d_{\text{FeMn}} < 10$ ML to $H_4 > 0$ at $d_{\text{FeMn}} > 10$ ML. This sudden change corresponds to the $H_C$ behavior, supporting the literature conclusion that the $H_C$ enhancement is due to the AFM induced 4-fold magnetic anisotropy in the FM layer. Note that $\theta_M$ is the angle between the magnetization and the [100] axis, thus the sign change of from $H_4$ shows that the easy magnetization axis of the Py 4-fold anisotropy changes from [110] at $d_{\text{FeMn}} < 10$ ML to [100] at $d_{\text{FeMn}} > 10$ ML. This result is the same as the result in FeMn/Co/Cu(001) system, confirming that the anisotropy change is due to the FeMn AFM order. In fact, thickness-dependent study in FeMn/Co/Cu(001) system indicates that the FeMn induced 4-fold anisotropy is indeed originated from the FeMn/Co interfacial interaction [71]. Compared to the dramatic change in $H_C$ and $H_4$, the $H_2$ change is less dramatic at $d_{\text{FeMn}} = 10$ ML. The presence of $H_2$ at $d_{\text{FeMn}} < 10$ ML shows that the uniaxial anisotropy is not originated from the FeMn AFM order. Then it is must be associated with the vicinal angle of the Cu(001) substrate. It was shown that vicinal surface on Cu(001) could induce a uniaxial anisotropy to a Co overlayer with $\sim 50$ Oe per degree of vicinal angle [70]. Then it is possible that the observed small uniaxial magnetic anisotropy ($\leq 50$ Oe) in Py/FeMn/Cu(001) is generated by a small vicinal angle ($\leq 1$ degree) of our Cu(001) substrate, which is very possible by a misalignment in substrate polishing. The $H_2$ only changes its magnitude slightly at $d_{\text{FeMn}} \sim 10$ ML as compared to the change of $H_4$, showing that the AFM order of the FeMn has little effect on the step-induced uniaxial anisotropy.

The FeMn induced magnetic anisotropy can be explained using the FeMn 3Q spin structure as shown in Fig. 5.5(b). Among the possible [100] and [110] steps on a FeMn(001) surface, the in-plane projection of spins has uncompensated spins only at [100]-type steps
from the top terrace at the [100] step edge. The uncompensated spin direction could take one of the [110], [110], [110], and [110] for [100] steps (or for [010] steps). Then as a FM overlayer is grown on top of the FeMn(001) surface, the coupling between FM and the uncompensated FeMn spins is equivalent to a random field applied along the four [110], [110], [110], and [110] directions in space so that the AFM/FM interfacial interaction will lead to a 4-fold magnetic anisotropy which favors the magnetization 45° from these four axes (e.g. magnetic easy axes in [100] and [010] axes). That is why the FeMn AFM order switches the Py 4-fold anisotropy from $H_4 < 0$ at $d_{FeMn} < 10$ ML to $H_4 > 0$ at $d_{FeMn} > 10$ ML. Since the FeMn uncompensated spins have equal probability in [110], [110], [110], and [110] directions for [100] steps, the FeMn uncompensated spins for [100] (or [010]) vicinal surface won’t induce uniaxial magnetic anisotropy. That explains why we don’t see a dramatic change of $H_2$ at $d_{FeMn} = 10$ ML. In another word, the small change of $H_2$ at $d_{FeMn} = 10$ ML is more likely associated with the electronic state change that modifies the Néel’s pair bonding strength at the AFM ordering point rather than due to the FeMn 3Q spin structure. Within Néel’s pair bond model, a Py atom at the Py/FeMn vicinal interface will have some of its Py-Py bonds replaced by Py-FeMn bonds which break the symmetry and induce a uniaxial magnetic anisotropy. The strength of $H_2$ is determined by the difference between the magnetic energy of Py atoms in a bulk site and the Py atoms at Py-FeMn interface, in particular those at the step-edge or step-corner sites.72 During the FeMn phase transition from PM to AFM, it is possible that the Py-FeMn bonds at the step-edge sites and/or step-corner sites at the Py/FeMn interface would experience a change in bond strength due to electronic state change of the FeMn at the Néel transition, akin to the vanishing exchange splitting as defined by Stoner’s model at the Curie temperature of an itinerant ferromagnet.73,74 Our measurements of $H_2$ in Py/FeMn/Ni/Cu(001) also show little change on $H_2$ compared to $H_4$ in the AFM regime of FeMn. In this paper, we will focus the discussion on $H_4$, and we will only give a brief discussion on $H_2$. 
Figure 5.4: (a) Schematic of the ROTMOKE experiments. The optics are set such that the Kerr signal measures the projection of magnetization in the [100] direction ($\approx \cos \theta_M$). Figure (b)-(g) are the magnetic torque [$l(\theta_M) \equiv H_{ROT} \sin (\theta_H - \theta_M)$] as a function of the magnetization angle ($\theta_M$) for Py(15ML)/FeMn/Cu(001) at (b) $d_{FeMn} = 6$ ML, (c) 8 ML, (d) 10 ML, (e) 12 ML, (f) 14 ML and (g) 16 ML. The red lines are theoretical fittings using Eqn. (5.2).
Figure 5.5: (a) $H_C$, $H_2$, and $H_4$ as a function of FeMn thickness for Py(15ML)/FeMn/Cu(001). (b) Schematic drawing of the in-plane component of FeMn 3Q spin-structure on (001) surface. Red and blue arrows are in-plane projection of spins from different layers. Black dashed lines are [100] steps. The circled spins are uncompensated spins as a result of the [100] steps. The inset in figure (b) shows the three dimensional drawing of the 3Q structure.
5.3.3 Magnetic Anisotropy of Py/FeMn/Ni/Cu(001)

We now discuss the result of Py/FeMn/Ni/Cu(001). Since we already know the Ni magnetization direction in Py/FeMn/Ni/Cu(001), we first studied Py/FeMn/Ni/Cu(001) at $d_{Ni} = 2$ ML, 10 ML, and 18 ML to show the effect of paramagnetic, in-plane, and out-of-plane Ni magnetization on the Py magnetic anisotropy. Fig. 5.6(a) shows the ROTMOKE result as a function of FeMn thickness at these three different Ni thicknesses. As compared to the $d_{Ni} = 2$ ML and 18 ML samples, there is an obvious difference for in-plane Ni ($d_{Ni} = 10$ ML) in the region where the FeMn establishes its AFM order ($d_{FeMn} \sim 12$ ML). The derived $H_4$ vs $d_{FeMn}$ is shown in Fig. 5.6(b). The onset of the $H_4$ enhancement in $d_{Ni} = 10$ ML and 18 ML samples is slightly thinner in FeMn thickness than in $d_{Ni} = 2$ ML sample. Although there is report on the effect of the Ni spin orientation on the FeMn Néel temperature [60], the change in the onset FeMn thickness is too small for us to claim the Néel temperature change in our sample. Therefore we here focus on the $H_4$ discussion only. First, we find that the $H_4$ values for paramagnetic Ni ($d_{Ni} = 2$ ML) and perpendicular Ni ($d_{Ni} = 18$ ML) are virtually the same in Py/FeMn/Ni/Cu(001), showing that the perpendicular Ni has little (or no) effect on the Py 4-fold anisotropy. For in-plane Ni ($d_{Ni} = 10$ ML), the $H_4$ in Py/FeMn/Ni/Cu(001) increases as that for paramagnetic Ni ($d_{Ni} = 2$ ML) and perpendicular Ni ($d_{Ni} = 18$ ML) but only in the region of $10$ ML $< d_{FeMn} < 15$ ML where the FeMn just becomes AFM ordered (just above the transition thickness). The small difference of the $H_4$ values of these three samples at $d_{FeMn} > 15$ M is more likely associated with the sample inhomogeneity.

To ensure the result of Fig. 5.6(b), we also measured $H_4$ of Py/FeMn/Ni/Cu(001) as a function of Ni thickness at fixed $d_{FeMn} = 8$, 12, and 18 ML, corresponding to paramagnetic FeMn, just-ordered FeMn, and well-ordered FeMn (Fig. 5.7). The $H_4$ of Py/FeMn/Ni/Cu(001) has little dependence on the Ni thickness for $d_{FeMn} = 8$ ML and 18 ML, showing that the Ni layer in Py/FeMn/Ni/Cu(001) has little effect on the 4-fold anisotropy of Py for paramagnetic and well-ordered FeMn. At $d_{FeMn} = 12$ ML where the AFM order of FeMn has just been established, the $H_4$ increases as the Ni changes from paramagnetic to in-plane ferromagnetic at $d_{Ni} \sim 5$ ML and then drops after the Ni changes from in-plane to out-of-plane at $d_{Ni} \sim 15$ ML. Therefore the result of Fig. 5.7 supports the result of Fig 5.6(b) that only in-plane Ni has an effect to enhance the Py 4-fold anisotropy in Py/FeMn/Ni/Cu(001) but only in the region where the FeMn just becomes AFM ordered.

Although wedged sample provides a systematic variation of film thickness, it has inherent inhomogeneity problem due to inhomogeneity of the substrate polishing (e.g., scratches, vicinal surface, etc). This could give abnormal data points in a single line scan on wedged sample. To ensure that the result of Fig. 5.6(b) and Fig. 5.7 are not associated with any defects or scratches of the substrate, we measured the $H_4$ of Py/FeMn/Ni/Cu(001) in the entire FeMn-Ni thickness plane using ROTMOKE. This tremendous amount of measurement ($\sim 1000$ ROTMOKE curves) allows us to construct a map of the $H_4$ in the FeMn-Ni thickness plane for Py(15ML)/FeMn/Ni/Cu(001). The result (Fig. 5.8) shows an obvious enhancement of $H_4$ for in-plane Ni ($5$ ML $< d_{Ni} < 15$ ML) in the region where the FeMn
Figure 5.6: Magnetic torque versus magnetization angle for (a)-(c) paramagnetic Ni \((d_{Ni} = 2\) ML), (d)-(f) in-plane ferromagnetic Ni \((d_{Ni} = 10\) ML), and (g)-(i) out-of-plane ferromagnetic Ni \((d_{Ni} = 18\) ML). (a), (d) and (g) are taken at paramagnetic FeMn \((d_{FeMn} = 8\) ML); (b), (e) and (h) are taken at \(d_{FeMn} = 12\) ML where the FeMn is just AFM ordered; and (c), (f) and (i) are taken at \(d_{FeMn} = 16\) ML where the FeMn is well AFM ordered. The anisotropy value \((H_4)\) from the fitting is shown in (j). The \(H_4\) is enhanced for in-plane Ni in the range of \(10\) ML < \(d_{FeMn}\) < 15 ML.

just becomes AFM ordered \((10\) ML < \(d_{FeMn}\) < 15 ML). There is also a significant change of \(H_4\) below 3 ML FeMn in the ferromagnetic regime of Ni \((d_{Ni} > 5\) ML and \(d_{FeMn} < 3\) ML). However, as discussed in section 5.3.1, the Py and Ni layers are strongly coupled in this region so that it doesn’t make too much sense to discuss the Ni spin orientation effect on the Py magnetic anisotropy in this regime.
Figure 5.7: $H_4$ of Py(15ML)/FeMn/Ni/Cu(001) as a function of Ni thickness at paramagnetic ($d_{FeMn} = 8$ ML), just ordered ($d_{FeMn} = 12$ ML), and well ordered ($d_{FeMn} = 18$ ML) FeMn film. While $H_4$ has little change in the $d_{FeMn} = 8$ ML and 18 ML samples, $H_4$ is enhanced by in-plane Ni magnetization ($5$ ML $< d_{Ni} < 15$ ML) in the $d_{FeMn} = 12$ ML sample.
Figure 5.8: Map of (a) $H_4$ and (b) $H_2$ of Py(15ML)/FeMn/Ni/Cu(001) as a function of both Ni and FeMn thicknesses. In the PM-AFM transition region (10 ML < $d_{FeMn}$ < 15 ML), only in-plane Ni magnetization (5 ML < $d_{Ni}$ < 15 ML) enhances the $H_4$ value as compared to Py(15ML)/FeMn/Cu(001).

The above result shows that perpendicular Ni in Py/FeMn/Ni/Cu(001) has no effect on the FeMn-induced magnetic anisotropy in the Py film. This can be understood from the FeMn/FM interfacial interaction mechanism. Looking back at the FeMn induced magnetic anisotropy; the analysis is based on the FeMn 3Q spin structure. Monolayer-scale roughness at the AFM-FM interface leaves the FeMn spins at the [100]-type step edges to be uncompensated. The FM spins at the interstitials will be coupled to the FeMn uncompensated spins, forcing the FM spins to align at 45 degrees to the AFM uncompensated spins. This coupling of the FM spins with the uncompensated FeMn spins induces a 4-fold anisotropy in the Py layer. Therefore the Py 4-fold anisotropy should depend only on the in-plane component of the uncompensated FeMn spins at [100] steps of the Py/FeMn interface. Since the perpendicular component of FeMn spin is always compensated regardless of atomic steps on the (001) surface, it is reasonable to assume that FeMn grown on top of perpendicular Ni should not experience any in-plane magnetic moment change, i.e., the in-plane uncompensated FeMn spins at the [100]-type steps should not undergo any changes when grown on top of a perpendicular Ni. That explains why the perpendicular Ni in Py/FeMn/Ni/Cu(001) has no effect on the Py 4-fold magnetic anisotropy. On the other hand, when a FeMn film is grown on top of an in-plane Ni film, the FeMn in-plane uncompensated spins at [100] steps are directly coupled to the Ni in-plane magnetization. If this interaction leads to a slight
deviation of the FeMn 3Q structure by increasing the FeMn in-plane projection compared to pure FeMn, it will lead to an effectively larger uncompensated magnetic moment even without changing the film roughness. This effect should be more pronounced at FeMn thicknesses just above the transition from PM to AFM where the AFM spins are not yet well ordered into the 3Q structure (‘soft AFM’) so that the large correlation length around the phase transition region would transfer the enhanced FeMn in-plane uncompensated spins from the FeMn/Ni interface to the Py/FeMn interface. This mechanism could explain a stronger 4-fold anisotropy of the Py for in-plane Ni sample in the region of 10 ML < \( d_{FeMn} < 15 \) ML where the FeMn just becomes AFM ordered. For thicker FeMn where the AFM order is well established, the modification of the uncompensated FeMn spins at the FeMn/Ni interface won't propagate to the Py/FeMn interface so that no enhancement of \( H_4 \) is observed as compared to perpendicular Ni sample. We would like to point out that our discussion are conjectures without a firm experimental proof because at the present moment there is no effective tool to directly probe the FeMn spin structure. Future works are needed to further resolve this issue.

5.4 Summary

In summary, we studied epitaxially grown Py/FeMn/Ni/Cu(001) films as a function of FeMn and Ni thicknesses. MOKE and XMCD hysteresis loop measurement shows that the Py magnetization is in the film plane and thicker Ni magnetization, where the FeMn/Ni/Cu(001) alone has a perpendicular magnetization, undergoes a pseudo SRT with FeMn thickness at \( d_{FeMn} \sim 5 \) ML. ROTMOKE was employed to study the Py anisotropy as a function of the FeMn thickness and the Ni thickness. We find that the FeMn AFM order changes the sign of the Py 4-fold anisotropy to switch its easy axis from [110] to [100], but has little effect on the step-induced uniaxial anisotropy. For FeMn in the paramagnetic state or well-ordered AFM state, the Ni layer has little effect on the Py 4-fold anisotropy. For FeMn in the ‘soft-AFM’ state, the in-plane Ni enhances the Py 4-fold anisotropy.
Chapter 6

Tailoring the Topology of Artificial Magnetic Skyrmion

Despite theoretical predictions, it remains an experimental challenge to realize an artificial magnetic skyrmion whose topology can be well controlled and tailored so that its topological effect can be revealed explicitly in a deformation of the spin textures. Here we report epitaxial magnetic thin films in which an artificial skyrmion is created by embedding a magnetic vortex into an out-of-plane aligned spin environment. By changing the relative orientation between the central vortex core polarity and the surrounding out-of-plane spins, we are able to control and tailor the system between two skyrmion topological states. An in-plane magnetic field is used to annihilate the skyrmion core by converting the central vortex state into a single domain state. Our result shows distinct annihilation behaviour of the skyrmion core for the two different skyrmion states, suggesting a topological effect of the magnetic skyrmions in the core annihilation process.

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6.1 Introduction

A skyrmion is a topological twist of a continuous field that was first proposed by Skyrme [75, 76] to describe discrete nucleons. In condensed-matter physics, skyrmions emerge as topological invariant spin textures in a two-dimensional Heisenberg spin lattice [16]. Evidence of skyrmions in condensed matter physics appeared after the discovery of the Quantum Hall Effect in which the lowest energy charged excitations can be mapped onto two-dimensional magnetic skyrmion states [77, 78]. Although great effort has been made thereafter to understand the skyrmion effect in condensed matter physics, it was only recently considered that magnetic skyrmions may exist in real materials. The breakthrough occurred when it was proposed [79] that the Dzyaloshinsky - Moriya (DM) interaction [80] could compete with the Heisenberg collinear interaction to stabilize a skyrmion lattice in magnetic materials lacking inversion symmetry. Soon after this theoretical prediction, magnetic skyrmions were
identified in a narrow part of a phase diagram by neutron diffraction [81] and later further confirmed by direct imaging in real space [82, 83]. These newly discovered skyrmion materials [84] exhibit many fascinating phenomena such as the topological electric [85, 86, 87] and spin [88, 89] Hall effects, specific heat anomaly [90], skyrmion motions [91, 92] and excitations [93, 94], dynamics [95, 96], switching [95] and the topological knots of spin textures [97, 98] and so on. It is well recognized that these newly discovered skyrmion materials have opened a new field for the study of topological effect in fundamental physics [99, 100]. Despite this rapid progress, however, constraints on the existence of the skyrmions (for example, the limited range of magnetic field, temperature, length scale and so on) have been demanding an experimental realization of robust skyrmions whose topology can be well controlled and tailored in a wider range of parameter space so that topological effects can be directly revealed in the deformation of the spin textures. As outlined in ref. [84], there exist four general approaches to realize skyrmions: magnetic dipolar interaction, the DM interaction, frustrated exchange interactions or four-spin exchange interactions. In particular, the method based on the dipolar interaction could lead to the formation of skyrmions in centrosymmetric magnetic thin films [18, 101]. For example, it is recently shown that the so-called magnetic bubbles [102] in M-type ferrite thin films are actually magnetic skyrmions with rich topological structures [101]. Recognizing that an out-of-plane magnetic bubble in a perpendicularly magnetized thin film can be deformed smoothly into a magnetic vortex (including the out-of-plane vortex core) surrounded by an out-of-plane magnetized background, it is easy to understand the recent theoretical proposal that embedding a magnetic vortex state [103] into an out-of-plane magnetized environment could lead to the formation of an artificial skyrmion [104]. The challenging questions are: is it possible to realize such an artificial skyrmion in experiment; how to control and tailor its topologies; and what kind of topological effect can be retrieved from this engineered state? In this study, we report our experimental result on the realization of this type of artificial skyrmion by growing an epitaxial Co vortex disk on top of an out-of-plane magnetized Ni film. We show that we can control the skyrmion topological index of this system and subsequently reveal the topological effect in a skyrmion core annihilation process.

6.2 Experimental Design

We fabricated 30-nm-thick Co circular disks (radius= 1µm) on top of a 30 monolayer (ML) thick Ni film, which was grown epitaxially on a Cu(001) substrate. It is well known [68] that epitaxial Ni/Cu(001) has an out-of-plane magnetization above ∼7 ML Ni thickness, Co/Cu(001) film has an in-plane magnetization and Co/Ni(30 ML)/Cu(001) magnetization undergoes a spin re-orientation transition from out-of-plane to in-plane direction as the Co thickness increases above ∼1 nm. Magneto-Optic Kerr Effect (MOKE) measurement on Co/Ni(30 ML)/Cu(001) continuous film shows that the Co/ Ni(30 ML) film exhibits only the polar hysteresis loop (magnetic field applied in the out-of-plane direction) with a full remanence for Co thickness < 1 nm, and only the longitudinal hysteresis loop (magnetic
field applied parallel to the film plane) with a full remanence for Co thickness thicker than 1 nm, confirming that the Co/Ni(30 ML)/Cu(001) magnetization changes from out-of-plane to in-plane direction as the Co thickness increases above \( \sim 1 \) nm (Fig. 6.1). Therefore, our Co(disk)/Ni(30 ML)/Cu(001) sample should consist of two distinct regions in terms of magnetization direction: (1) the Co/Ni disk has an in-plane magnetization and (2) the Ni surrounding the disk has an out-of-plane magnetization. Element-specific X-ray magnetic circular dichroism (XMCD) measurements (Fig. 6.2) confirm this spin configuration that the Co exhibits an in-plane magnetic hysteresis loop with a full remanence and the surrounding Ni exhibits an out-of-plane magnetic hysteresis loop with a full remanence (the Ni XMCD measures only the Ni region surrounding the Co disks because of the surface sensitivity of the XMCD measurement).

**Figure 6.1:** Low-energy electron diffraction (LEED) patterns and hysteresis loops of Co/Ni(30 ML)/Cu(001). (a) LEED patterns confirm the formation of epitaxial single crystalline films. (b) MOKE hysteresis loops show that the Co/Ni(30 ML)/Cu(001) film has an out-of-plane magnetization for Co film thinner than 1 nm and an in-plane magnetization for Co film thicker than 1 nm.
6.3 Results

6.3.1 Static measurements of skyrmion.

The Co magnetic domain image from photoemission electron microscopy (PEEM) measurement clearly shows the formation of magnetic vortex state (spins curling around the centre of the vortex). The vortex contrast changes accordingly after changing the in-plane projection of the X-ray beam by 90° (Fig. 6.2d), confirming that the vortex state is an in-plane curling of the spin texture (the observation of the out-of-plane vortex core is beyond the PEEM spatial resolution). It should be mentioned that the 30-nm Co disk prevents a direct imaging of the Ni domain beneath the Co disk because of the PEEM surface sensitivity. However, the MOKE result (Fig. 6.1) and previous works [105, 106] in the literature show that the strong Co/ Ni interfacial coupling makes the Co/Ni bilayer behave as a single ferromagnetic layer with the same domain pattern. This assertion is further confirmed by our micromagnetic simulations (see below). Thus, we conclude that the Ni region beneath the Co disk follows the same Co vortex structure. This creates a magnetic system in which the Co disk (together with the Ni below the disk) forms an in-plane magnetic vortex with the Ni spins surrounding the disk having an out-of-plane magnetization. Note that an isolated magnetic vortex is half of a skyrmion [107] and that a vortex connected to an out-of-plane spin environment is a full skyrmion; our system of the central vortex connected to the out-of-plane surrounding Ni spins thus corresponds exactly to the skyrmion spin texture proposed in ref. [104]. The topology of a skyrmion is characterized by an integer index (the so-called skyrmion number) [84]

\[ N = \frac{1}{4\pi} \int dx dy \mathbf{n} \cdot (\partial_x \mathbf{n} \times \partial_y \mathbf{n}) \]  

where \( \mathbf{n}(x,y) \) is the normalized magnetization field (\( \mathbf{n} = \mathbf{M}/|\mathbf{M}| \)). The beauty of the skyrmion topology is that the skyrmion number is topologically invariant against a smooth deformation of the spin texture. In particular, the two skyrmion textures for a central vortex surrounded by out-of-plane spins have indices of \( N = 0 \) and \( N = 1 \), which are determined only by the central vortex core polarity relative to the surrounding out-of-plane spins (\( N = 0 \) for parallel alignment and \( N = 1 \) for antiparallel alignment) regardless of the spin texture details [84] [108]. In our experiment, we realize the \( N = 0 \) state by first applying a \( H = 20,000 \) Oe magnetic field in the out-of-plane direction and then turning off the field. As both the Ni film and the Co disk have an out-of-plane saturation field < 20,000 Oe (Fig. 6.2), the central vortex core polarity after turning off the magnetic field will be parallel to the surrounding Ni spins to form the \( N = 0 \) state (Fig. 6.3b). To prepare the \( N = 1 \) skyrmion state, we apply an 800 Oe magnetic field to the \( N = 0 \) state in the opposite direction of the original 20,000 Oe direction (for example, opposite to the core polarity direction) and then turn off the field. As the magnetic coercivity of the surrounding Ni is 500 Oe and the core polarity reversal field for a permalloy disk is > 3,000 Oe [109] (our simulation result shows that the vortex core switching field for our Co/Ni disk
should be $\sim 6,000 \text{ Oe}$), the 800 Oe field will only reverse the surrounding Ni spin direction without reversing the central vortex core polarity. Therefore, the ending state has opposite orientation between the central vortex core polarity and the surrounding Ni spins, that is, the $N = 1$ skyrmion state (Fig. 6.3b). We performed micromagnetic simulations using the parameters of our experimental system. Figure 6.3c shows the simulation result of the Co and Ni remanent domains after applying a 20,000-Oe out-of-plane magnetic field in the $-z$ direction and subsequently a 1,000-Oe out-of-plane magnetic field in the $+z$ direction. The simulation result clearly shows the formation of skyrmions in the Ni film. In fact, numerical calculation of the skyrmion number using equation (1) from the Ni spin texture in Fig. 6.3c yields $N = 0.0001$ for skyrmion core parallel to the surrounding Ni spins and $N = 0.9965$ for skyrmion core antiparallel to the surrounding Ni spins.
Figure 6.2: **Element-specific magnetic measurement of the Co disk and the surrounding Ni.** (a) PEEM image of the Co(disk)/Ni(30 ML)/Cu(001) sample fabricated using shadow mask. (b) X-ray absorption spectra (XAS) of Co and Ni at 2p levels for left circular polarized X-ray. The difference of the XAS for magnetization parallel (red colour) and antiparallel (blue colour) to the X-ray beam represents the element-specific XMCD signal. (c) Element-specific hysteresis loops shows that the Co disk (together with the Ni below the disk) has an in-plane magnetization and the Ni surrounding the disk has an out-of-plane magnetization. (d) Co PEEM images with different X-ray incident directions show that the Co disk in Co(disk)/Ni(30 ML)/Cu(100) forms a magnetic vortex state. The central vortex plus the surrounding out-of-plane Ni spins correspond to a magnetic skyrmion32.
Figure 6.3: A magnetic skyrmion consists of a central in-plane vortex and the surrounding out-of-plane spins. (a) $N = 0$ state and $N = 1$ skyrmion state are characterized by a parallel and antiparallel alignment of the central vortex core polarity relative to the surrounding out-of-plane spins, respectively. (b) $N = 0$ state and $N = 1$ skyrmion state in Co(disk)/Ni(30 ML)/Cu(100) can be prepared by switching the surrounding Ni spin direction without switching the central vortex core polarity using an 800-Oe out-of-plane magnetic field. (c) Micromagnetic simulation confirms the formation of $N = 0$ state and $N = 1$ skyrmion state by switching the surrounding Ni spin direction without changing the core polarity (blue dot at the centre).
6.3.2 Annihilation of skyrmion.

To reveal the topological effect of the skyrmion, we studied the skyrmion core annihilation by applying an in-plane magnetic field of various strengths. Within an in-plane magnetic field, the skyrmion core moves sideways to expand the domain parallel to the field [110]. If the in-plane field is not strong enough to push the core out of the disk (for example, annihilate the skyrmion core), the skyrmion core will move back to the disk central region after turning off the field. On the other hand, if the in-plane field reaches a critical field to annihilate the skyrmion core by pushing it out of the disk region, the disk region will be converted from a vortex state to a single domain state. As the Co dots have the radius of 1 µm and thickness of 30 nm, which lead to a bistable phase of Co domain within the disk region (vortex state and single domain state are both stable under zero magnetic field) [111], the single domain state will remain so after turning off the magnetic field. An interesting question is whether the \( N = 0 \) state and \( N = 1 \) skyrmion state need the same critical field to annihilate the skyrmion core (for example, to change the central vortex disk into a single domain state).

Figure 6.4 shows the PEEM images of the central vortex state after application of an in-plane magnetic field pulse of different strengths. The Co spin texture is imaged but, as discussed above, is also representative of the strongly exchange coupled Ni spins. For the \( N = 0 \) state, the central vortex state remains after the application of an in-plane magnetic field pulse up to \( \sim 100 \) Oe and switches to the single domain state for field greater than \( \sim 110 \) Oe (Fig. 6.4a). In contrast, the \( N = 1 \) skyrmion central vortex state remains up to a field of \( \sim 140 \) Oe and switches to the single domain state for field greater than \( \sim 160 \) Oe (Fig. 6.4b). As the PEEM images in Fig. 6.4a,b are taken from the same disk, we attribute the different critical fields in annihilating the skyrmion core to the different topologies of the skyrmion: a lower critical field for \( N = 0 \) state as opposed to a greater field for \( N = 1 \) skyrmion. We fabricated and measured 15 samples. Although the critical field varies from sample to sample due to inhomogeneity, we observe the same result that the critical field for \( N = 1 \) skyrmion is greater than the critical field for \( N = 0 \) state.

6.4 Discussion

The result can be understood once it is recognized that the spin texture, after the central vortex of a skyrmion becomes a single domain, has a skyrmion number of \( N = 0 \). Changing the \( N = 0 \) state into a single-domain disk corresponds to a smooth deformation of the spin texture within the same topological structure, but changing the \( N = 1 \) skyrmion into a single-domain disk requires a topological change of the spin texture. Therefore, the greater critical field for \( N = 1 \) skyrmion than \( N = 0 \) state could be interpreted as the extra field needed to break the topological knot of the \( N = 1 \) skyrmion. This interpretation is conclusive if the \( N = 0 \) state and \( N = 1 \) skyrmion state are energetically degenerate so that the extra field for the \( N = 1 \) skyrmion has to be associated with a topological barrier. If the \( N = 0 \) state has a higher energy than the \( N = 1 \) state (for example, due to the interaction between the
The skyrmion PEEM images after applying an in-plane magnetic field pulse. The central vortex is surrounded by out-of-plane Ni spins as indicated by the yellow symbols. (a) $N = 0$ state for parallel alignment between the central vortex core and the surrounding Ni spins, and (b) $N = 1$ skyrmion for antiparallel alignment between the central vortex core and the surrounding Ni spins. The critical field needed to switch the central vortex disk to the single-domain disk ($N = 0$ state) is weaker for $N = 0$ state than $N = 1$ skyrmion, suggesting a topological effect of in the skyrmion core annihilation process.

Microscopically, as the in-plane magnetic field pushes the skyrmion core to the vortex edge, the $N = 0$ vortex core can enter and disappear in the surrounding out-of-plane spins smoothly, because the core has the same spin direction as the surrounding spins (the initial and final states have the same topology). The $N = 1$ skyrmion core, however, cannot disappear smoothly inside the surrounding spins because of its opposite polarity relative to the environment (the initial and final states have different topologies). That explains why it takes higher critical field to annihilate the $N = 1$ skyrmion core than the $N = 0$ core. From this point of view, our case is similar to the process discussed in refs [95, 97] and where a spin-polarized current creates/annihilates a single skyrmion [98, 112]. Finally, we would like to point out that the circulation of the vortex (clockwise or counterclockwise spin curling around the vortex core) is not the topological winding number, which characterizes the spin rotation direction following a counterclockwise
contour around the vortex core. The topological winding number is always +1 for vortex state regardless of vortex circulation and is −1 only for an antivortex \([100]\). We confirmed this assertion by PEEM measurement that \(N = 1\) skyrmions of different circulations have the same critical field for skyrmion core annihilation.

In conclusion, we realized the artificial magnetic skyrmion proposed in ref. \([104]\) by epitaxially growing a Co disk on top of a perpendicularly magnetized Ni film. We show that the central vortex and the surrounding out-of-plane Ni spins form a magnetic skyrmion whose topology can be tailored between the \(N = 0\) and \(N = 1\) states by switching the surrounding Ni spin direction relative to the central vortex core polarity. The critical in-plane magnetic field to annihilate the skyrmion core is weaker for \(N = 0\) state than for \(N = 1\) state, suggesting a topological effect in the skyrmion core annihilation process.

### 6.5 Methods

#### 6.5.1 Sample fabrication.

The sample was prepared in an ultrahigh vacuum chamber with a base pressure of \(5 \times 10^{-10}\) Torr. A Cu(001) substrate was mechanically polished using diamond paste down to 0.25 \(\mu\)m followed by electropolishing. The Cu substrate was treated in the ultrahigh vacuum chamber by cycles of Ar ion sputtering at 2 keV and annealing at 600°C. Ni film (30 ML) was grown epitaxially on top of the Cu(001) substrate at room temperature. The Co was deposited epitaxially at room temperature on top of the 30 ML Ni film either as a continuous film for MOKE characterization or as disks for skyrmion study, by placing a shadow mask in situ on top of the Ni film. After the Co growth, 2 nm Cu protection layer was grown on top of the sample to prevent contamination. Low-energy electron diffraction (LEED) measurement confirms the formation of high-quality single crystalline epitaxial films (Fig. 6.1).

#### 6.5.2 Experimental setup.

MOKE was used to determine the easy magnetization axis of the continuous Co/Ni(30 ML)/Cu(001) film. Magnetic hysteresis loops were recorded for magnetic field applied in the out-of-plane and the in-plane directions. A loop with a full remanence shows the easy magnetization character and a loop with zero remanence shows the hard magnetization axis. As the penetration depth of light in metal is \(\sim 10\) nm, MOKE signal in Fig. 6.1 represents the hysteresis loop of the whole Co/Ni magnetization. Element-specific XMCD \([113]\) was measured at the Advanced Light Source of Lawrence Berkeley National Lab. on the Co and Ni 2p level at BL6.3.1 for hysteresis loop measurement and at BL11.0.1.1 for PEEM magnetic imaging. Circular polarized X-rays experience different absorption at the 2p \(L_{3,2}\) edges for magnetization parallel and antiparallel to the X-ray beam, respectively (Fig. 6.2). Therefore, one can image spin textures with PEEM by taking images of the same area using left- and right-circularly polarized light at an appropriate energy and dividing one image by
the other. The resulting PEEM XMCD image exhibits intensity contrast for different spin orientations within the sample plane (bright, grey and dark for spins parallel, perpendicular and antiparallel, respectively, to the X-ray beam). The lateral magnetic spatial resolution of PEEM is $\sim 50 \text{ to } 100 \text{ nm}$ so that PEEM measurement cannot resolve the out-of-plane magnetic vortex core.

### 6.5.3 Micromagnetic simulation.

Micromagnetic simulation was carried out using the Object Oriented MicroMagnetic Framework code [114] based on the LandauLifshitz Gilbert equation. The model contains a Co disk on top of a continuous Ni film with the same parameters as our experimental system: saturation magnetization $M_{S,Co} = 1.4 \times 10^6 \text{Am}^{-1}$; exchange stiffness $A_{Co} = 3 \times 10^{-11} \text{Jm}^{-1}$; Co thickness $t_{Co} = 30 \text{nm}$; in-plane fourfold magnetic anisotropy [71] $K_{Co} = 8 \times 10^4 \text{Jm}^{-3}$ for Co and $M_{S,Ni} = 4.9 \times 10^5 \text{Am}^{-1}$; $A_{Ni} = 1.2 \times 10^{-11} \text{Jm}^{-1}$, $t_{Ni} = 5 \text{nm}$ for Ni layer. The cell size is $2 \times 2 \times 2.5 \text{ nm}$ in our simulation. The Ni film perpendicular anisotropy, $K_\perp = 3.5 \times 10^5 \text{Jm}^3$ is obtained from the equation of $H_S + 4\pi M_{S,Ni} = \frac{2K_\perp}{M_{S,Ni}}$, where $H_S$ is the saturating field of Ni in-plane hysteresis loop (Fig. 6.2). A Co/Ni interfacial coupling of $A_{Co/Ni} = 2 \times 10^{-11} \text{Jm}^{-1}$ is adapted, but the result is insensitive to $A_{Co/Ni}$ value at least in the range of $A_{Ni} < A_{Co/Ni} < A_{Co}$. Figure 6.5a shows the simulation result after removing $2.0 \text{ T}$ magnetic field in the $-z$ direction. The Co disk forms the expected vortex state with its core polarity in the $-z$ direction. The Ni spins form a skyrmion with the central vortex following the vortex structure of the Co disk and the Ni spins outside the disk region in the $-z$ direction (plus a domain wall between the central vortex and the outside spins). Subsequent simulations are performed when $H = 1,000 \text{ Oe}$ static magnetic field is applied in the $+z$ direction. It can be clearly seen that the outside out-of-plane Ni spins are switched from $-z$ to $+z$ directions, but the central vortex core remains in the $-z$ direction (Fig. 6.5). After this state, we performed two simulations. One simulation is to release the $H = 1,000 \text{ Oe}$ field to get the remanent state ($N = 1$ skyrmion shown in Fig. 6.3c). The other is the simulation by increasing the magnetic field in the $z$ direction. We find that the central vortex core remains in the $-z$ direction (blue dot at the core position, see Fig. 6.5) until $H = 6,000 \text{ Oe}$ where the central vortex core switches to the $+z$ direction (red dot at the core position, see Fig. 6.5).
Figure 6.5: Simulation result of the Co and Ni domains within different out-of-plane magnetic fields. (a) $H = 0$ Oe; (b) $H = 1000$ Oe; (c) $H = 3000$ Oe; (d) $H = 6000$ Oe. The surrounding out-of-plane Ni spins switch from $-z$ to $+z$ directions above $H = 800$ Oe, but the central vortex core switches its polarity from $-z$ to $+z$ directions at $H = 6,000$ Oe. This allows the switching of the skyrmion topology between $N = 0$ and $N = 1$ states by switching the surrounding Ni spin direction using an 800-Oe out-of-plane magnetic field without switching the central vortex core polarity.
Chapter 7

Topology of Spin Meron Pairs

Meron is a special topological object that carries only one-half of the topological charge unit. In condensed matter physics, a spin meron corresponds to one-half of a spin skyrmion. As compared to the many fascinating topological properties of skyrmion materials, little is known of the properties of spin merons especially on their formations. It was confirmed only recently that hedgehog merons could exist in pairs via a spin flux closure with opposite helicities. However, it is unclear on whether a single hedgehog meron could ever exist by pairing with another type of meron. Using element-resolved magnetic imaging measurement on epitaxial trilayer disks, we show that a spin meron with a full range of helicity, including the hedgehog meron, can be stabilized by pairing with another vortex meron with a fine tuning of the magnetic coupling between the two merons. Furthermore, the meron divergence is fully controlled by the polarity of the vortex meron, independent of the vortex helicity.

7.1 Introduction

Meron was described originally in the context of quark confinement as one-half of a topological unit and can exist only in pairs [115]. In condensed matter physics, a spin meron corresponds to one-half of a spin skyrmion which carries one unit topological charge [116, 117]. While spin skyrmions have been proposed [79, 104] and realized recently in experiments [81, 118] with many fascinating topological properties [12, 84], it remains a mystery on why single hedgehog spin meron has never been discovered although a magnetic vortex [103] has been argued to be a vortex-type spin meron [107]. Noticing that vortex-type (spins curling around a center) and hedgehog-type (spins diverge/converge from a center) skyrmions have different helicities (γ, defined as the whirling angle of a spin texture from a divergent structure) [12], the role of the helicity in the formation of the spin merons has been speculated on. Although a helicity change doesn’t change the topology of a spin texture, it was found recently that the helicity actually plays a critical important role in many skyrmion topological properties. For example, Nomura et al. showed that electric charge can be induced in hedgehog-type (γ = 0, π) spin textures on top of a topological insulator.
and subsequently affects the domain wall motions [119]. Yokoyama et al. proposed that the supercurrent in a superconductor/magnetic skyrmion/superconductor junction can be controlled by the helicity of the magnetic skyrmion [120]. Rowland et al. suggest that the skyrmion phase in a skyrmion crystal could be significantly expanded by changing the helicity from the vortex-type ($\gamma = \pm \pi/2$) in the Dresselhaus limit to the hedgehog-type ($\gamma = 0, \pi$) in the Rashba limit [121]. The above results on skyrmion research suggest that the helicity may also play a critical role in the formation of spin merons. Since the helicity is directly related to the magnetic charge in a magnetic system especially the magnetic surface charge at the boundary of a finite magnetic system, it has been speculated that hedgehog merons can only exist in pairs via a spin flux closure between the two merons. First evidence on the existence of hedgehog meron pairs comes from a study on a magnetic trilayer disk where two ferromagnetic (FM) disks are antiferromagnetically (AFM) coupled through a spacer layer [122]. Direct proof of the hedgehog-like meron pair structure, however, was provided only by using element-resolved magnetic imaging [123]. It was shown that the spin flux closure leads to an antisymmetric relation in the divergence and circulation of the two merons. On the other hand, recent result on magnetic bilayers showed that spin merons in one magnetic film can only be partially replicated in the second magnetic layer [124]. The above results raise a critical issue: does a hedgehog meron have to be paired with another hedgehog meron of opposite helicity (i.e., opposite divergence and circulation)? Or alternatively, can a hedgehog meron be stabilized by pairing it with another type of meron (e.g., vortex meron)? In this chapter, we report our result on the study of epitaxially grown trilayer disks in which two FM disks are coupled through a spacer layer. By synthesizing asymmetric trilayer disks in which one FM disk is forced to form a vortex meron, we are able to fully explore the meron state of the second FM disk as a function of the magnetic interlayer coupling between the two FM disks. We demonstrate that the second meron can be stabilized from the vortex-type to the hedgehog-type merons in the full range of helicity. In particular, a hedgehog meron could be stabilized by pairing with a vortex meron without the need of spin flux closure. In addition, we show that the divergence of the hedgehog meron is solely determined by the polarity of the vortex meron, independent of the vortex circulation.

7.2 Experimental design

The Ni/Fe/Co trilayers were grown epitaxially at room temperature in an ultrahigh vacuum (UHV) chamber with a base pressure of $1 \times 10^{-10}$ Torr. A Cu(001) substrate was treated in the ultrahigh vacuum chamber by cycles of Ar ion sputtering at 2 keV and annealing at 600°C. The Ni and Co thicknesses were fixed at 5 ML and 150 ML, respectively. The Fe spacer layer was grown into a wedge shape with its thickness varying continuously from 5 ML to 6 ML over $\sim 0.5$ mm lateral distance. Circular disks of 0.75 $\mu$m radius were deposited in the central area of the Cu substrate using a contact shadow mask, and continuous films were deposited outside the shadow mask region. Because of the small slope of the wedge (2 ML/mm), the Fe spacer layer thickness is virtually the same in each disk but varies...
from disk to disk continuously from 5 ML to 6 ML. It is well known that Ni and Co on Cu(001) have face-centered-cubic (fcc) structure exhibiting a FM order independent of layer thickness \( [68, 125] \). Fe films on Cu(001), however, have an fcc structure below 11 ML with its magnetic phase being FM for \( d_{Fe} < 4 \) and AFM for \( 4 < d_{Fe} < 11 \) ML \( [126] \). Low Energy Electron Diffraction was used to characterize the sample, and confirms the formation of fcc Ni, Fe, and Co films on Cu(001) \([\text{FIG. } 7.1(c)]\). Element-resolved X-ray Magnetic Circular Dichroism (XMCD) measurement was performed on the Co, Fe, and Ni 2p level at the Advanced Light Source of Lawrence Berkeley National Laboratory. Hysteresis loop measurement was taken at BL4.0.2 and magnetic imaging was taken at the Photoemission Electron Microscopy (PEEM) endstation at BL11.0.1.

![Figure 7.1](image)

Figure 7.1: (a) Schematic diagram of the Ni/Fe/Co/Cu(001) trilayer disks in which the ferromagnetic Ni and Co are magnetically coupled across the antiferromagnetic fcc Fe spacer layer. (b) PEEM image of the Ni/Fe/Co/Cu(001) trilayer disk. (c) LEED patterns from Ni/Fe/Co/Cu(001) films confirm the epitaxial growth of fcc Ni, Fe, and Co single crystalline films. The LEED patterns were taken at the energy of 130 eV for Cu, 142 eV for Co, 124 eV for Fe, and 123 eV for Ni.

### 7.3 Magnetic Interlayer Coupling

It is well known that magnetic interlayer coupling between two FM films across a non-FM spacer layer oscillates between FM and AFM couplings with the spacer layer thickness \( [127] \). Thus the interlayer coupling between the FM Ni and Co layers in our Ni/Fe/Co/Cu(001) trilayers is expected to oscillate with increasing the AFM fcc Fe spacer layer thickness \( [128] \). We studied the interlayer coupling by performing element-resolved hysteresis loop measurements.
of the Co and Ni layers at different Fe thickness (FIG. 7.2). At $d_{Fe} = 6$ ML, the hysteresis loops of both Co and Ni have the same shape and identical coercivity, showing a FM coupling between the Ni and Co magnetizations. At $d_{Fe} = 5$, the Co magnetization shows the typical hysteresis loop with its magnetization aligned to the magnetic field direction above the coercivity. The Ni hysteresis loop, however, saturates in the opposite direction of the magnetic field (e.g., the Ni magnetization is aligned antiparallel to the Co magnetization), showing an AFM coupling between the Ni and Co magnetizations. Moreover, the opposite alignment between the Ni magnetization and magnetic field shows that the Ni-Co AFM coupling is stronger than the Ni Zeeman energy for magnetic field at least up to 0.1 T. The Ni magnetization actually increases linearly above the coercivity with increasing magnetic field, and should eventually align to the magnetic field direction at strong enough field when the Zeeman energy overcomes the Ni-Co AFM coupling.

Figure 7.2: Element-specific hysteresis loops of Ni(5ML)/Fe/Co(150ML)/Cu(001). (a) $d_{Fe} = 5$ ML, (b) $d_{Fe} = 5.5$ ML, (c) $d_{Fe} = 5.75$ ML and (d) $d_{Fe} = 6$ ML. The linear slopes in Ni hysteresis loops above the coercivity are described by eqn. (S1).

From the linear slope of the Ni magnetization above the coercivity in FIG. 7.2a, we could extrapolate that it takes about $H \sim 3.5$ T to overcome the Ni/Co AFM coupling to align the Ni magnetization to the magnetic field direction. We estimate the strength of the magnetic interlayer coupling constants as follows. The interlayer coupling energy per unit area can be
written phenomenologically as:

\[ E_{IC} = -J_L \frac{\vec{M}_{Ni} \cdot \vec{M}_{Co}}{M_{Ni}M_{Co}} - J_Q \left( \frac{\vec{M}_{Ni} \cdot \vec{M}_{Co}}{M_{Ni}M_{Co}} \right)^2 = -J_L \cos(\theta_{Co} - \theta_{Ni}) - J_Q \cos^2(\theta_{Co} - \theta_{Ni}) \] (7.1)

Here \( \theta_{Co} \) and \( \theta_{Ni} \) denote the Co and Ni magnetization orientations, respectively. The bilinear coupling \( J_L > 0 \) (or \( J_L < 0 \)) favors a ferromagnetic (or antiferromagnetic) alignment and the so-called biquadratic coupling of \( J_Q < 0 \) favors an orthogonal (\( \theta_{Co} - \theta_{Ni} = \pm \pi/2 \)) alignment of the Ni and Co magnetizations, respectively. Then the total energy per unit area of the trilayer within an applied magnetic field \( H \) is:

\[ E = -M_{Ni}d_{Ni}H \cos(\phi_H - \theta_{Ni}) - M_{Co}d_{Co}H \cos(\phi_H - \theta_{Co}) - J_L \cos(\theta_{Co} - \theta_{Ni}) - J_Q \cos^2(\theta_{Co} - \theta_{Ni}) \] (7.2)

We have ignored the anisotropy terms here to best single out the effect of the magnetic interlayer coupling. This is justified when the interlayer coupling dominates the anisotropy energy. Since the thickness of Co is much greater than Ni, the Co magnetization is approximately parallel to the applied field direction. Then by setting \( \theta_{Co} = \phi_H \) we have:

\[ E = -M_{Ni}d_{Ni}H \cos(\phi_H - \theta_{Ni}) - M_{Co}d_{Co}H \] (7.3)

Minimizing the total energy by \( \frac{dE}{d\theta_{Ni}} = 0 \), we derive the Ni magnetization angle:

\[ -M_{Ni}d_{Ni}H - J_L - 2J_Q \cos(\phi_H - \theta_{Ni}) = 0 \] (7.4)

Note that \( \phi_H = 0 \) for \( H > 0 \) and \( \phi_H = \pi \) for \( H < 0 \) during the hysteresis loop measurement, then we have:

\[ \cos \theta_{Ni} = \begin{cases} \frac{-J_L - M_{Ni}d_{Ni}H}{2J_Q}, & \text{for } H > 0 \\ \frac{J_L - M_{Ni}d_{Ni}H}{2J_Q}, & \text{for } H < 0 \end{cases} \] (7.5)

provided that \( |\cos \theta_{Ni}| \leq 1 \) (e.g., bounded by the saturation field). This equation gives us the component of Ni magnetization along the applied field direction, which is the linear portion of the Ni hysteresis loops in Figure 7.2. With known values of \( M_{Ni} = 4.9 \times 10^5 \text{A/m} \) and \( d_{Ni} = 1 \text{ nm} \), we fit the linear portion of the Ni hysteresis loops at four different Fe thicknesses and obtain the following bilinear and biquadratic coupling strengths (Table 7.1):

At the remanent state, minimizing the interlayer coupling energy leads to different coupling angles of \( \Delta \theta = \theta_{Ni} - \theta_{Co} \) at different relative strengths of \( J_L \) and \( J_Q \), summarized as follows for \( J_Q < 0 \):

\[ J_L > 2J_Q \rightarrow \Delta \theta = 0 \]
\[ J_L < -2J_Q \rightarrow \Delta \theta = \pi \]
\[ -2J_Q < J_L < 2J_Q \rightarrow 0 < \Delta \theta < \pi \] (7.6)
### Table 7.1: Bilinear and biquadratic coupling strengths

<table>
<thead>
<tr>
<th>$d_{Fe}$ (ML)</th>
<th>$J_L (mJ/m^2)$</th>
<th>$J_Q (mJ/m^2)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>&lt; -0.68</td>
<td>-0.34</td>
</tr>
<tr>
<td>5.5</td>
<td>-0.13</td>
<td>-0.093</td>
</tr>
<tr>
<td>5.75</td>
<td>0.058</td>
<td>-0.073</td>
</tr>
<tr>
<td>6</td>
<td>&gt; 1.02</td>
<td>-0.51</td>
</tr>
</tbody>
</table>

From the $J_L$ and $J_Q$ values obtained from the hysteresis loops, we expect a full range of $0 < \Delta \theta < \pi$ between the Ni and Co magnetizations as the Fe thickness changes from 5 ML to 6 ML, permitting a full exploration of the Ni-Co meron pair formation in Ni/Fe/Co disks as we will demonstrate. Note that adding a 4-fold magnetic anisotropy will modify the formula a little but won’t alter the result that, as the bilinear coupling changes from ferromagnetic to antiferromagnetic coupling, the coupling angle $\Delta \theta$ changes continuously from 0 to $\pi$ at the presence of a biquadratic coupling [129].

#### 7.4 Meron Pair Formation

In our sample of Ni(5 ML)/Fe/Cu(150 ML) disks, the thick Co disk in the trilayer should behave similar to a single Co disk to form a vortex meron. PEEM imaging measurement shows that the Co disk indeed forms the magnetic vortex meron ($\gamma_{Co} = \pm \pi/2$) in the trilayer disks with the Co spins curling around the disk center along the four equivalent [110] magnetization easy axes (FIG. 7.3a e). No hedgehog merons were found in the Co disk for all Fe thicknesses studied.

In contrast, the Ni disk exhibits a variety of meron states at different Fe thicknesses. At $d_{Fe} = 5$ ML, the Ni disk forms a vortex meron (FIG. 7.3a) with it helicity opposite to that of Co disk ($\gamma_{Ni} = -\gamma_{Co}$). This is because the strong AFM coupling between the Ni and Co magnetizations at $d_{Fe} = 5$ ML (FIG. 7.2) favors an antiparallel alignment of the Ni and Co magnetization directions. Similarly, the Ni disk also forms a vortex meron (FIG. 7.3i) at $d_{Fe} = 6$ ML with its helicity the same as that of Co ($\gamma_{Ni} = +\gamma_{Co}$) because of the strong FM interlayer coupling between Ni and Co magnetizations. The most interesting observation is that the Ni meron state undergoes a drastic change (FIG. 7.3gi) with increasing Fe thicknesses. From the Ni domain XMCD contrast, we find that the Ni spin direction (the colored arrows in Figure 7.3) changes continuously from $\pm \pi$ (AFM coupling) to 0 (FM coupling) relative to the Co spin direction with increasing the Fe thicknesses from 5 ML to 6 ML. In particular, we found that Ni hedgehog meron ($\gamma_{Ni} = 0, \pm \pi$) could be stabilized with the Co vortex meron ($\gamma_{Co} = \pm \pi/2$) to form a hedgehog/vortex meron pair which has never been discovered before. The distortion in some of the Ni domains (e.g., the edge in Figure 7.3) is perhaps due to the imperfection of the sample fabrication using the shadow mask.
which disturbs the domain more easily in thin film (Ni) than thick film (Co). Our result shows that the Ni helicity can be tuned in the full range of 0 to $\pm \pi$ by the magnetic interlayer coupling without the need of Dzyaloshinskii-Moriya interaction \cite{130}. Furthermore, we found no correlation between the divergence ($d = +1$ for divergence, and $d = -1$ for convergence) of the Ni hedgehog meron and the circulation ($c = 1$ for counter-clockwise, and $c = -1$ clockwise) of the Co vortex meron.

The above result can be understood qualitatively by recalling that different helicities lead to different magnetic charges $\rho_B = -\nabla \cdot \vec{M}$, $\sigma_B = \vec{M} \cdot \hat{n}$ in a FM disk, where $\vec{M}$ is the magnetization, $\hat{n}$ is the unit vector in the normal direction of the disks edge surface, and $\rho_B$ and $\sigma_B$ are the volume and surface magnetic charge densities, respectively. It is known that a curling structure of spins (vortex meron) could minimize the magnetostatic energy by minimizing the magnetic charges at the disk boundary, thus permitting the formation of single vortex meron in a FM disk. In contrast, a divergent structure of spins (hedgehog meron) would increase the magnetostatic energy due to the exposure of the magnetic charges at the disk boundary, thus forbidding the formation of single hedgehog meron in a FM disk. However, magnetostatic energy could be lowered by the magnetic charge-charge attraction between two hedgehog merons carrying opposite magnetic charges, thus favoring the formation of hedgehog meron pairs in a trilayer disk. In literature, such spin textures
with radially diverging/converging spins have been referred to as a meronlike-state \cite{122} or effective merons \cite{123}, and indeed obey the spin flux closure relation in the divergence and circulation of the two merons \((d_1 = -d_2; \ c_1 = -c_2)\) in trilayer disks of similar FM disk thicknesses. On the other hand, this mechanism also limits the accessible helicity of meron pairs because both merons have to change simultaneously with respect the change of the interlayer coupling.

In our system, the Co disk is much thicker than the Ni disk so that the Co vortex meron is virtually unaffected by the Ni meron state. In contrast, the Ni disk has a much less magnetic surface charge at the disk edge because of its ultrathin thickness so that its magnetization direction is almost completely determined by its magnetic coupling to the Co layer. That is why a single hedgehog meron in the Ni disk can be paired with the Co vortex meron in our system under a biquadratic coupling between the Ni and Co layers. In fact, as shown in the supplemental materials, the interlayer coupling angle between Ni and Co magnetizations changes continuously from 0 to \(\pi\) as the Fe thickness changes between 5 and 6 ML. Thus the Ni meron helicity could change continuously in the full range without the need of an opposite circulation to that of Co. Using micromagnetic simulations, we confirmed that these configurations are energetically stable states (Figure 7.4). Hence we showed that it is not necessary for both merons to have non-zero divergence as long as the interlayer exchange coupling dominates the total energy of the trilayer in an asymmetric sample structure.

![Figure 7.4: (a) Sketch of the sample geometry used in the micromagnetic simulation [23]. (b) - (d) shows typical micromagnetic simulation results using Object Oriented MicroMagnetic Framework (OOMMF) package. Both divergent and convergent meron states can be stabilized in the Ni disk while the much thicker Co disk retains a vortex meron state. The grayscale color is used here to emulate the measurement results from PEEM, which is sensitive only to magnetization along x-ray direction. Simulation parameters are detailed in section 7.5.](image)

### 7.4.1 Topological Imprinting

Since the interlayer coupling energy depends only on the angle between the two FM layer magnetizations, there are two degenerate configurations at any given interlayer coupling which correspond to a clockwise and counter-clockwise rotation of the Ni magnetization with respect to the Co magnetization (or \(\gamma_{Ni} = \gamma_{Co} + \Delta \gamma\) and \(\gamma_{Ni} = \gamma_{Co} - \Delta \gamma\) for a given
Except the special cases of a complete FM or AFM alignment between the Ni and Co magnetizations ($\Delta \gamma = 0$ or $\pi$), there should always exist two possible Ni configurations with $+\Delta \gamma$ or $-\Delta \gamma$ rotation at any local position of the trilayer disk. In all simulation results, we never find coexistence $\pm \Delta \gamma$ in the Ni disk, i.e., $\Delta \gamma$ never switch its sign within the Ni disk from location to location. For example, at the 90° coupling angle between the Ni and Co magnetizations, we find that all spins in the Ni disk are either divergent ($\gamma_{Ni} = 0$) or convergent ($\gamma_{Ni} = \pm \pi$) but not a mixture of divergent/convergent (e.g., not an antivortex where the spins are divergent in two directions and convergent in the orthogonal two directions), supporting our experimental observation (Figure 7.5). This result shows that as the Co disk imprints the spin texture into the Ni disk through the magnetic interlayer coupling, the topological winding number [12] of the Co vortex meron is preserved and carried over into the Ni meron, similar to bilayer skyrmion imprinting [19, 131]. Then the interesting question is: what determines the Ni meron divergence?

Figure 7.5: Hedgehog/Vortex meron pairs in Ni/Fe/Co/Cu(001) disks. (a) and (c) Co disk exhibits vortex meron state, and (b) and (d) Ni disk exhibits hedgehog meron state with either divergent structure for $p = +1$ core polarity or convergent structure for $p = -1$ core polarity.
7.4.2 Divergence/Core correlation

In a trilayer with comparable FM disk thicknesses, the divergence of one meron is always accompanied with the convergence of another meron. Magnetic simulation result shows that the divergence of the meron is determined by the polarity of the meron pairs. The above results has been attributed to the spin flux closure mechanism \[123\]. In our system, the Co disk is always a vortex meron and we find no correlation between the Ni divergence and the Co vortex circulation direction. Therefore the in-plane spins of the Co vortex do not play any role in the Ni meron divergence. Noting that a single meron should not have the divergence/polarity correlation because of the up/down symmetry, we believe that the divergence/polarity correlation in the trilayer disk originates from the interaction between the Ni in-plane spins and the Co vortex core. We recall that a magnetic vortex meron consists of in-plane spins curling around an out-of-plane spin core. Our simulation result (section 7.5.1) shows that a vortex meron generates a stray magnetic field similar to a field from a magnetic dipole moment that is located at the vortex core and points in the same direction as the vortex core polarity. Therefore the Ni meron in the Ni/Fe/Co disk is actually placed above a dipole moment from the Co vortex core. This Co dipole moment, whose magnitude scales with the core volume (core area times the Co thickness), below the Ni disk should align not only the Ni out-of-plane core polarity to the same direction of the Co polarity but also the in-plane radial component of the Ni spins to the radial component of the Co dipole field at the Ni disk, leading to a positive or negative divergence of the Ni meron \((d_{Ni} = +1\) or \(-1\)) for Co out-of-plane core polarity pointing towards the Ni disk \((p_{Co} = +1)\) or away from the Ni disk \((p_{Co} = -1)\). Note that Co dipole field has no effect on the tangential component of the Ni spins, neither does it depend on its own tangential spins. That is why we observed no correlation between the circulations, neither any correlation between the Ni divergence and the Co circulation, in our Ni/Co meron pair. To test the divergence/polarity correlation in experiment, we applied a \(\pm 2\) T out-of-plane magnetic field to align the vortex core before collecting the domain images \[19\]. Since the Curie temperature of \(\sim 5\) ML Ni film is very close to room temperature, we demagnetized the sample at \(\sim 100\) K to reduce thermal fluctuations in order to reach the ground state of the merons. The result shows that the Ni merons are divergent for the \(+2\)T pre magnetic field, and convergent for the \(-2\)T pre magnetic field. Micromagnetic simulation (section 7.5.2) shows that divergent and convergent Ni merons have different energies at a fixed Co vortex polarity, confirming the divergence/polarity correlation.

7.5 Micromagnetic Simulation

Micromagnetic simulations were carried out using the Object Oriented MicroMagnetic Framework code based on the Landau Lifshitz Gilbert equation. The following magnetic materials parameters are used in all simulations presented in this work: Saturation magnetization \(M_{Co} = 1.4 \times 10^6 Am^{-1}\), exchange stiffness \(A_{Co} = 3 \times 10^{-11} Jm^{-1}\) for the Co layer, and
$M_{Ni} = 4.9 \times 10^5 Am^{-1}$, $A_{Ni} = 1.2 \times 10^{-11} Jm^{-1}$ for the Ni layer. The diameter of the coupled disks are set to be 1 $\mu$m. For the simulation results shown in FIG. 7.4, a bilinear coupling of $J_L = 4 \times 10^{-4} Jm^{-2}$, a biquadratic coupling of $J_Q = -4 \times 10^{-4} Jm^{-2}$, and a fourfold magnetic anisotropy of $K_{Co} = 8 \times 10^4 Jm^{-3}$ were used in the simulation to resemble the experimental conditions more closely.

7.5.1 Stray magnetic field from a Co vortex meron.

To show that the stray field from a vortex meron is similar to a field from a dipole moment, we performed a micromagnetic simulation to calculate the stray magnetic field from a 50 nm thick Co disk of 500 nm radius. We chose a polarity of $p=+1$ for the simulation and a cell size of $2 \times 2 \times 2$ nm for the stray field calculation. The stray field resulting from this Co disk at a position 1.5 nm above the disk top surface (where the Ni disk would have been in the actual experiment) is shown in FIG. 7.6. First, we find that both right- and left-circulated vortex merons produce the same stray magnetic field (FIG. 7.6a and 3b), confirming that the stray field is mostly generated by the vortex core and is independent of the circulation. Second, the in-plane stray field is always divergent for $p = +1$ even when the out-of-plane component of the stray field could change from $+z$ direction (red colored region) to $z$ direction (blue colored region). FIG. 7.6c shows the stray field from a single magnetic dipole moment (pointing to the $+z$ direction) placed $\sim 17$ nm below the calculated surface. The result represents the actual stray field from the Co vortex meron well, confirming that the stray field of a vortex meron is equivalent to the stray field from a dipole moment. The position of 17 nm distance below the surface is above the center of the Co disk which is 26.5 nm below the calculated surface. This is because a magnetic moment closer to the surface contributes to the field more than a moment further away from the surface.

7.5.2 Energy difference between divergent and convergent merons.

To calculate the energy difference between the divergent and convergent Ni meron with a fixed Co meron vortex core polarity, we performed micromagnetic simulations with various thicknesses of Ni. The cell size used for these set of simulations is $5 \times 5 \times 0.5$ nm. The bilinear and biquadratic interlayer exchange coupling constants are set to $J_L = 0$ and $J_Q = -5 \times 10^{-4} Jm^{-2}$ so that the coupling angle between Co and Ni is set at $\Delta \gamma = \pi/2$ to manifest the divergence/polarity correlation (the result won’t change at $\Delta \gamma \neq \pi/2$). For a fixed Co vortex core polarity, we found that the Ni meron always has its core polarity the same as the Co, i.e., a Ni meron cannot be stabilized with its core polarity opposite to the Co core polarity. Under the condition of the same Ni and Co core polarity, we found that both $+\Delta \gamma$ and $-\Delta \gamma$ Ni meron states can be stabilized as metastable states. However, the lower energy state always has a divergent (convergent) structure for up (down) polarity of the meron core. FIG. 7.7 shows the energy difference ($\Delta E$) between these two states for different Ni thickness ($d_{Ni}$) at a fixed Co thickness of $d_{Co} = 50$ nm. The $\Delta E$ value increases almost
linearly with the Ni thickness. Note that the anisotropies of Co and Ni are ignored in this set of simulations since we were only interested in a qualitative picture of the energy difference between the two states $\pm \Delta \gamma$.

The linear dependence in FIG. 7.7 is consistent with the physical picture described earlier that the Ni spin divergence (convergence) is a result of the dipole interaction between the Ni spins and the Co vortex core dipole moment. To further confirm this physical picture, we consider the following over simplified model where the effective magnetic dipole moment of the Co vortex core is described approximately by $\vec{m}_{\text{Co}} = M_{\text{Co}} \pi \delta^2 d_{\text{Co}} \hat{z}$, where $\delta$ is the radius of the Co vortex core, and $d_{\text{Co}}$ is the thickness of the Co disk. This Co effective dipole moment is located at a distance $s$ below the Ni core. Noticing that the difference between the convergent and divergent Ni meron states is simply reversing the in-plane spin direction outside the core, then the energy difference between the convergent and divergent Ni meron
states is approximately:
\[
\Delta E = \int_{\delta}^{R} 2\pi \rho d\rho \frac{\mu_0}{4\pi |r|^3} \left( 3(\mathbf{m}_{\text{Co}} \cdot \hat{r})(\Delta \mathbf{m}_{\text{Ni}} \cdot \hat{r}) - \mathbf{m}_{\text{Co}} \cdot \Delta \mathbf{m}_{\text{Ni}} \right)
\]
\[
= \frac{3\mu_0}{2\pi} M_{\text{Co}} \pi \delta^2 d_{\text{Co}} M_{\text{Ni}} d_{\text{Ni}} \int_{\delta}^{R} \frac{s^2 \pi \rho^2}{(\rho^2 + s^2)^{3/2}} d\rho
\]
\[
= 3\pi \mu_0 M_{\text{Co}} M_{\text{Ni}} \pi \delta^2 d_{\text{Co}} d_{\text{Ni}} \frac{1}{s} \left( \frac{\delta^2}{(\delta^2 + s^2)^{3/2}} - \frac{R^2}{(R^2 + s^2)^{3/2}} \right)
\]
(7.7)

This equation gives rise approximately to a linear dependence on \(d_{\text{Ni}}\). Using \(R = 500\) nm for the disk radius, \(d_{\text{Co}} = 50\) nm, and \(s = 17\) nm + \(d_{\text{Ni}}/2\) as the approximate distance of the effective Co moment below the Ni disk, the calculated \(\Delta E\) as a function of \(d_{\text{Ni}}\) (blue curve in FIG. 7.7) could reproduce the simulation result very well at the value of \(\delta \approx 5.2\) nm which agrees well with the Co vortex core size. For this energy difference to be able to separate the convergent and divergent meron states in experiment, the energy difference (\(\Delta E_l\)) by flipping a block of spins from divergent to convergent over a characteristic length scale of \(l\) must be greater than the thermal excitation energy of \(k_B T\). At 5 ML Ni thickness, \(\Delta E \approx 1\) eV so that the order of magnitude of \(\Delta E_l\) can be roughly estimated to be \(\Delta E_l \approx \Delta E_l^2 / R^2\). In experiment, we found that the divergence/polarity correlation is more significant at low temperature (\(T \sim 100\)K) than room temperature (\(T = 300\)K) where the thermal excitations start to fluctuate the meron state. Therefore, taking \(T = 300\)K, we estimate that the length
scale of the Ni spin fluctuation needs to be at least \( l \sim R\sqrt{k_B T/\Delta E} \sim 80 \text{ nm} \) to manifest the divergence/polarity correlation. Of course the real physical process of thermal flipping of the Ni spins should be more complicated than the above over simplified estimation. Future theoretical studies are obviously needed to address such thermal flipping process of the meron states.

7.6 Summary

In summary, we synthesized single crystalline Ni/Fe/Co/Cu(001) trilayer disks and investigated the formation of meron pairs in the Co and Ni disks. By employing asymmetric thickness to force the Co into a vortex meron state, we successfully achieved a full-range control of the helicity of the Ni meron by tuning the Ni-Co magnetic interlayer coupling across the fcc Fe spacer layer. In particular, we find that a single hedgehog meron could exist by pairing with a vortex meron. In addition, we find that the divergence/convergence of the Ni hedgehog meron is determined by the Co vortex core polarity, independent of its circulation.
Chapter 8

Vortex Circulation Switching in FM/FE System

We demonstrate a non-volatile magnetic vortex circulation switching induced by piezoelastic strain in nanoscale Co disks. Magnetic uniaxial anisotropy measurements reveal that the magnitude of strain transferred from a ferroelectric PMN-PT(011) substrate to an adjacent ferromagnetic Co film strongly depends on the thickness of the spacer material between the ferromagnetic layer and the substrate. We observed that inserting Cu spacer layer will reduce the magnitude of transferred strain by more than 20 times compared to the strain transferred with the ferromagnetic layer in direct contact with the substrate. We speculate that the switching of magnetic vortex circulation is a dynamical process, which requires strain pulses of appropriate magnitude and duration to be delivered to the magnetic vortex.

8.1 Introduction

Devices based on magnetic architectures are intrinsically more energy-efficient than conventional transistors because there is no leakage current. In addition, the non-volatility of magnetic memory makes it an ideal candidate for storing information [132]. Achieving a fast, low-energy-dissipation means of switching the magnetization state using external voltage/electric field is thus highly desirable from fundamental and technological viewpoint [133]. Multiferroic-magnetoelectric composite systems such as ferromagnetic-ferroelectric heterostructures are ideal candidates to realize a robust electrical means of controlling magnetization due to the coupling between ferroelectric and magnetic orders [134]. One main route through which magnetoelectric effect can be established is through strain, where strain-induced change on the ferromagnet is caused by applying electric field to a piezoelectric/ferroelectric layer in the ferromagnet/ferroelectric heterostructures. It was proposed that the magnetization of a single-domain nanomagnet can be switched with very small voltages that generate strain in the magnetostrictive layer of a magnetostrictive/piezoelectric system [135, 136]. Devices based on this phenomena have also been proposed with the
promise of being 3-5 orders of magnitude more energy efficient compared to conventional transistors [137, 138].

Phenomenologically, the influence of strain on the magnetostrictive layer is a spin reorientation transition (SRT) of the thin-film magnetization from in-plane to out-of-plane [139, 140]. More recently however, ferroelastic (71°, 109°) domain switching was observed in new generation single-crystal relaxor ferroelectrics Pb(Zn$_{1/3}$Nb$_{2/3}$)O$_3$-PbTiO$_3$ (PZN-PT) and Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$-PbTiO$_3$ (PMN-PT). The unique switching pathways in these materials allow for an in-plane spin reorientation transition of the magnetization as a function of out-of-plane applied electric field [141, 142, 143, 144, 145]. Reproducible switching between two single domain states have been proposed [146] and demonstrated recently in experiment [147]. Similar manipulation of a magnetic vortex state in Ni nanostructured squares have also been demonstrated [148]. However, the use of 50 nm thick Pt between the ferromagnetic layer and the ferroelectric substrate in these works raises the question of how exactly does strain get transferred from the substrate to the ferromagnetic film through such a thick spacer layer. In this chapter, I will show that the insertion of a spacer layer between the ferromagnetic layer and the ferroelectric substrate will greatly reduce the magnitude of shear strain that can be transferred to the ferromagnetic layer. Furthermore, I will demonstrate that the circulation of magnetic vortices can be switched by applying strain pulses of certain magnitudes and length.

8.2 Experiments

8.2.1 Sample preparation

PMN-PT(011) substrate was annealed in an ultrahigh vacuum with base pressure of 2 × 10$^{-10}$ Torr to a temperature of 600°C. Next, Copper (Cu) steps of various thicknesses (0, 2, 4, 15 nm) are grown at room temperature as a spacer layer on a single substrate. 30 nm thick continuous film Co was then grown on top of the copper spacer. The Co film is finally capped with 2 nm Cu protection layer. This metallic upper layer also serve as a top electrode, while a 15 nm thick Cu layer was grown on the back-side of the PMN-PT(011) substrate to serve as the back electrode. The electric field was set by applying a constant voltage between the top electrode and the back electrode across the 0.5 mm thick PMN-PT(011) substrate. The experimental geometry for the uniaxial anisotropy measurement is shown in Figure 8.1(a). The optical plane of the magneto-optical Kerr effect (MOKE) is set up such that the Kerr signal measures the projection of the in-plane magnetization along the [110]-axis (x-axis in Figure 8.1(b)) of the PMN-PT crystal as a 300-Oe magnetic field rotates the magnetization within the film plane. The difference between the field angle and the magnetization angle relative to the [110] axis reveals information on the magnetic anisotropy of the Co film, specifically by calculating the magnetic torque equilibrium condition from the measured Co Kerr angle as a function of applied field angle [67]. The advantage of measuring magnetic anisotropy rotating-field MOKE (ROTMOKE) compared to analyzing the squareness of a
magnetization hysteresis loop is that with ROTMOKE we can obtain both positive and negative anisotropy values without having to rotate the sample geometry.

Figure 8.1: (a) Experimental geometry for uniaxial anisotropy measurements under a perpendicularly applied electric field using ROTMOKE. (b) Sketch of PMN-PT(011) crystal structure.

8.2.2 ROTMOKE measurements

Figure 8.2 shows the results of MOKE measurements of the Co(30 nm)/ PMN-PT(011) sample. Figure 8.2(a) and (b) show the hysteresis loops and magnetic torque on the Co film at two different applied electric field values. We can clearly observe a sign change in the induced anisotropy when the strength of electric field applied perpendicular to the sample is changed from 0.35 to 1.0 MV/m. This sign change is well known and has been reported before, although the details are lacking [145]. The magnetic anisotropy field constant $H_2$ was measured using the ROTMOKE as a function of electric field and the full bipolar loop is shown in Figure 8.2(c). Starting from zero applied field, the ferromagnetic thin film first obtained a positive uniaxial anisotropy as the electric field is increased. The uniaxial anisotropy then switches sign to negative at around the 0.35 MV/m (the coercivity of the ferroelectric substrate polarization). As the electric field is continuously increased, the magnitude of the uniaxial anisotropy also continues to increase until it saturates at around 0.6 MV/m aplied electric field. When the electric field is reduced after saturation, the uniaxial anisotropy displays a hysteretic behavior as it does not trace the same curve as with increasing-electric-field case. The magnetic anisotropy response to the applied electric field is symmetric between positive and negative electric field values. Overall the uniaxial anisotropy has a butterfly-shaped hysteresis, similar to the hysteresis loop of the strain/lattice constant observed in
To confirm this result, we repeated this systematic measurements with the sample rotated by 90 degrees in-plane. The measured uniaxial anisotropy values are the same except for a sign change, as expected. This behavior strongly suggest that the observed uniaxial anisotropy is induced by strain that is transmitted from the piezo-response of the substrate to the applied electric field. The magnetic uniaxial anisotropy is related to strain via the following relation:

$$K_2 = -\frac{3}{2}\lambda_S Y |\epsilon_x - \epsilon_y| = (\mu_0 M_S H_2)/2$$  \hspace{1cm} (8.1)

where $K_2$ denotes the induced uniaxial magnetoelastic anisotropy energy, $\lambda_S = 60$ ppm is the magnetostrictive constant of Co, $Y = 209$ GPa is the Youngs modulus of Co, $M_S = 1.4 \times 10^6 A/m$ is the saturation magnetization of Co, and $|\epsilon_x - \epsilon_y|$ is the in-plane strain difference between the piezoelectric response in the $x-$ and $y-$ direction due to the transverse electric field applied in the $z-$ direction. Using this equation, we estimate that the average in-plane strain difference $|\epsilon_x - \epsilon_y|$ is about 623 ppm throughout the Co film when a saturating electric field is applied on the substrate (corresponds to an induced magnetic uniaxial anisotropy energy of about $K_2 = 12 kJ/m^3$). This is smaller than the values reported in literature [7], which indicate that some relaxation of the strain occurs throughout the thickness of Co layer.

The behavior of this uniaxial anisotropy can be understood as follows [145]. The ferromagnetic Co film was grown on top of the PMN-PT(011) substrate in its remanent state, hence the relaxed state of the ferromagnetic film correspond to ferroelectric domains that are randomly distributed along the 8 different [111] easy-direction orientation. In this state, the polycrystalline Co film contains no (or very small) in-plane magnetic anisotropy. When an electric field is applied, say in the $-z$ direction, the polarization of the ferroelectric substrate prefer to point along the two out-of-plane [111] directions (i.e. [111] and [111] directions in Figure 8.1). This causes the ferromagnetic film that is grown over ferroelectric domains with in-plane orientation to experience contraction along one of the two in-plane axis, since the transverse piezoelectric response is different in the $x-$ and $y-$ direction [142]. As a result, the ferromagnetic film obtained a uniaxial magnetic anisotropy when measured using the same geometry as previous measurements.

Next we inserted a copper spacer layer between the ferromagnetic Co layer and the ferroelectric PMN-PT(011) substrate to study the effect on the induced magnetic uniaxial anisotropy of the ferromagnetic layer. We showed in Figure 8.3, the uniaxial anisotropy of Co film as a function of applied out-of-plane electric field for different copper spacer thicknesses. As expected, the insertion of copper spacer dramatically reduces the magnitude of the induced magnetic anisotropy. We observed that the behavior of the uniaxial anisotropy versus electric field appear to be identical between 0, 2 and 4 nm of copper spacer thicknesses, only with different magnitudes. The induced magnetic anisotropy and, by extension, transferred strain is the largest when the ferromagnetic Co film is in direct contact with the ferroelectric PMN-PT(011), that is at $d_{Cu} = 0$ nm, $H_2 = 2K_2/M_S = -167.5$ Oe at saturation (1 MV/m applied electric field). For $d_{Cu} = 2$ nm, $H_2 = -57.4$ Oe at saturation, and for $d_{Cu} = 4$ nm, $H_2 = -55.7$ Oe at saturation.
Figure 8.2: (a) Hysteresis loops measured at 0.35 MV/m and 1.0 MV/m applied electric field. (b) Magnetic torque experienced by the magnetization as a function of magnetization angle. (c) Uniaxial magnetic anisotropy field as a function of applied electric field strength.

When the copper spacer layer is thick enough, the induced magnetic uniaxial anisotropy is negligible in comparison. Assuming a constant $\lambda_S$ for the Co film with different copper spacer thicknesses, the smaller induced uniaxial anisotropy also means that the strain transferred to the Co film is weaker. From Figure 8.3, we can conclude that the strain transferred to the ferromagnetic layer from the ferroelectric substrate will be significantly reduced when it reaches the ferromagnetic layer if the Cu spacer thickness is greater than $\sim 10$ nm, for example at $d_{Cu} = 15$ nm, the induced uniaxial anisotropy is $H_2 = -8.2$ Oe at saturation, corresponding to a transferred strain of about 30 ppm, which is more than 20 times smaller than the magnitude of strain transferred when Co is in direct contact with the substrate.
Figure 8.3: The magnitude of the induced uniaxial anisotropy is greatest when Co is in direct contact with the substrate. With 2 or 4 nm Cu spacer between Co and the PMN-PT(011) substrate, the shape of the uniaxial anisotropy as a function of applied electric field is similar to that with Co in direct contact with PMN-PT, but with smaller magnitude.

This is in disagreement with previously observed results whereby it was claimed that significant shear strain can be transmitted across a 50 nm thick Pt layer \[147, 148\]. It leaves an interesting question on whether the choice of spacer layer material will be critical in the transfer of strain. Note that our samples with different spacer thicknesses were grown on the same substrate, hence we can assume the same behavior of strain from the substrate. Measurements done on different substrates with similar sample structure would have variations in the shape of the hysteresis, but with the same magnitude of induced uniaxial anisotropy. This may be due to the variations in the growth condition of the substrate, or different shape of ferroelectric hysteresis due to different annealing history.

8.2.3 PEEM measurements

To study the effect of induced uniaxial anisotropy on the vortex state, we prepared Co (30 nm)/Cu spacer (0, 4 nm)/PMN-PT(011) sample. This time the Co layer is patterned into a \(\sim 1.5\) \(\mu\)m disks by evaporating Co through a contact shadow mask (Figure 8.4). Element-resolved XMCD was measured at the Advanced Light Source of Lawrence Berkeley National
Laboratory on the Co 2p level at BL11.0.1. Circular polarized X-rays experience different absorption at the 2p $L_{3,2}$ edges for magnetization parallel and antiparallel to the X-ray beam. Element-resolved PEEM images were taken from the Co disks using left- and right-circularly polarized x-rays. From the contrast, one can retrieve the Co domain patterns as well as their relative magnetization directions.

Figure 8.4: (a) A sketch of the sample structure for PEEM measurements. (b) A PEEM image of Co L3 edge showing the formation of Co disks.

Figure 8.5(a)-(b) shows the domain images as a function of different applied electric fields for Co disks in direct contact with the PMN-PT(011) substrate for two different disks. We can see transitions from a vortex state to single domains and vice versa. The behavior appears random, but is consistent with the ROTMOKE data where the uniaxial anisotropy first becomes positive and then negative before fully saturating. Note that to ensure the upper electrode is conductive, and to provide a good contact between the PEEM optics and the thin film, copper capping layer (~ 10 nm) was added after the magnetic Co disks are formed. This relatively thick layer causes in fewer secondary electron reaching the CCD camera that reduces the image quality. We can still observe contrast in the circular dichroism which allows us to make some qualitative observation, however quantitative evaluation of the domain size area changes due to the induced uniaxial anisotropy is not possible. Figure 8.5(c)-(d) shows the domain images of Co (30 nm disks)/Cu (4 nm)/PMN-PT(011) at different applied electric field for two different Co disks. We observed that the circulation of a Co magnetic vortex switches from counter-clockwise to clockwise when the applied electric field is increased from 0.4 MV/m to 0.6 MV/m (Figure 8.5(c)). On another disk (Figure 8.5(d)), we observed that the circulation switches from clockwise to counter-clockwise from 0.2 MV/m to 0.4 MV/m, but returns to clockwise circulation when the electric field is increased further from 0.4 MV/m to 0.6 MV/m. This behavior was observed for many different magnetic disks, with apparently random switching behavior. Measurements done on Py disks with similar sample
structure have been done, and vortex circulation switching was also observed with the same random behavior. Note that the XMCD contrast images are better for Figure 8.5(c)-(d), since the presence of spacer layer allow us to grow thinner copper capping layer as the top electrode, allowing more secondary electrons to escape.

![Figure 8.5: A series of XMCD images taken using PEEM at increasing electric fields of (a, b) Co/PMN-PT(011) and (c, d) Co/Cu(4 nm)/PMN-PT(011)](image)

The non-uniform behavior of these ferromagnetic disks can be attributed to the size of the disk which is comparable to the typical ferroelectric domain size of PMN-PT(011). Since the ROTMOKE measurement is done with a He-Ne laser, the measured uniaxial anisotropy represents the property of a rather large area covered by the laser beam size (approximately 300 µm diameter). The measured induced uniaxial anisotropy represents the collective behavior of many ferroelectric domains, whereas the strain transferred to a single ferromagnetic disk would depend on the details of the ferroelectric domain wall motion.
Symmetry arguments lead to no motion of the vortex core as there are equal numbers of spins aligning in opposite directions along the anisotropy axis. This means that the process of circulation switching must be a dynamic one, i.e. gyroscopic motion of the vortex core driven by to time-dependent strain as described by Ostler et al [150]. It was also noted that the shape of the structure is not critical to the physics of strain driven gyroscopic motion. We speculate that applying the electric field across the sample is equivalent to applying a pulsed strain on the magnetic disks. This may result in a similar circulation switching process that has been proposed in literature recently for magnetic field pulses [151, 152], or for the core orientation switching by time-varying strain [150]. Because of the randomness involved during the ferroelectric domain switching process the exact profile (strength, duration, shape) of these strain pulses is unknown. Further theoretical and/or simulation work is needed to confirm this observation.

8.3 Summary

In summary, we have shown that strain can be transferred from the ferroelectric PMN-PT(011) substrate to an adjacent Co ferromagnetic layer. The magnitude of the transferred strain can be tuned by inserting copper spacer of various thicknesses between the ferromagnetic layer and the ferroelectric substrate. When the ferromagnetic layer is patterned into nano-scale disks such that magnetic vortices are formed, non-volatile switching of the vortex circulation can be achieved by applying strain pulses of appropriate magnitude and/or length.
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