Magnetic Skyrmions in Multilayers with Interfacial Dzyaloshinskii-Moriya Interactions

Xiao Wang

Bryn Mawr College

Follow this and additional works at: https://repository.brynmawr.edu/dissertations

Custom Citation

This paper is posted at Scholarship, Research, and Creative Work at Bryn Mawr College. https://repository.brynmawr.edu/dissertations/215

For more information, please contact repository@brynmawr.edu.
MAGNETIC SKYRMIONS IN MULTILAYERS
WITH INTERFACIAL DZYALOSHINSKII-MORIYA
INTERACTIONS

by

Xiao Wang

May, 2020

Submitted to the Faculty of Bryn Mawr College
in partial fulfillment of the requirements for
the degree of Doctor of Philosophy
in the Department of Physics
MAGNETIC SKYRMIONS IN MULTILAYERS
WITH INTERFACIAL DZYALOSHINSKII-MORIYA INTERACTIONS

Approved by:

Dr. Xuemei May Cheng, Advisor
Department of Physics
Bryn Mawr College

Dr. Mike Noel
Department of Physics
Bryn Mawr College

Dr. Michael Schulz
Department of Physics
Bryn Mawr College

Dr. David Schaffner
Department of Physics
Bryn Mawr College

Dr. Leslie Cheng
Department of Mathematics
Bryn Mawr College

Date Approved: April 13, 2020
ABSTRACT

Magnetic skyrmions, topologically stable spin textures that can be driven by electric currents efficiently, have both rich physics and potential applications in future technology such as spintronic devices for data storage. Magnetic skyrmions in multilayers with interfacial Dzyaloshinskii-Moriya interaction (DMI) were investigated in three aspects: stabilization, current-driven motion and antiferromagnetically coupled skyrmion pairs.

The formation and stabilization of magnetic skyrmions without external magnetic fields are determined by the competition of the Heisenberg exchange interaction, dipolar interaction, magnetic anisotropy, and interfacial DMI. In the [Pt/Co/HM]_n (HM = W, Mn, Ir, Au, n = 1, 3, 5, 8, 10, 12) multilayers, DMI and the dipolar interaction can be tuned by changing the heavy metal (HM) and the repetition number n, respectively. The size of the stabilized skyrmions decreases as the DMI or the dipolar interaction increases.

Current-driven skyrmion motion was investigated in a Ta/CoFeB/TaO_x multilayer and the skyrmion Hall effect was observed. The transverse motion of the skyrmions, driven by an electric current in a perpendicular magnetic field, can be quantized by the skyrmion Hall angle, the angle between the current direction and the skyrmion motion direction. A significant current/velocity dependence of the skyrmion Hall angle was observed. Combining the experimental results and the theoretic derivation based on the Thiele equation, we attribute the skyrmion Hall effect to the topological Magnus force and random pinning potentials in the materials.

To avoid the undesired skyrmions motion towards the device edge, antiferromagnetically coupled skyrmion pairs were realized and investigated in [Co/Gd/Pt]_10 multilayers. These skyrmion pairs are stable over a broad temperature range from about 300 K to 55 K. Below 50 K, the magnetization of the core area of the skyrmion pairs starts to rotate from the original direction of out-of-plane to in-plane. However, the domain wall regions are preserved during this spin reorientation transition (SRT). After the temperature is in-
creased back to above 50 K, skyrmions recover to their initial state before the SRT. The recovery of the skyrmions can be attributed to the persisted chiral domain walls due to the strong topological protection by DMI, confirmed by micromagnetic simulations.

This research work was conducted under the guidance of the author’s research advisor, Professor Xuemei May Cheng.
To my parents for their unconditional love and support
ACKNOWLEDGEMENTS

First and foremost, I would like to thank my advisor, Professor Xuemei May Cheng. As her first Ph.D student, I have received an enormous amount of valuable guidance, support, encouragement, and opportunities. Without her, my growth as a researcher and a person would not have been possible. I have learned so much through working with her and I am very grateful for the experience.

I deeply appreciate the invaluable help and advice from Prof. Xiaoshan Xu, Prof. Wanjun Jiang, Dr. Yaohua Liu, Dr. Suzanne te Velthuis, and Dr. Axel Hoffmann. I am grateful to have collaborated some projects with Dr. David Keavney, Prof. Kristen Buchanan, Dr. John Pearson, Prof. Xifan Wu, Prof. Weigang Wang, Dr. Anthony DiChiara, Dr. Rajesh Chopdekar, Dr. Sheng Zhang, Dr. Charudatta Phatak and Dr. Amanda Petford-Long. It has also been a pleasure to work with the following colleagues and fellow graduate students: Prof. Wei Zhang, Prof. Qiang Wang, Dr. Kishan Sinha, Dr. Xuanyuan Jiang, Dr. Pavel Lapa, Dr. Hilal Saglam, Zhuyun Xiao and Dr. Xichao Zhang.

I would like to thank all the faculty members of the Department of Physics: Prof. Michael Noel, Prof. Michael Schultz, Prof. Elizabeth McCormack, Prof Peter Beckmann, Prof. Mark Matlin, Prof. David Schaffner, and Prof. Leslie Cheng in the Department of Mathematics for their help along my way. I am very thankful for all the support from Maria Dantis, Patricia Lausch and Dr. Sharon Burgmayer. I would also like to express my thanks to the graduate students in the department Kristen Recine, Vincent Gregoric, Alex Chartrand and Andrew Clark for their friendships.

Finally, I would like to thank my parents, Xiuqin Shen and Zhaolin Wang and my dearest friend Barbara Tomczak for their encouragement, support, and caring.

This dissertation work was supported by the National Science Foundation under grants NSF-DMR 1708790 and NSF CAREER-DMR 1053854.
# TABLE OF CONTENTS

Abstract ................................................................. iii

Acknowledgments ......................................................... v

List of Figures .......................................................... x

Chapter 1: Introduction ................................................... 1

References of Chapter 1 ................................................... 7

Chapter 2: Magnetic skyrmions and Nanomagnetism in multilayers ............. 8
  2.1 Introduction ......................................................... 8
  2.2 Magnetic skyrmions ................................................. 8
  2.3 Nanomagnetism ...................................................... 15
    2.3.1 Heisenberg exchange interaction ............................ 17
    2.3.2 Interfacial Dzyaloshinskii-Moriya Interaction ............. 17
    2.3.3 Dipolar Interaction ........................................... 21
    2.3.4 Magnetic Anisotropy ......................................... 23
    2.3.5 Zeeman energy ............................................... 23
  2.4 Magnetic multilayers ............................................ 25

References of Chapter 2 ................................................ 30
### Chapter 3: Experimental Methods of Sample Fabrication and Characterization

<table>
<thead>
<tr>
<th>Section</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.1</td>
<td>Introduction</td>
<td>31</td>
</tr>
<tr>
<td>3.2</td>
<td>Fabrication of Multilayer Thin Films</td>
<td>31</td>
</tr>
<tr>
<td>3.2.1</td>
<td>Sputtering Deposition</td>
<td>31</td>
</tr>
<tr>
<td>3.2.2</td>
<td>Thickness Characterization</td>
<td>34</td>
</tr>
<tr>
<td>3.3</td>
<td>Device Patterning by Photolithography</td>
<td>36</td>
</tr>
<tr>
<td>3.4</td>
<td>Magnetic Property Characterization</td>
<td>38</td>
</tr>
<tr>
<td>3.4.1</td>
<td>Magnetometry</td>
<td>38</td>
</tr>
<tr>
<td>3.4.2</td>
<td>X-ray Magnetic Circular Dichroism Spectra</td>
<td>39</td>
</tr>
<tr>
<td>3.5</td>
<td>Magnetic Imaging Methods</td>
<td>40</td>
</tr>
<tr>
<td>3.5.1</td>
<td>X-ray Photoemission Electron Microscopy</td>
<td>40</td>
</tr>
<tr>
<td>3.5.2</td>
<td>Lorentz -Transmission Electron Microscopy</td>
<td>43</td>
</tr>
<tr>
<td>3.5.3</td>
<td>Magneto-optical Kerr Effect Microscopy</td>
<td>46</td>
</tr>
</tbody>
</table>

**References of Chapter 3**

<table>
<thead>
<tr>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>49</td>
</tr>
</tbody>
</table>

### Chapter 4: Magnetic skyrmions in [Pt/Co/HM]ₙ Multilayers with interfacial DMI

<table>
<thead>
<tr>
<th>Section</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.1</td>
<td>Motivation and Experiments</td>
<td>50</td>
</tr>
<tr>
<td>4.2</td>
<td>Confirmation of Néel-type skyrmions at room temperature and zero field</td>
<td>51</td>
</tr>
<tr>
<td>4.3</td>
<td>The effect of interfacial DMI on skyrmions</td>
<td>53</td>
</tr>
<tr>
<td>4.4</td>
<td>The effect of the dipolar interaction on skyrmions</td>
<td>62</td>
</tr>
<tr>
<td>4.5</td>
<td>Conclusions</td>
<td>66</td>
</tr>
</tbody>
</table>

**References of Chapter 4**

<table>
<thead>
<tr>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>69</td>
</tr>
</tbody>
</table>
# Chapter 5: Direct observation of Skyrmion Hall effect

5.1 Motivation and Experiments

5.2 Observation of skyrmion transverse motion

5.3 Topological Magnus force and Thiele equation

5.4 Current-dependent skyrmion dynamics

5.5 Skyrmion Hall angle diagram

5.6 Conclusion

References of Chapter 5

# Chapter 6: Persistence of chiral domain walls in Antiferromagnetically-coupled bubble Skyrmion pairs during spin reorientation transition

6.1 Motivation and experiments

6.2 AFM-coupled bubble skyrmion pairs at room temperature and zero field

6.3 Spin reorientation of the core area of the AFM-coupled bubble skyrmion pairs

6.4 Persistence of chiral domain walls

6.5 Conclusions

References of Chapter 6

# Chapter 7: Conclusions

List of Publications

Appendix A: Calculation of topological charge of a magnetic skyrmion

Appendix B: Sum rules
Vita ................................................................. 119
LIST OF FIGURES

2.1 Illustration of the Néel-type magnetic skyrmions in multilayers with the interfacial DMI. Arrows represent the atomic spin. Different colors correspond to spin component along the $z$ direction as indicated in the color scale bar. (a) Schematic of spin texture of an ideal magnetic skyrmion followed by the cross section of the spin texture highlighted as a grey dashed line. (b) Spin texture of a bubble skyrmion with a uniformly magnetized core area. The center core area of a skyrmion bubble with magnetization pointing in the positive $z$ direction is shown as red arrows. Magnetization at the boundary area, represented by blue arrows, is along the negative $z$ direction. The transition between magnetization $M_z = 1$ and $M_z = -1$ is the Néel-type domain wall shown in (b) highlighted by the frame with dashed outlines. Cross section of the spin texture marked as a grey dashed line is also presented. (c) Relation of concepts of skyrmion, bubble and bubble skyrmion. .......................... 10

2.2 Illustration of Néel-type magnetic skyrmion with $Q = \pm 1$ and $\chi = \pm 1$. Arrows represent atomic spins. Different color corresponds to different net magnetization along the $z$ direction as indicated in the color scale bar. The spins at the center of the skyrmions in (a) and (c) point along the positive $z$ direction indicating the topological charge $Q$ being $+1$ while (b) and (d) are opposite situations. The transition between spin up and spin down is counterclockwise (CCW) for skyrmions in (a) and (b) giving rise to the different sign of chirality $\chi$ comparing to the clockwise (CW) transition in (c) and (d). The CCW and CW transitions are indicated as black arrows in (a) and (c). .......................... 12
2.3 Differences in topological charge between Néel-type skyrmion bubbles and other circular-shape magnetic domains with the emphasize on the domain wall area. Blue areas indicate uniform magnetization pointing out of the paper plane. Light brown areas are magnetization pointing into paper plane. Red arrows shows the trend of magnetization direction of the domain wall area between blue and light brown areas. (a) and (b) are two different bubble skyrmions with topological charges $Q = 1$ but $\chi = \pm 1$, respectively. (c) and (d) are two examples of magnetic bubbles with topological charges $Q = 0$. (e) and (f) are two examples of magnetic bubbles with topological charges $Q = 2$.

2.4 Schematic of current-driven skyrmion motion. Arrows represent the atomic spins. Different colors of the arrows correspond to different net magnetization along the $z$ direction as indicated in the color scale bar. Skyrmion move along $+y$ direction as marked by the large and thick blue arrow. The two opposite sides of skyrmions experience opposite direction of forces illustrated as thick green arrows due to the different local spin texture.

2.5 Illustration of the parallel and antiparallel alignment of neighboring spins when $J$ is (a) positive and (b) negative.

2.6 Mechanism for an interfacial DMI and spins at an interface of magnetic multilayers. Diagram redrawn in the style of Figure 1f in Nature Nanotechnology 8, 152-156 (2013). Spins $S_i$ and $S_j$ in the ferromagnetic (gray, upper) layer couple to each other through overlap of their wave functions with an atom with large spin-orbit coupling (blue, lower layer). This overlap gives rise to a contribution to the energy of the form $D_{ij} \cdot (S_i \times S_j)$, where $D_{ij}$ lies in the plane of the interface, in the direction normal to the plane defined by the three atoms [57]. The dark grey lines between atoms indicates the plane defined by the three atoms.

2.7 Mechanism of the relationship between the direction of $D_{ij}$ and spin canting direction. When the interfacial DMI vector $D_{ij}$ points out (into) of the paper plane, spin cants in counterclockwise (clockwise) direction.

2.8 Explanation of magnetic domains on the basis of energy minimization: (a) single domain with magnetic energy/flux around the magnetized body in the air, has the largest dipolar energy. (b) two domains decreases the total dipolar energy by containing part of the magnetic flux within the magnetic material and shorting the distance that the magnetic field has to travel in the air. (c) multiple domains with closure domains further reduce the magnetic energy due to shorter way the magnetic field within the magnetic material.
2.9 The effect of external magnetic field on topological charge of a skyrmion. Different colors correspond to different net magnetization along the $z$ direction as indicated in the color scale bar. (a) When the external magnetic field points at positive $z$ direction, the spins at the boundary of a magnetic skyrmion point at positive $z$ direction giving rise to a negative topological charge. (b) The direction of spins and the sign of topological charge reverse when the direction of the external magnetic field reverses. .......................... 24

2.10 Magnetic multilayers with tunable parameters and the associated properties. 25

3.1 Schematic of sputtering deposition. ................................. 32

3.2 Illustration of multilayered structures. (a), (b) and (c) are structures of multilayered samples studied in this thesis project in Chapters 4, 5 and 6 respectively. ................................................................. 33

3.3 Growth rate in sputtering deposition of elements in the multilayer film samples measured using crystal oscillator thickness monitor. ................................. 33

3.4 Schematic of x-ray diffraction in a thin film with thickness $t$. .......................... 35

3.5 X-ray small-angle reflectivity results and fitting results of multilayered films: (a) [Pt/Co/W]$_{n=8}$ (c) [Pt/Co/Ir]$_{n=8}$ and (d) [Co/Gd/Pt]$_{n=10}$. (b) The fitted results of the thickness and roughness of each layer of the results in (a). ... 35

3.6 One example of the designed pattern for studying current-driven skyrmion motion. .............................................................................. 36

3.7 Illustration of photolithography and ion milling procedures for patterning devices. ............................................................... 37

3.8 Schematic of XMCD spectra at $L_2$ and $L_3$ edges. (a) X-ray absorption spectra excited by LCP and RCP x-rays as well as the XMCD spectrum of Co metal at $L_2$ and $L_3$ edges. (b) Mechanism of XMCD. $2p_{3/2}$ and $2p_{1/2}$ are two different $2p$ levels (core level) with different spin-orbit couplings. Blue and Green electrons correspond to spin-up and spin-down electrons in the $2p$ states. Photons with LCP or RCP angular momentum, shown in blue or red circular arrows, excite electrons from core level to valence band above Fermi energy, corresponding to the absorption spectra in blue and red in (a). Electrons in $3d$ level are filled with spin-up and spin-down electrons at energy below Fermi energy $E_F$, shown as blue and green area respectively. Empty states in $3d$ level with energy above Fermi energy, shown as white area are the valence states for electrons excited from core level $2p$. ........... 41
3.9 Illustration of PEEM imaging experiment set-up and the mechanism. Left panel shows x-ray absorption spectra and XMCD spectrum of Ni $L_{2,3}$ edges. The dashed line marks the energy where XMCD has the highest intensity. PEEM images correspond to the XMCD contrast by taking the difference of an image taking with a LCP and RCP x-ray. X-ray incident angle is $30^\circ$. The contrast showing in the PEEM image is the magnetization of Ni projected along the x-ray propagation direction. The red arrows in the vortex image indicate the in-plane magnetization direction within the vortex. PEEM images are taken at beamline 4-ID-C of the APS at ANL.

3.10 Simulations of Lorentz TEM image assuming different spin textures. (a) Simulated Lorentz TEM images of Néel-type magnetic skyrmions with spins pointing towards [a-(1)] or out from [a-(2)] the core direction. a-(3-4): sample tilted by $\phi = -30^\circ$, (a)-(5) and (a)-(6): no sample tilt $\phi = 0^\circ$, and a-(7-8): sample tilted by $\phi = +30^\circ$. $x$ axis is the rotation axis indicated by the black arrow and the rotation direction is illustrated by the red arrow. Magnetic contrast is only visible when the sample is tilted, but the configurations seen in [a-(1)] and [a-(2)] appear indistinguishable, as shown in the intensity line profiles seen in a-(9). (b) Simulated Lorentz TEM images of topologically-trivial bubbles with spins pointing to the left or to the right: b-(1) and b-(2) respectively. Sample tilt provides further contrast between the different spin configurations to be distinguished.

3.11 Main parts and the schematics of a MOKE microscope.

4.1 Sample structure of multilayers Ta (2.0nm) /[Pt (1.5nm) /Co (1.0nm) /HM (1.0nm)]$_n$ /Pt (2.0nm) (HM = Mn, W, Ir and Au, $n = 1, 2, 3, 4, 5, 6, 8, 10$ and 12). The sandwich structure in red box is the unit of the repetition number $n$. 
4.2 Experimental identification of room-temperature Néel-type skyrmions in the absence of magnetic field in [Pt/Co/W]$_{n=8}$ multilayers. a, L-TEM image of a $\phi = -30^\circ$ tilted sample with zoomed-in features in the blue box. The black and red arrows indicate the sample tilt axis and the rotation direction, respectively. The bubble-like and stripe-like features are magnetic contrast. Dark and bright contrast come from domain walls. The magnetic contrast of these domain walls is dark in the upper left corner and bright in the lower right corner. b, L-TEM image of an un-tilted sample. The round features come from the grains in the film but not magnetic contrast. c, L-TEM image of a $\phi = +30^\circ$ tilted sample with zoomed-in features in the blue box. The magnetic contrast of each feature is bright in the upper left corner and dark in the lower right corner, opposite from a. d, Intensity profile of slices, highlighted as dotted green and red lines, along the diagonal direction of the zoomed-in feature in blue boxes. e-g, Simulation of L-TEM images of a Néel-type bubble skyrmion when tilted at $\phi = -30^\circ$, 0 and $\phi = +30^\circ$, respectively. The center grey region corresponds to the uniformly magnetized core area of a Néel-type bubble skyrmion. The white and black edges are the Néel domain walls.

4.3 L-TEM imaging of $+30^\circ$ tilted [Pt/Co/HM]$_{n=8}$ (HM=Mn, W, Ir, Au) multilayers. a, Stipe domains in [Pt/Co/Mn]$_{n=8}$. b, c and d, Néel-type skyrmions in [Pt/Co/W]$_{n=8}$, [Pt/Co/Ir]$_{n=8}$ and [Pt/Co/Au]$_{n=8}$ multilayers, respectively.

4.4 Calculation of DMI based on the field-dependence evolution of the skyrmion size in [Pt/Co/W]$_{n=8}$. a-d, L-TEM imaging at external magnetic fields of 7.6 mT, 21.0 mT, 25.0 mT and 29.0 mT, respectively. The increasing of perpendicular magnetic fields results in the shrinking/annihilation of skyrmions. e, The change of diameter of selected skyrmions, marked by green arrows in a, as a function of field. f, The reduced single skyrmion energy $\hat{U}$ calculated for three different values of the DMI strength $D_k$ (1.3, 1.5 and 1.7 mJ/m$^2$) at applied magnetic fields of 25.0 mT, 21.0 mT, 12.1 mT, and 7.6 mT. The sizes of the individual skyrmions from e that were labeled as $S_{k1}$-$S_{k5}$, are shown at each field as vertical colored long dashed lines. A DMI value $D_k = 1.5$ mJ/m$^2$ (black thick dotted curves) is inferred, as at this value the experimental skyrmion sizes lie within the minimum energy valley for all applied field.
4.5 Calculation of DMI in $[\text{Pt/Co/Au}]_{n=4}$ multilayers based on the evolution of the Néel-type skyrmions sizes as a function of perpendicular magnetic fields. a-d MOKE images at external magnetic fields of 131 Oe, 169 Oe, 201 Oe and 211 Oe. The increasing of perpendicular magnetic fields results in the shrinking/annihilation of skyrmions. e, The change of diameter of selected skyrmions, marked by different colors of circles in a, as a function of field. f, the reduced single skyrmion energy $\hat{U}$ calculated for three different values of the DMI strength $D_k$ (1.9, 2.1 and 2.3 mJ/m$^2$) at applied magnetic fields of 211 Oe, 201 Oe, 169 Oe, and 131 Oe. The sizes of the individual skyrmions from e that correspond to the color of the circles in a-d, are shown at each field as vertical colored long dashed lines. A DMI value $D_k = 2.1$ mJ/m$^2$ (red short dotted curves) is inferred, as at this value the experimental skyrmion sizes lie within the minimum energy valley for all applied fields. ................................................................. 58

4.6 Calculation of DMI in $[\text{Pt/Co/Ir}]_{n=4}$ multilayers based on the evolution of the Néel-type skyrmions sizes as a function of perpendicular magnetic fields. a-d, MOKE images at external magnetic fields of 0 Oe, 50 Oe, 60 Oe and 80 Oe. The increasing of perpendicular magnetic fields results in the shrinking/annihilation of skyrmions. e, The change of diameter of selected skyrmions, marked by different colors of circles, as a function of field. f, the reduced single skyrmion energy $\hat{U}$ calculated for different values of the DMI strength $D_k$ (2.1, 2.3 and 2.5 mJ/m$^2$) based on the effective medium model at applied magnetic fields of 80 Oe, 60 Oe, 50 Oe, and 40 Oe. A DMI value $D_k = 2.3$ mJ/m$^2$ (black short dotted curves) is inferred, as at this value the experimental skyrmion sizes lie within the minimum energy valley for all applied fields. ................................................................. 59

4.7 Parameters used in DMI calculation and the calculated DMI strength. ................................. 61

4.8 Field-dependent magnetization of $[\text{Pt/Co/HM}]_n$ (HM = W, Mn, Ir, Au) with an external magnetic field applied along a out-of-plane direction and b in-plane direction. ......................................................... 61

4.9 L-TEM imaging of $+30^\circ$ tilted $[\text{Pt/Co/Ir}]_n$ ($n = 8, 10$ and 12) multilayers and MOKE imaging of $[\text{Pt/Co/Ir}]_n$ ($n = 3, 5$). The black and red arrow indicate the sample tilt axis and the rotation direction, respectively. a and b, Néel-type skyrmions imaged by MOKE in $[\text{Pt/Co/Ir}]_n$ ($n = 3, 5$). In a and b, black contrast indicates the direction of magnetization is along $-z$ direction pointing into paper plane. c, d and e, Néel-type skyrmions imaged by L-TEM in $[\text{Pt/Co/Ir}]_n$ ($n = 8, 10, 12$) multilayers, respectively. f, The evolution of skyrmion size as repetition number $n$ in $[\text{Pt/Co/Ir}]_n$ multilayers. 63
4.10 Field-dependent magnetization of [Pt/Co/Ir]$_n$ (n = 3, 5, 8, 12) with an external magnetic field applied along a out-of-plane direction and b in-plane direction. .................................................. 65

4.11 Magnetic properties of [Pt/Co/Ir]$_n$ (n = 1, 2, 3, 4, 5, 6, 8, 12) multilayers. . 66

5.1 Illustration of ordinary Hall effect and skyrmion Hall effect. $J$ is the current. $j_e$ denotes the electron density. The negative sign means the electron motion is along negative $x$ direction from right to left. a, Ordinary Hall effect of an electron. The red arrow indicates the moving trajectory of an electron when there exists an magnetic field ($H$) orthogonal to the sample surface and the current direction. b, Skyrmion Hall effect of a skyrmion with topological charge $Q = -1$. The red arrow refers to the deviation of an skyrmion towards the edge of device from electron motion direction. Notice that an external magnetic field is not necessary for skyrmion Hall effect. .................................................. 71

5.2 Magneto-optical Kerr effect (MOKE) imaging of current driven skyrmion motion that deviates from the current direction towards the device edge. The blue area is substrate. The black area is the device. The dark contrast indicates the magnetization in this area is along $-z$ direction, the same direction as the external magnetic field of -5.2 Oe. The dimensions of the Hall bar device are 100 $\mu$m in width and 500 $\mu$m in length, but the area shown in the figure is about 100 $\mu$m $\times$ 160 $\mu$m. The red arrow on top of the device refers to electron motion direction. The magnitude of electron current density $j_e$ equals to $6.0 \times 10^6$ A/cm$^2$. The negative sign of $-j_e$ means the electron motion is along negative $x$ direction from right to left. The white contrast of the skyrmions suggests that the magnetization of the core area is along $+z$ direction and the topological charge of the skyrmion is +1. A few $Q = +1$ skyrmions moved approximately along a straight line, marked as a dashed grey line for eye guidance, forming an angle with the current direction.[30] .................................................. 73

5.3 Accumulation of skyrmions at the device edge. The dimensions of device is 60 $\mu$m in width and 500 $\mu$m in length, but the are shown in the image is about 60 $\mu$m $\times$ 140 $\mu$m. The red arrow refers to electron motion direction. The negative sign of $-j_e$ means the electron motion is along negative $x$ direction from right to left. a, Demonstration of skyrmion ($Q = -1$) accumulation at the edge of the device. This is done by repetitively applying 50 pulsed currents of 50 $\mu$s duration at a frequency of 1 Hz with a current density $j_e = 6 \times 10^6$ A/cm$^2$ and an applied field of +5.4 Oe along $+z$ direction. b, Reversing the magnetic field from $+z$ direction (+5.4 Oe) to $-z$ direction (-5.2 Oe) leads to the accumulation of skyrmions with positive topological charge $Q = +1$ at the opposite edge.[30] .................................................. 74
5.4 MOKE imaging of current-dependent skyrmion dynamics. All experiments were done by using 50 µs pulsed currents. The red arrow refers to electron motion direction. The plus sign of $+j_e$ indicates the electron motion is along positive $x$ direction from left to right. **a-e**, Snapshots of $Q = -1$ skyrmion motion captured after applying successive current pulses of amplitude $j_e = 1.3 \times 10^6$ A·cm$^{-2}$ and external perpendicular magnetic field $H_{\perp} = +4.8$ Oe. **f**, Summary of the skyrmion trajectory from **a-e**, showing no net transverse motion along the $y$ direction. **g-k**, Snapshots of $Q = +1$ skyrmion motion at $j_e = 1.3 \times 10^6$ A·cm$^{-2}$ and magnetic field $H_{\perp} = -5.2$ Oe. **l**, Stochastic trajectory from **g-k**, again showing no net transverse motion. **m-q**, Snapshots of $Q = -1$ skyrmion motion at $j_e = 2.8 \times 10^6$ A·cm$^{-2}$ and magnetic field $H_{\perp} = +5.4$ Oe. **r**, Summary of the trajectory from **m-q**. Nearly straight and diagonal trajectory indicates the presence of transverse motion along the $+y$ direction. The size of skyrmion shrinks slightly as compared to **a-e** due to the larger perpendicular magnetic fields. Two other skyrmions that moved into the frame, marked with green circles in **m** and **o**, were not studied. **s-w**, Snapshots of $Q = +1$ skyrmion motion at $j_e = +2.8 \times 10^6$ A·cm$^{-2}$ and magnetic field $H_{\perp} = -5.2$ Oe. **x**, Summary of the trajectory from **s-w**. Again, there is a nearly straight and diagonal trajectory. However, the slope is opposite, indicating that the presence of transverse motion is along the $-y$ direction and opposite to **r**.[30] 77

5.5 Current-dependence transverse motion of a single skyrmion. **a**, The average skyrmion velocity ($\bar{v}$) as a function of electron current density $j_e$. The light blue region corresponds to the skyrmion-pinning regime, where $j_e < (0.6 \pm 0.1) \times 10^6$ A·cm$^{-2}$. The beige region corresponds to the regime of stochastic motion without net transverse motion, where $(0.6 \pm 0.1) \times 10^6$ A·cm$^{-2} < j_e < 1.5 \times 10^6$ A·cm$^{-2}$. **b**, Evolution of the skyrmion Hall angle $\varphi_{sk}$ and the ratio between the transverse and longitudinal velocities of the skyrmion $v_y/v_x$. The green region corresponds to the regime without net transverse motion, where $j_e < 1.5 \times 10^6$ A·cm$^{-2}$. When $j_e > 1.5 \times 10^6$ A·cm$^{-2}$, both $\varphi_{sk}$ and $v_y/v_x$ are monotonically increasing as a function of current density.[30] 79
5.6 Skyrmion Hall effect diagram. Diagram of the skyrmion Hall angle as a function of current density/sign of topological charge in a modified device of dimensions $80 \, \mu\text{m} \times 100 \, \mu\text{m}$, obtained by tracking the motion of several skyrmions. In the low-current-density regime, the skyrmion Hall angle $\varphi_{sk}$ exhibits a linear dependence similar to that shown in b. A further increase of current density $j_e > 8 \times 10^{6} \, \text{A/cm}^2$ results in the saturation of the skyrmion Hall angle. By alternating the sign of the driving electron current density ($\pm j_e$) and the sign of topological charge ($\pm Q$), a phase diagram for the four different regimes was determined. Namely, for negative topological charge (under positive magnetic fields), regime I ($+j_e, -Q$) with positive $\varphi_{sk}$ and regime III ($-j_e, -Q$) with negative $\varphi_{sk}$ were identified by changing the polarity of the electron current. For skyrmions with positive topological charge (under negative magnetic fields) a positive $\varphi_{sk}$ in regime II ($-j_e, +Q$), and negative $\varphi_{sk}$ in regime IV ($+j_e, +Q$) were detected. The decrease of skyrmion Hall angle from $|\varphi_{sk}| \approx 32 \pm 2^\circ$ to $|\varphi_{sk}| \approx 28 \pm 2^\circ$ is also demonstrated by increasing the skyrmion diameter from $d = 800 \pm 300 \, \text{nm}$ ($+5.4 \, \text{Oe}$-$5.2 \, \text{Oe}$) to $d = 1100 \pm 300 \, \text{nm}$ ($+4.8 \, \text{Oe}$-$4.6 \, \text{Oe}$).[30] 

6.1 Illustration of an AFM-coupled bubble skyrmion pair, as a perspective view (at the top) and a view of the spin configuration along radial direction (at the bottom). Arrows represent the atomic spins. Color difference corresponds to different magnetization along the $z$ direction as indicated in the color scale bar. The topological charge $Q$ of the top (bottom) skyrmion is -1 (+1) and therefore the total $Q = 0$. Spins in the top skyrmion are one-on-one AFM-coupled to the spins in the bottom skyrmion, as shown by the opposite colors in the color scale.

6.2 PEEM images of AFM-coupled bubble skyrmion pairs at room temperature and zero field. a, Schematic of PEEM geometry. The yellow arrow indicates the x-ray propagation direction ($\mathbf{k}$) with an incident angle of $30^\circ$ degree. b and c, PEEM images acquired at the Co $L_3$ and Gd $M_5$ absorption edges at room temperature without external fields. The contrast of bright and dark corresponds to magnetization oriented along $-z$ and $+z$ directions, respectively. The field of view is 20 $\mu\text{m}$. Note that the bright areas in Co layer and the dark areas in Gd layer are the core area of the bubble skyrmions with uniformly magnetized spins in out-of-plane direction. The Néel domain wall is beyond the imaging resolution.
6.3 Observation of bubble skyrmion core area disappearance and recovery during spin reorientation at zero field in Gd layer. Scale bar in the PEEM image is 5 µm. PEEM images with blue (red) background are the images taken at different temperatures as temperature decreases (increases). All images are taken at Gd M₅ edge. a-d, XMCD results of calculated magnetic moment per Co and Gd atom along x-ray propagation direction at different temperatures and zero field. Each data point is the calculated magnetic moment per atom based on XMCD spectra and sum rules. The lines are for eye guidance. The open and solid symbols are the results when x-ray is incident at an angle of 20 and 70 degrees, respectively. Green and red symbols in a and c are the results of Gd moment during cooling down and warming up, respectively. Orange and purple symbols in b and d are the results of Co moment during cooling down and warming up, respectively. a and b are the results when x-ray is incident at 20 degrees, therefore are the magnetic moment projected to almost in-plane direction. c and d are the results when x-ray is incident at 70 degrees, therefore are the magnetic moment projected to almost out-of-plane direction. The curving arrows in a-d indicates the temperature changing direction of different curves with different colors.

6.4 Observation of the persisted chiral domain walls by PEEM imaging. Scale bar in the top left image is 1 µm. The images with blue background in the top two rows are the PEEM images of one skyrmion taken at Gd M₅ and Co L₃ edges at various temperatures during cooling down from room temperature to 33 K. Only some of the results at the representative temperatures are shown here. The blue arrow from left to right indicates the temperature cooling direction. The PEEM images with red background at the bottom row are the images taken at Gd M₅ during warming up from right to left as indicated by the red arrow. The dotted red arrows in all the images surround the bubble skyrmion and highlight the wall area of the bubble skyrmions.

6.5 Micromagnetic simulations: persistence of in-plane magnetization at the wall area. a Magnetic domain pattern at 160 K in Gd layer. b Magnetization configuration at 30 K in Gd layer. c The overlapping of 40% transparency of the area within dotted red lines and the area within the black dotted lines in b. Note that the highlighted area in b is the corresponded area of the highlighted area in a at 30 K. The small triangles indicate the local magnetization direction. The in-plane magnetization at the wall area (boundary) of the bubble in a and the magnetic configuration at the same position in b are very similar. d is the same as a. e is the z component of b. f Magnetic domain pattern at 160 K in Gd layer after going through spin reorientation. g The overlapping of e and 40% transparency of d. h The overlapping of e and 40% transparency of f.
6.6 Micromagnetic simulations: persistence of topological charge during spin reorientation. a-c, The $x$, $y$ and $z$ component of the skyrmion bubble at 160 K in Gd layer before going through spin reorientation, respectively. d-f, The $x$, $y$ and $z$ component of the recovered skyrmion bubble after spin reorientation in Gd layer. The calculated topological charge is 1 for both skyrmion bubbles before and after going through spin reorientation even though the shape is slightly different. 

6.7 Micromagnetic simulations: the effect of DMI when $K_{eff} = 4.9 \times 10^5$ J/m$^3$. First row: relaxed magnetic domain patterns at 160K. Second row: relaxed magnetic domain pattern after cool down to 30 K. Bottom row: relaxed magnetic domain patterns at 160 K after warm up from 30 K. Orange and green boxes are for Co and Gd respectively. Three DMI values were investigated and the left two, middle two and right two columns of Co and Gd are the results with the DMI = 0, DMI = 0.5 and 2.5 mJ/m$^2$ respectively.

B.1 Sum rules for 3d metals 

B.2 Sum rules for 5d metals
CHAPTER 1
INTRODUCTION

Magnetic skyrmions are chiral spin textures that have particle-like behavior [1]. As a result of the chiral rotation of the magnetic spins, skyrmions are topologically protected against continuous transition into topologically trivial states such as a ferromagnetic state [2, 3]. Magnetic skyrmions can have an extremely small size in the nanometre range, and be efficiently driven by an electric current [4, 5]. Therefore, they have attracted great interest because of their rich physics related to topological properties and their potential applications in technology such as data storage [1, 6].

Although theoretically predicted in the 1980s, magnetic skyrmions were first experimentally observed in 2009 as skyrmion lattice in single crystals of magnetic compounds with a non-centrosymmetric system, such as MnSi [7, 8]. Skyrmions were later observed in ultrathin magnetic films epitaxially grown on heavy metals, such as Fe monolayers and PdFe bilayers on Ir(111) in 2011 [9]. However, the stabilization of the skyrmion lattice in these materials requires large magnetic fields (∼ 1 Tesla) and cryogenic temperatures, which make them impractical for technological applications. Moreover, the skyrmion lattice ground state does not allow the specific properties of individual skyrmions to be exploited. Recent studies have found that magnetic skyrmions can also be stabilized at room temperature in magnetic multilayers with the interfacial Dzyaloshinskii-Moriya Interaction (DMI) [10, 11, 12, 13]. Magnetic multilayers with the interfacial DMI provide a promising room-temperature system with individual skyrmions for physics properties investigation and practical applications [6, 12, 13, 14]. Therefore, this thesis investigates magnetic skyrmions in multilayers with the interfacial DMI.

Magnetic skyrmions become an energetically favored magnetic state mainly due to the DMI, which is an asymmetric exchange interaction that tends to align the neighboring
spins perpendicular to each other [15]. By studying the magnetic properties of skyrmions, further knowledge about DMI can be obtained [10, 16]. On the other hand, the engineering of magnetic skyrmions can be enabled by better understanding of DMI and the interplay between DMI and other interactions such as the dipolar interaction and antiferromagnetic exchange interaction [17, 18, 19].

As topological spin textures, magnetic skyrmions also exhibit many unique physics properties when interacting with electric currents [2, 20]. For example, generating/writing magnetic skyrmions with a pulsed electric current [21, 22, 23], current-driven skyrmion motion [24, 25, 26, 27, 28, 29, 30], and topological Hall effect [31, 32] have been investigated by many researchers. Therefore, magnetic skyrmions have promising applications as information carriers in the next-generation data storage and spin logic devices [4, 5]. Instead of incorporating electron charges to produce functional devices, spintronic devices aim to manipulate electron spins, in addition to the electron charges [33, 34, 35]. A great example of spintronics is the discovery of the giant magnetoresistance (GMR), which was awarded the Physics Nobel Prize in 2007. Spintronic technologies memory such as magnetoresistive random-access memory (MRAM) [36, 37], and domain wall race-track devices [38], have been proposed as the solutions to non-volatile, energy-efficient, high-density, and high-performance data storage. However, magnetic skyrmions, as topologically protected spin textures, are better candidates. Magnetic skyrmions have a size as small as a few nanometers [9]. Magnetic skyrmions can also be more efficiently driven by electric currents, which can be a few orders of magnitude smaller than those needed for MRAM and domain wall race-track memory devices [5, 39]. Moreover, they are topologically protected from being pinned or destroyed by the defects encountered when moving in the device [20, 2]. The development of skyrmion-based topological spintronics holds promise for the future non-volatile data storage with ultra high density and speed, as well as ultra low energy, even though there are still many critical physics phenomena needed to be better understood [1].
This dissertation focuses on the investigation of skyrmions in magnetic multilayers with the interfacial DMI and their current-driven dynamics. The dissertation is organized as follows. Chapter 2 provides the details about magnetic skyrmions and the theoretical background concerning how to stabilize skyrmions in magnetic multilayers with the interfacial DMI. Chapter 3 starts with the deposition of magnetic multilayers, and then focuses on the experimental methods used for characterization and imaging of magnetic skyrmions. Chapter 4 presents the study of the effects of DMI and the dipolar interaction on magnetic skyrmions. Chapter 5 describes the skyrmion Hall effect, which is a phenomenon observed in the current-driven motion of magnetic skyrmions. Chapter 6 is devoted to antiferromagnetically-coupled skyrmion pairs, which are expected to resolve the issue of undesired skyrmion Hall effect in the spintronics application.

References of Chapter 1


CHAPTER 2
MAGNETIC SKYRMIONS AND NANOMAGNETISM IN MULTILAYERS

2.1 Introduction

To understand the physics phenomena related to magnetic skyrmions and realize the potential applications for data storage, it is important to study magnetic skyrmions and nanomagnetism concerning their formation, stabilization, and manipulation, as well as the relevant material systems. This chapter first presents the concepts and properties of magnetic skyrmions in section 2.2. Then explains the mechanism for the formation and stabilization of magnetic skyrmions, which is the energy minimization in section 2.3. Among all the interactions, the interfacial Dzyaloshinskii-Moriya interaction (DMI) is the key in stabilizing magnetic skyrmions. Section 2.4 proposes a model materials system, multilayers with the interfacial DMI, for studying magnetic skyrmions.

2.2 Magnetic skyrmions

Skyrmions were originally postulated by Tony Skyrme in the 1960s as topologically protected non-linear vector fields to explain the stable states of some particles, such as neutrons in nuclear physics [40]. The concept of "topological protection" implies that those non-linear vector fields have some unique configurations, characterized by integer topological numbers (-1, 0, 1, 2, -2, 3 etc), and can not be transformed continuously into each other. Hence, skyrmions and those particles are stable [40].

Though Skyrme’s idea about stable non-linear vector fields due to topological protection was not agreed upon and applied by the majority of nuclear physicists at that time, researchers have adapted his "skyrmion" concept into other fields. Some topologically protected non-linear spin textures were proposed or experimentally observed in research
fields, such as Bose-Einstein condensates [41, 42], high-Temperature superconductors [43, 44], chiral liquid crystals [45, 46] and magnetic thin films [1, 47, 48, 49, 50].

To the interest of this thesis work, in magnetic multilayers with the interfacial DMI, a magnetic skyrmion is a chiral spin texture characterized by an integer topological charge \( Q \). A topological charge \( Q \), also referred to as the winding number, identifies how many times the local unit magnetization is wrapped around a sphere. It is the result of magnetization rotation. Mathematically, it is the integration of the unit vector \( \mathbf{m}(r) \) over the \( x - y \) plane, as

\[
Q = \frac{1}{4\pi} \int \mathbf{m}(r) \cdot (\partial_x \mathbf{m}(r) \times \partial_y \mathbf{m}(r)) \, d^2r.
\]  

(2.1)

The derivation and calculation of the topological charge of a magnetic skyrmion is discussed in details in Appendix.

One example of a magnetic skyrmion in magnetic multilayers with the interfacial DMI is a Néel-type skyrmion, a chiral spin texture characterized by topological charge \( Q = +1 \), with a central out-of-plane magnetization and a 180-degrees gradual rotation of magnetization along the radial direction, as illustrated in Figure 2.1(a). The transition between the center and the boundary has the same spin configuration of a Néel-type domain wall, a type of transition between two different uniformly magnetized areas, as the reason for the name "Néel-type skyrmion".

The size of experimentally reported skyrmions are varied from \( \sim 50 \) nm to a few hundreds \( \mu \)m [1, 47, 48, 49, 50]. The large \( \mu \)m size skyrmions are Néel-type bubble skyrmions, as illustrated in Figure 2.1(b) with the transition between the core area and boundary as Néel-type domain walls. The spins in the center regions (shown by the red arrows) of bubble skyrmions are uniformly magnetized and form a magnetic domain, where magnetization are uniformly aligned in one direction. Magnetic domains can have different shapes. A circular shaped domain is called a magnetic bubble.

The relations among magnetic bubbles, skyrmions and bubble skyrmions are shown as the Venn diagram in Figure 2.1(c). Conceptually, bubble skyrmions correspond to the inter-
Figure 2.1: Illustration of the Néel-type magnetic skyrmions in multilayers with the interfacial DMI. Arrows represent the atomic spin. Different colors correspond to spin component along the $z$ direction as indicated in the color scale bar. (a) Schematic of spin texture of an ideal magnetic skyrmion followed by the cross section of the spin texture highlighted as a grey dashed line. (b) Spin texture of a bubble skyrmion with a uniformly magnetized core area. The center core area of a skyrmion bubble with magnetization pointing in the positive $z$ direction is shown as red arrows. Magnetization at the boundary area, represented by blue arrows, is along the negative $z$ direction. The transition between magnetization $M_z = 1$ and $M_z = -1$ is the Néel-type domain wall shown in (b) highlighted by the frame with dashed outlines. Cross section of the spin texture marked as a grey dashed line is also presented. (c) Relation of concepts of skyrmion, bubble and bubble skyrmion.
section of magnetic skyrmions and magnetic bubbles. Conventional magnetic bubbles are any circular-shaped magnetic domains, studied in the 1980s for the applications of bubble memory [51]. They do not have any particular defined about topological protection nor the spin configuration but only the shape of the magnetic domain. However, the concept of skyrmions emphasizes on the topological protection and stability. They are ideally circular because circular boundary are energetically favorable. Therefore, Néel-type bubble skyrmions have the advantages of both bubbles and skyrmions with a circular shape domain and a specific topological charge $Q$.

The topological charge $Q$ of a Néel-type skyrmion is $+1$ or $-1$ when the magnetization at the center is along $+z$ or $-z$ direction, respectively, as shown in Figure 2.2(a) and (b). Because $Q$ is proportional to $m(r)^3$, as shown in Equation 2.1, topological charge of the skyrmion in Figure 2.2(b) has an opposite sign to that of (a) when the magnetization is reversed. Also, because the magnetization only wraps around a sphere one time, the magnitude of the topological charge is 1. Note that, the center core area of a bubble skyrmion is uniformly magnetized and will be integrated as zero. Therefore, no matter what the sizes of the core areas are, bubble skyrmions have the same topological charge as the corresponding skyrmion without a uniformly magnetized core.

For Néel-type skyrmions with $Q$ being $+1$, though knowing that the magnetization at the center is pointing up, we still need to define how it rotates to the boundary. $\chi$ defines the fixed chirality, continuously changing in one direction, as either clockwise or counter-clockwise within the $x - z$ plane, as shown in Figure 2.2(a) and (c) or (b) and (d). $\chi = +1$ ($\chi = -1$) represents the counter clockwise (clock wise). The signs of $\chi$ and $Q$ are determined by the sign the DMI factor and the external magnetic field, explained in detail below.

One might wonder why the author is interested in Néel-type skyrmions with $Q = \pm 1$ but not other integer topological charges. This question can be addressed by comparing Néel-type bubble skyrmions with $Q = +1$ and magnetic bubbles with other topological
Figure 2.2: Illustration of Néel-type magnetic skyrmion with $Q = \pm 1$ and $\chi = \pm 1$. Arrows represent atomic spins. Different color corresponds to different net magnetization along the $z$ direction as indicated in the color scale bar. The spins at the center of the skyrmions in (a) and (c) point along the positive $z$ direction indicating the topological charge $Q$ being $+1$ while (b) and (d) are opposite situations. The transition between spin up and spin down is counterclockwise (CCW) for skyrmions in (a) and (b) giving rise to the different sign of chirality $\chi$ comparing to the clockwise (CW) transition in (c) and (d). The CCW and CW transitions are indicated as black arrows in (a) and (c).
integers. To focus on the domain wall areas of these circular-shape magnetic domains, indicating how spins rotate at 180 degrees, the schematics of bubbles with different topological charges are presented as 2 dimensions, as shown in Figure 2.3. As discussed above, the transition between spin up and spin down has a unique chirality; it can be counterclockwise Figure 2.3(a) or clockwise Figure 2.3(b). Clearly, Néel-type skyrmion bubbles with \( Q = +1 \), Figures 2.3(a) and (b), have continuously rotation and a fixed chirality everywhere (indicates as the arrow direction) within the domain areas. They are topologically protected spin configurations that can be well defined. Due to this uniform topology, the interactions between the spins in a skyrmion with other media, such as current and external magnetic fields, is less complicated to study and control. Magnetic bubbles with \( Q = 0 \) shown in Figure 2.3(c) is one example of trivial bubbles. It has a simple spin configuration but is not topologically protected. Magnetic bubbles with \( Q = 0 \) or \( Q = 2 \) shown in Figure 2.3(d), (e) and (f) have complex spin configurations that are not good candidates for studying physics and technology applications.

It should be mentioned that the gyrotropic motion of purely dipolar interaction stabilized magnetic bubbles driven by magnetic field gradients has been extensively studied in insulating yttrium iron garnets [52]. However, due to the lack of a fixed chirality originating from DMI in these materials, a uniform spin topology of magnetic bubbles is absent which, in turn, results in a random transverse motion [52]. In contrast, inversion asymmetric multilayers, such as those studied in the present work, are technologically appealing by hosting chiral skyrmion bubbles with a uniform topological charge that gives rise to a well-defined motion driven by the current-induced spin Hall spin torques [12, 13, 21, 53].

The unique spin texture of skyrmions gives rise to some important properties and one of them is topological protection. Note that, though magnetic skyrmions can be generated or annihilated by currents and magnetic fields, they are topologically protected against a transition into topologically trivial states, such as a ferromagnetic state, by defects or minor perturbations. The transition between different \( Q \) states is topologically forbidden.
Figure 2.3: Differences in topological charge between Néel-type skyrmion bubbles and other circular-shape magnetic domains with the emphasize on the domain wall area. Blue areas indicate uniform magnetization pointing out of the paper plane. Light brown areas are magnetization pointing into paper plane. Red arrows shows the trend of magnetization direction of the domain wall area between blue and light brown areas. (a) and (b) are two different bubble skyrmions with topological charges $Q = 1$ but $\chi = \pm 1$, repsectively. (c) and (d) are two examples of magnetic bubbles with topological charges $Q = 0$. (e) and (f) are two examples of magnetic bubbles with topological charges $Q = 2$. 
because it needs the generation and motion of a micromagnetic singularity to achieve the transition. A micromagnetic singularity can be considered as a pointy spin configuration with topological charge $Q$ being 1. This topological protection of a magnetic skyrmion is physically magnificent because it implies that magnetic skyrmions cannot be deformed continuously into another state with a different topological charge $Q$ without changing the local magnetization. Topological protection is very important in terms of race-track memory application because when a skyrmion moves along a patterned device, which can never be perfect, it will not be affected or annihilated due to the defects in the device.

Another important property of Néel-type skyrmions due to the continuous chiral spin texture is the high efficiency of current-driven motion. In current-driven devices, skyrmions exhibit higher mobility than magnetic domain walls. The critical current density required is $\sim 10^9 - 10^{12}$ A$^{-2}$ to drive domain wall, which is several orders of magnitude larger [4, 5, 21, 30, 54] than that needed for skyrmion motion ($\sim 10^5 - 10^6$ A$^{-2}$). The high mobility of the skyrmions is related to the opposite direction of effective torques at their opposite sides [2, 5, 54] and the resulting gyration of skyrmions, as shown in Figure 2.4. This can be understood by thinking about separating two disk-shape magnets with large surface areas connecting together (similar to the 2D structure of magnetic domains and skyrmions). It is almost impossible to separate them along the opposite direction of the attraction force, perpendicular to the surfaces. However, when applied with a sliding force, within the surface plane, the magnets can be separated more easily. The required lower current density to move/depin skyrmions is analogous to the smaller force in separating the magnets. Therefore, skyrmions are considered as promising candidates for skyrmion-based logic devices and race-track memories [1, 55].

2.3 Nanomagnetism

A magnetic skyrmion reaches static equilibrium, determined by minimizing the magnetic energy in magnetic materials. In general, the total energy in a skyrmion system can
Figure 2.4: Schematic of current-driven skyrmion motion. Arrows represent the atomic spins. Different colors of the arrows correspond to different net magnetization along the $z$ direction as indicated in the color scale bar. Skyrmion move along $+y$ direction as marked by the large and thick blue arrow. The two opposite sides of skyrmions experience opposite direction of forces illustrated as thick green arrows due to the different local spin texture.

be given as

$$E_{\text{total}} = E_{\text{Heisenberg}} + E_{\text{DMI}} + E_{\text{dipolar}} + E_{\text{anisotropy}} + E_{\text{Zeeman}},$$

(2.2)

where $E_{\text{Heisenberg}}$ is the Heisenberg exchange energy, $E_{\text{DMI}}$ is the DMI energy, $E_{\text{dipolar/demag}}$ is the dipolar energy/demagnetizing energy, $E_{\text{anisotropy}}$ is the anisotropy energy and $E_{\text{Zeeman}}$ is the Zeeman energy. To generate, stabilize, and further manipulate Néel-type magnetic skyrmions with an electric current, it is necessary to go into more details about the basic concepts of nanomagnetism. The following sections discuss all the related energies in magnetic multilayers with the interfacial DMI for stabilizing magnetic skyrmions.
2.3.1 Heisenberg exchange interaction

The exchange energy can be explained by Heisenberg model, as described in equation

$$E_{\text{exch}} = -\sum_{i,j} J_{ij} S_i \cdot S_j,$$

where $J_{ij}$ is the exchange integral determined by the overlap of the electronic wavefunctions of the two atoms with spins $S_i$ and $S_j$. The sign of $J_{ij}$ can be positive or negative. If $J_{ij}$ is positive, to lower the total energy, $S_i$ and $S_j$ tend to align parallel to each other so the dot product yields a maximum positive value and the energy $-J_{ij} S_i \cdot S_j$ minimizes. As shown in Figure 2.5, the parallel alignment of magnetic moments results in a ferromagnetic ordering. Similarly, if $J_{ij}$ is negative, $S_i$ and $S_j$ tend to align antiparallel to each other and results in an antiferromagnetic ordering.

Fe, Co, and Gd are the materials of interest in this dissertation work, and they have all ferromagnetic exchange interaction within themselves. However, the interaction between Gd and Co is antiferromagnetic, which will be discussed in detail in Chapter 6.

2.3.2 Interfacial Dzyaloshinskii-Moriya Interaction

The key to stabilizing the chiral structure of magnetic skyrmions is the Dzyaloshinskii-Moriya Interaction (DMI), which is relevant in material systems exhibiting large spin-orbit coupling and lacking inversion symmetry [15]. DMI is an antisymmetric exchange interaction. First postulated by Igor Dzyaloshinskii in 1958 [15], DMI was then studied in
Figure 2.6: Mechanism for an interfacial DMI and spins at an interface of magnetic multilayers. Diagram redrawn in the style of Figure 1 in Nature Nanotechnology 8, 152-156 (2013). Spins $S_i$ and $S_j$ in the ferromagnetic (gray, upper) layer couple to each other through overlap of their wave functions with an atom with large spin-orbit coupling (blue, lower layer). This overlap gives rise to a contribution to the energy of the form $D_{ij} \cdot (S_i \times S_j)$, where $D_{ij}$ lies in the plane of the interface, in the direction normal to the plane defined by the three atoms [57]. The dark grey lines between atoms indicates the plane defined by the three atoms.

more detail by Toru Moriya in 1960 [56], who identified the microscopic mechanism of the interaction. It is a super-exchange interaction between two magnetic spins in a system that needs a third non-magnetic atom to realize the exchange interaction. Even though it is smaller than direct exchange interaction, DMI has gained great research interest recently because it is crucial for forming and stabilizing magnetic skyrmions.

The interfacial DMI between two atomic spins $S_i$ and $S_j$ in the magnetic materials in contact with a non-magnetic atom can be expressed as

$$E_{DMI} = \sum_{i,j,i\neq j} D_{ij} \cdot (S_i \times S_j),$$

where $D_{ij}$ is the DMI vector and $S_i$ and $S_j$ are the spins of atom $i$ and $j$, as illustrated in Figure 2.6 [15]. The interfacial DMI vector $D_{ij}$ lies in the plane of two adjacent layers, and perpendicular to the triangle defined by the three-site exchange between $S_i$ and $S_j$ and neighboring atom, as shown in Figure 2.6.

The mechanism behind the changing of the spin canting direction is shown in detail in
Figure 2.7. When $D_{ij}$ points out-of-paper-plane and $S_i$ lies within the paper plane pointing from up to down, the adjacent $S_j$ will tilt counterclockwise, which can be understood easily by analyzing the equation. To minimize the $E_{\text{DMI}(ij)}$, $S_i \times S_j$ should be in the direction that is opposite to $D_{ij}$. Knowing the direction of $S_i$, and the direction of $S_i \times S_j$, $S_j$ can be figured using the right hand rule. Therefore, the direction of DMI vector determines the adjacent spin canting direction, domain wall transition direction and the chirality of magnetic skyrmions, giving rise to skyrmions with different chiralities shown in Figure 2.2.

In contrast to Heisenberg exchange interaction, which tends to align the spins in parallel or antiparallel, DMI energy is minimized if $S_i$ and $S_j$ are perpendicular to each other due to the cross product. This is the reason that DMI is the key in forming and stabilizing the swirling spin textures with a fixed chirality. Notice that, the magnitude of DMI vector also affects the strength of spin canting. The larger the DMI vector is, the more the adjacent spin tilts. Since Néel-type domain wall of a magnetic skyrmion is the finite region where spin rotates from pointing up (down) to sown (up), a larger DMI vector will result in a smaller domain wall width.

In order for the interfacial DMI to exist in a magnetic system, two conditions need to be fulfilled: large spin-orbit coupling in non-magnetic atoms and a broken inversion symmetry [7, 8, 9]. The spin-orbit coupling in an atom is the interaction between a single electron’s intrinsic spin and its orbiting motion, which functions as an effective magnetic field and can be described by the equation:

$$E_{\text{SOC}} \propto Z^4 \cdot S \cdot L,$$  \hspace{1cm} (2.5)

where $Z$ corresponds to the atomic number, $S$, and $L$ are the spin angular momentum of an electron and its angular momentum around the nucleus. Therefore, the spin-orbit coupling is stronger in heavy metals with larger $Z$. Understandably, the electron in the non-
\[ E_{\text{DMI}(ij)} = \vec{D}_{ij} \cdot (\vec{S}_i \times \vec{S}_j) \]

Figure 2.7: Mechanism of the relationship between the direction of \( D_{ij} \) and spin canting direction. When the interfacial DMI vector \( D_{ij} \) points out (into) of the paper plane, spin cants in counterclockwise (clockwise) direction.

magnetic atom needs to interact with both of the electrons in the magnetic atoms. During spin and orbital motion of the electron in the non-magnetic atom, the stronger the spin-orbit coupling is, the better it can serve as the intermedia in passing the interaction from one magnetic spin to another. When the DMI is strong enough to save energy by forming a canting/swirling spin structure, in combat with the Heisenberg exchange interaction, the neighboring spins tend to align perpendicular instead of parallel/antiparallel. However, the detailed correlation between the spin-orbit coupling and the magnitude of DMI remains debatable to this date.

Another necessary factor for the existence of DMI is the broken inversion symmetry [15]. Due to the reduction in symmetry near the surface, the antisymmetry exchange is enhanced at an interface. Therefore, a magnetic layer/heavy metal interface with large spin-orbit coupling can fulfill all DMI requirements and can exhibit a strong DMI for stabilizing magnetic skyrmions, theoretically. One restriction for these magnetic layer/heavy
metal interfaces is that the layers have to be extremely thin. The interfacial DMI decays to almost zero after three or four atomic layers because of the short-range nature of exchange interactions. For the magnetic spins that are a few Å away from the interface with the non-magnetic layer, the electrons do not have the broken inversion symmetry and can barely interact with the non-magnetic atoms.

The key reason behind the uniform topology spin configuration of Néel-type bubble skyrmions can be attributed to the existence of interfacial DMI in the system. The swirling in the magnetization profile of a skyrmion leads to a decrease in energy with respect to a homogeneously magnetized due to the interfacial DMI [15, 56]. The stabilization of magnetic bubbles, circular uniformly magnetized domain, doesn’t require DMI. The long-range dipolar interaction, discussed below can serve as the key in stabilizing magnetic bubbles. However, without the interfacial DMI, the non-linear spin configuration does not have a fixed chirality resulting in different numbers of topological charges.

2.3.3  Dipolar Interaction

Dipolar interaction is the interaction between two magnetic dipoles \( \mathbf{m}_i \) and \( \mathbf{m}_j \). It can be described by

\[
E_{\text{dipolar}} = -\sum_{i,j,i\neq j} \frac{\mu_0}{4\pi|\mathbf{r}|^3} \left( 3 \left( \mathbf{m}_i \cdot \hat{\mathbf{r}} \right) \left( \mathbf{m}_j \cdot \hat{\mathbf{r}} \right) - \mathbf{m}_i \cdot \mathbf{m}_j \right),
\]

where \( \mu_0 \) is the magnetic permeability in vacuum, \( \hat{\mathbf{r}} \) is the unit vector parallel to the line connecting the centers of the two dipoles, and \( |\mathbf{r}| \) corresponds to the distance between the centers of \( \mathbf{m}_i \) and \( \mathbf{m}_j \). \( E_{\text{dipolar}} \) is proportional to the inverse cube of inter nuclear distance and decreases as the distances increases.

Unlike exchange interactions discussed above, dipolar interaction is a long-range interaction, favors breaking the large area of the magnetic domain, an area of magnetic materials with the same magnetization, and helps forming magnetic flux-closure domains with
smaller areas [58], shown as Figure 2.8. Magnetic domains can have different sizes and shapes, for example circular, labyrinth and stripe. Both theory and experiments suggest that these patterns appear in distinct sequences as a function of temperature or film thickness [58]. Therefore, the dipolar interaction is important in stabilizing different sizes and shapes of magnetic domains such as circular-shape (bubble) domains.

The bubble domains stabilized by the dipolar interaction have random-oriented domain walls. Examples can be found in Figure 2.3(c)-(f). As discussed in section 2.3.2, the interfacial DMI is the key in forming domain walls with a fixed canting/swirling direction (chirality). Without DMI, the transition of the spins are random and do not have a fixed chirality. Because the chiral domain walls with the fixed chirality are the source of the topological properties, these magnetic domains stabilized by the dipolar interaction do not always have integer topological charges.

However, considering the fact that the dipolar interaction helps stabilizing circular-shape magnetic domains, the combination of the dipolar interaction and interfacial DMI has great potential in realizing and stabilizing Néel-type skyrmion. This hypothesis has already attracted research interest. Part of this thesis work (Chapter 4) investigates the effect of DMI and the dipolar interaction on the stabilization of magnetic skyrmions bubbles in multilayers.
2.3.4 Magnetic Anisotropy

Magnetic anisotropy determines the preferential direction of magnetization in a magnetic material. Owing to the reduced symmetry at the surface or interface, the magnetic anisotropy in magnetic multilayers is usually within the surface of the film. However, by tuning the magnetocrystalline direction at the interfaces and the thickness of different layers, the preferential direction of the magnetization can be changed from the commonly observed in-plane orientation to the perpendicular direction, achieving perpendicular magnetic anisotropy (PMA). For example, Pt deposited on Ta will have the crystalline direction along $z$ axis. By depositing an ultra-thin layer of Co (a few Co atoms along $z$ axis), Pt/Co interface will form an out-of-plane magnetic anisotropy.

In magnetic multilayers with the interfacial DMI, PMA is required for the system to potentially host magnetic skyrmions. As discussed in section 2.3.2, the broken inversion symmetry is along the out-of-plane direction and the direction of DMI is within the plane of the interfaces. To minimize the dot product of $D_{ij}$ and $S_i \times S_j$, shown in Equation (2.4), the direction of $S_i \times S_j$ should be antiparallel to the direction of $D_{ij}$. That is to say the plane of $S_i$ and $S_j$ should be perpendicular to the direction of $D_{ij}$. Therefore, the spin directions of the magnetic atoms are required to be perpendicular to the interface. In other words, PMA plays a role in stabilizing magnetic skyrmions in multilayers with the interfacial DMI.

2.3.5 Zeeman energy

$E_{\text{Zeeman}}$ is the potential energy of a magnetized volume in an external magnetic field, it can be described as

$$E_{\text{Zeeman}} = -\mu_0 \int_V \mathbf{M} \cdot \mathbf{H}_{\text{Ext}} \, dV,$$

(2.7)

where $\mathbf{M}$ is the local magnetization, $\mathbf{H}_{\text{Ext}}$ is the external field, and the integral is done over the magnetized volume. For simplification, the energy can be described as $E_{\text{Zeeman}} =$
Figure 2.9: The effect of external magnetic field on topological charge of a skyrmion. Different colors correspond to different net magnetization along the $z$ direction as indicated in the color scale bar. (a) When the external magnetic field points at positive $z$ direction, the spins at the boundary of a magnetic skyrmion point at positive $z$ direction giving rise to a negative topological charge. (b) The direction of spins and the sign of topological charge reverse when the direction of the external magnetic field reverses.

$MH \cos \theta$ where $\theta$ is the angle between $\mathbf{M}$ and $\mathbf{H}$. $E_{\text{Zeeman}}$ depends only on the average magnetization and not on the particular domain structure or the sample shape. To lower the Zeeman energy, $\mathbf{M}$ tends to align in the same direction as $\mathbf{H}_{\text{Ext}}$ with $\theta$ being zero.

Magnetic skyrmions usually are not the magnetic spin textures at ground state in multilayers with the interfacial DMI. An out-of-plane external magnetic field is often required to stabilize skyrmions. To lower the Zeeman energy with the existence of an external magnetic field, magnetization at the boundary of a magnetic skyrmion is aligned in the same direction as the external magnetic fields, as shown in Figure 2.9. This can be understood by considering a skyrmion as a particle. The magnetization at the boundary is closest to the external field comparing to the center. Hence, the magnetization at the boundary is aligned in the same direction as the external field and the magnetization in the center results to be in the opposite direction. In this case, the direction of local magnetization can be reversed when the direction of the external magnetic field is reversed. Furthermore, the sign of the topological charge of the magnetic skyrmions can be changed. This is because the topolog-
Figure 2.10: Magnetic multilayers with tunable parameters and the associated properties.

The charge $Q = 1/4\pi \int \mathbf{m} \cdot (\partial_x \mathbf{m} \times \partial_y \mathbf{m}) \, dxdy$ is an odd function of the magnetization vector $\mathbf{m}$.

### 2.4 Magnetic multilayers

To experimentally study the rich physics and realize the applications of magnetic skyrmions for future technology, one ultimately needs a system that is easy to fabricate and able to host nanoscale magnetic skyrmions without a large external magnetic field or low temperatures. With the knowledge of the fundamental concepts about magnetic skyrmions and the nanomagnetism, we consider magnetic multilayers with the interfacial DMI as good candidates for four reasons listed below. First, magnetic skyrmions have been experimentally observed at room temperature in multilayers with the interfacial DMI [10, 11, 12, 13]. Second, multilayers can be deposited using sputtering, a technique that is commonly used in industry for thin film deposition. Third, the interfacial DMI, crucial for stabilizing Néel-type skyrmions, exists at the interfaces of multilayers [6, 12, 13, 14]. Finally, mag-
magnetic multilayers can be engineered with tunable parameters such as thickness, elements, interfaces, and repetition numbers, resulting in different magnetic properties, as shown in Figure 2.10. Multilayer systems can have a different thickness ranging from 0.1 to a few hundred Å, where 0.1 Å difference can enable different magnetic properties [59, 19], such as magnetic anisotropy and dipolar interaction. By choosing different ferromagnetic layers, the damping (relates to the energy wasted in current-driven skyrmion motion), magnetization, and anisotropy can be changed. Different interfaces between heavy metal (HM) and the ferromagnetic layer can induce different signs and amplitudes of the interfacial DMI. The different repetition number $n$ gives rise to different strength of the dipolar interaction.

With all the tunable parameters, one can engineer the system to shrink the size of the currently observed bubble skyrmions from $\mu$m to nm, study the interfacial DMI and further investigate the nanomagnetism behind skyrmion and its current driven motion. Therefore, Néel-type skyrmions in multilayers with the interfacial DMI are the interest of this thesis work.

**References of Chapter 2**


CHAPTER 3

EXPERIMENTAL METHODS OF SAMPLE FABRICATION AND CHARACTERIZATION

3.1 Introduction

This chapter describes the main relevant experimental methods used in fabricating and charactering the magnetic multilayers. First, section 3.2 presents the methods related to fabrication of multilayer thin films. Then, the deposited multilayers were patterned into Hall bar devices for current-driven measurements by photolithography, explained in section 3.3. After samples were prepared, the magnetic properties were characterized using methods discussed in section 3.4, and the magnetic domains were imaged by different magnetic imaging methods described in section 3.5.

3.2 Fabrication of Multilayer Thin Films

3.2.1 Sputtering Deposition

Sputtering deposition is a commonly used technique to grow thin films in the study of nanomagnetic materials. In sputtering deposition, the target atoms are ejected into gas phase by incident energetic ions and then deposited onto a substrate to form a film. The schematic view of sputtering deposition is illustrated in Figure 3.1.

Before sputtering occurs, the chamber is initially pumped down to high or ultra-high vacuum pressure, with a typical base pressure of $0.1 - 9.9 \times 10^{-8}$ Torr, by the subsequent use of a roughing pump and a high vacuum pump [59]. The high base pressure is very important because it avoids surface contamination of sample films by residual gas molecules. For depositing the films studied in this dissertation work, the base pressure of the sputtering was $\sim 5 \times 10^{-8}$ Torr.
Then argon (Ar) gas of a controlled flow, 30 standard cubic centimeters per minute (sccm) in this thesis work, was sent into the chamber, so the chamber pressure dropped. By adjusting the valve between the pump and sputtering chamber, the pressure was adjusted to 5 mTorr for deposition. This deposition pressure is a very important parameter because it controls the average gas density, the chances of energy loss of particles through collision and the average mean free path of particles [59]. Within this stable deposition pressure, Ar gas was ionized by stray electrons, formed a stable gaseous plasma after a voltage was applied, as illustrated in Figure 3.1. The Ar$^+$ ions in the plasma were then accelerated by an electric field towards the negatively charged target, bombarding and ejecting neutral atoms from the target. The target atoms were transported through the Ar gas and plasma environment, before finally depositing onto the substrate, where they formed a layer of relatively uniform thickness.

Multilayers with three different structures were deposited and characterized to the interest of this thesis, and their structures are shown in Figure 3.2 with the sputtering rates for different metals shown in Figure 3.3. In the first multilayer film, as shown in Figure 3.2(a), 1.0 nm Co sandwiched by 1.5 nm Pt and 1.0 nm heavy metal (HM) forms one repeat and the repeat number $n$ varies across different samples as shown in Figure 3.2(a). Different HMs, such as Ir, Au, W, and Mn, were used to tune the strength of the DMI. Different repetition numbers ($n$) of the sandwich structure were used to tune the dipolar interaction. The multilayers are deposited on Si/SiO$_2$ substrates and transmission electron microscopy
Figure 3.2: Illustration of multilayered structures. (a), (b) and (c) are structures of multilayered samples studied in this thesis project in Chapters 4, 5 and 6 respectively.

<table>
<thead>
<tr>
<th>Element</th>
<th>Ir</th>
<th>Au</th>
<th>W</th>
<th>Co</th>
<th>Pt</th>
<th>Ta</th>
<th>Gd</th>
<th>Al</th>
<th>Mn</th>
</tr>
</thead>
<tbody>
<tr>
<td>Growth Rate (Å/s)</td>
<td>0.4</td>
<td>0.4</td>
<td>0.2</td>
<td>0.2</td>
<td>0.5</td>
<td>0.4</td>
<td>0.2</td>
<td>0.4</td>
<td>0.4</td>
</tr>
</tbody>
</table>

Figure 3.3: Growth rate in sputtering deposition of elements in the multilayer film samples measured using crystal oscillator thickness monitor.
(TEM) grid with a 50 nm thick silicon nitride (SiN) membrane window for magneto-optical Kerr effect (MOKE) microscopy and Lorentz-TEM measurements, respectively. The bottom 2 nm Ta was grown as a seeding layer. In second multilayer film, as shown in Figure 3.2(b), Ta (50Å) /Co_{20}Fe_{60}B_{20} (CoFeB) (11Å) /TaO_{x} (30Å) trilayer was grown onto a Si substrate with 300-nm-thick thermally formed SiO_{2} layer. To prepare the TaO_{x} layer, one would oxidize the top Ta layer with an oxygen plasma of ten W for sixty seconds and then anneal the trilayers in a vacuum for around thirty minutes to induce perpendicular magnetic anisotropy. To generate the antiferromagnetically coupled skyrmion pairs, engineered films Ta (5 nm) /Pt (4 nm) /[Co (0.5 nm) /Gd (1.0 nm) /Pt (1.0 nm)]n=10 /Al (2.0 nm) were deposited on Si/SiO_{2} (300 nm). In the structure of this sample, a unit of tri-layer [Pt (1.0 nm) /Gd (1.0 nm) /Co (0.5 nm)] was repeated 10 times on top of Ta (5.0 nm) /Pt (4.0 nm) seeding layers, as shown in Figure 3.2(c). 2.0 nm Al is used to protect the sample from being oxidized and allow x-ray penetrating the film when performing x-ray spectroscopy and microscopy measurements.

3.2.2 Thickness Characterization

The thickness information of the multilayers or thin films can be obtained according to Bragg’s law from the peak positions of the small-angle reflectivity scan [59]. Bragg’s law describes the condition for the constructive interference and the angle θ where diffraction peaks form, as shown in Figure 3.4

\[ n\lambda = 2t \sin \theta, \tag{3.1} \]

where \( n \) is the diffraction order, \( \lambda \) is the wavelength of x-ray, \( t \) is the thickness of a layer of thin film or the total thickness of the multilayer, and \( 2\theta \) is the angle between the diffracted and incident x-ray beams [59].

The small-angle reflectivity scan measures the intensity of the diffracted x-ray beam
Figure 3.4: Schematic of x-ray diffraction in a thin film with thickness $t$.

Figure 3.5: X-ray small-angle reflectivity results and fitting results of multilayered films: (a) $[\text{Pt/Co/W}]_{n=8}$ (c) $[\text{Pt/Co/Ir}]_{n=8}$ and (d) $[\text{Co/Gd/Pt}]_{n=10}$. (b) The fitted results of the thickness and roughness of each layer of the results in (a).
with a small incident x-ray angle $\theta$, usually between 0.2 to 5 degrees. The results of different multilayers studied in this dissertation are shown in Figure 3.5. The thickness can be calculated based on the periodicity of the peaks. The fitting using software Gen-X yields the roughness ($\Delta \ t$), and the thickness ($t$) of each individual layer, which is consistent with the expected nominal thicknesses ($t_n$), as summarized in Figure 3.5 (b).

### 3.3 Device Patterning by Photolithography

To conduct the current-driven skyrmion motion experiments as a function of current density, Hall bar devices, as shown in Figure 3.6 were patterned on the multilayer films. The solid blue regions are the film, and the dotted green areas are the electrodes for connecting the wires. The devices have a length of 60-500 $\mu$m and a width of 20-150 $\mu$m.

Laser writer photolithography was used to pattern the devices. The process flow for device patterning is depicted in Figure 3.7. First, the positive photoresist is spin-coated on the surface of a deposited film, 1813 used in this dissertation work. The UV light from the laser writer scans the material in the white areas with yellow dash edges. The area of posi-
Figure 3.7: Illustration of photolithography and ion milling procedures for patterning devices.

Positive photoresist that are exposed to the scanning laser beam has different chemical structure than the unexposed area and can be dissolved by the developer solvent (solvent 351: DI water = 1:4), exposing the underneath materials. The un-exposed areas are therefore left insoluble to the developer solvent, to protect the underneath magnetic film from being etched by ion milling, which is a physical etching process utilizing accelerating Argon atoms at the surface of a sample in order to physically remove atoms from the surface. After the ion milling, a remover (solvent 1165 for example) was used to clean the positive photoresist on top of the magnetic film, and the patterned devices will be achieved.

Several photoresist exposures and development steps are required to pattern the metal
contact structures, marked as dotted green areas in Figure 3.6. Still, the positive photoresist is used, so the area that is going to be exposed by laser writer can be dissolved after development. 20 nm of Cr and 80 nm of Au films are then deposited onto the substrate, which is then followed by a lift-off process. During the lift-off procedure, the substrate is submerged in acetone for 24 hours in order to dissolve the photoresist. The gold film that was deposited directly on the photoresist is removed, leaving the gold film that was deposited directly onto the substrate as contacts. The metal contacts are constructed. After performing wire bonding, electric current can be injected into Hall bars for studying current-driven skyrmion motion.

3.4 Magnetic Property Characterization

3.4.1 Magnetometry

Magnetometry studies the magnetic properties of materials as a function of magnetic field and temperature. A magnetometer is often used to conduct the measurements, which detects the change in magnetic flux due to the motion of a magnetic sample [59]. The two most common ones, the Vibrating Sample Magnetometry (VSM) and Superconducting Quantum Interference Device Magnetometry (SQUID) were utilized to study the magnetic properties of the multilayers studied in this dissertation.

The mechanism of detecting the change in magnetic flux slightly varies for different magnetometers, giving rise to different sensitivity. The VSM measures the magnetization of the sample based on detecting the voltage induced in the detection coils when the magnetic sample is vibrating. SQUID can measure magnetic signal as low as $10^{-8}$ emu, which is 3 order of magnitude more sensitive than a VSM due to the SQUID sensor [59].

Both VSM and SQUID can measure magnetic properties of samples at cryogenic temperatures and room temperature and above. However, the range of the magnetic fields that can applied to the measured sample are different. The magnet in a VSM is a regular electromagnet with a giant permanent magnet core which cools by cooling water. On the other
hand, the magnet in a SQUID is a superconducting magnet which usually achieves 5-6 Tesla magnetic field, much higher than the 2 Tesla capability of a VSM.

VSM and SQUID are very common methods in characterizing the magnetic properties of the whole sample. However, for a magnetic system with more than one magnetic components contributing to the total magnetization, element specific X-ray magnetic circular dichroism spectra is the best method.

3.4.2 X-ray Magnetic Circular Dichroism Spectra

X-ray magnetic circular dichroism (XMCD) spectra measure the difference between the absorption of left circularly polarized (LCP) and right circularly polarized (RCP) x-rays of a magnetic sample [60, 61, 62, 63]. Typical x-ray absorption and XMCD spectra of a Co metal are shown in Figure 3.8(a). In XMCD measurements, core electrons are excited by the LCP and RCP x-rays to the empty states in valence band above the Fermi level to probe magnetic properties, as illustrated in Figure 3.8(b). When excited by a polarized x-ray beam, core electrons absorb not only the energy but also the angular momentum of the x-ray photons.

Depending on the energy difference between the core electron level and the empty state in the valence band, photons with different energies that correspond to specific allowed energy differences can be absorbed by the core electrons. For example, as shown in Figure 3.8(a), the $L_3$ edge absorption of Co, corresponding to electron excitation from $2p_{3/2}$ level (core level) to 3d level (valence band), is maximized at energy $\sim 778$ eV while the $L_2$ edge, corresponding to electron excitation from $2p_{1/2}$ level (core level) to 3d level (valence band) is maximized at a higher energy $\sim 795$ eV. Those absorption energies vary for different elements. Therefore, the x-ray absorption and XMCD spectra are element-specific.

The magnetic moment of an atom originates from the unbalanced numbers of spin-up and spin-down electrons, which can be probed through the difference in the absorption of x-rays with different angular momentum (LCP and RCP). During excitation, angular
momentum of the x-ray photons can be transferred to electrons through spin-orbit coupling, following the selection rules. As shown in Figure 3.8(b), there are more available states for spin-up electrons than spin-down electrons above Fermi energy; therefore, there is a higher chance to excite electrons to spin-up states. As a result, the amount of the LCP and RCP x-rays absorbed by core electrons is different. Simply, because valence states can host more spin-up electrons, the x-ray photons with a specific circular polarization that can transfer core electrons into the spin-up states are absorbed more than the photons the other circular polarization, giving rise to the XMCD intensity. Moreover, the spin moment and orbital moment can be determined from the XMCD results according to sum rules, as discussed in Appendix [64, 65, 66].

All the XMCD experiments discussed in the thesis were performed at beamline 4 ID-C of the Advanced Photon Source (APS) at Argonne National Laboratory (ANL).

3.5 Magnetic Imaging Methods

3.5.1 X-ray Photoemission Electron Microscopy

X-ray photoemission electron microscopy (PEEM) forms images of electrons emitted from a sample surface, excited by circularly polarized x-ray, accelerated by a high voltage and focused with a set of electron lenses [60]. A magnetic PEEM image that shows the magnetization distribution in an area is formed similar to XMCD spectroscopy, by taking the difference of two images excited by LCP and RCP x-rays at certain energies. An example of a PEEM image of a vortex is shown as Figure 3.9. XMCD spectra was first taken to determine the photon energy that has the largest XMCD intensity. $L_3$ peak usually is the energy gives rise to the largest signal. When the energy of x-ray is tuned to Ni $L_3$ peak, a image excited by LCP photon, shown as a picture with the blue frame, is subtracted by a image excited by RCP photon, shown as picture in red frame. The resulting image corresponds to the magnetic configuration of the area being imaged.

PEEM is sensitive to the in-plane magnetization. The x-ray propagation direction is
Figure 3.8: Schematic of XMCD spectra at $L_2$ and $L_3$ edges. (a) X-ray absorption spectra excited by LCP and RCP x-rays as well as the XMCD spectrum of Co metal at $L_2$ and $L_3$ edges. (b) Mechanism of XMCD. $2p_{3/2}$ and $2p_{1/2}$ are two different $2p$ levels (core level) with different spin-orbit couplings. Blue and Green electrons correspond to spin-up and spin-down electrons in the $2p$ states. Photons with LCP or RCP angular momentum, shown in blue or red circular arrows, excite electrons from core level to valence band above Fermi energy, corresponding to the absorption spectra in blue and red in (a). Electrons in $3d$ level are filled with spin-up and spin-down electrons at energy below Fermi energy $E_F$, shown as blue and green area respectively. Empty states in $3d$ level with energy above Fermi energy, shown as white area are the valence states for electrons excited from core level $2p$. 
Figure 3.9: Illustration of PEEM imaging experiment set-up and the mechanism. Left panel shows x-ray absorption spectra and XMCD spectrum of Ni $L_{2,3}$ edges. The dashed line marks the energy where XMCD has the highest intensity. PEEM images correspond to the XMCD contrast by taking the difference of an image taking with a LCP and RCP x-ray. X-ray incident angle is $30^\circ$. The contrast showing in the PEEM image is the magnetization of Ni projected along the x-ray propagation direction. The red arrows in the vortex image indicate the in-plane magnetization direction within the vortex. PEEM images are taken at beamline 4-ID-C of the APS at ANL.
always perpendicular to the set of electron lenses due to the instrumentation. To make sure x-ray is incident onto the sample surface and the emitted electrons go through the electron lenses, there is usually a small angle (less than 30°) between x-ray direction and sample surface. Because XMCD intensity is proportional to the magnetization along the x-ray prorogation direction, the contrast difference in the PEEM image indicates the intensity of magnetization along the direction of x-ray. That is to say the brightest/white (darkest/black) contrast indicates the magnetization is along the same (opposite) direction as x-ray. The grey region is where the magnetization perpendicular to the x-ray direction. Since x-ray is almost along instead of perpendicular to the surface, PEEM images probe mainly the in-plane magnetization.

Because XMCD is element-specific and PEEM images are based on XMCD, PEEM images are element specific as well, meaning it can take images for different elements in a magnetic system. For example, when imaging the magnetization of a Py (FeNi) disk, the energy can be tuned to Ni $L_3$ peak or Fe $L_3$ peak to probe the magnetization for different elements in the sample. This is very useful in studying samples with multiple resources of magnetization to investigate the interaction among different elements.

All the PEEM images presented in Chapter 6 of this thesis were taken at beamline 11.0.1 of the Advanced Light Source (ALS) at Lawrence Berkeley National Laboratory (LBNL).

3.5.2 Lorentz -Transmission Electron Microscopy

Lorentz -transmission electron microscopy (L-TEM) is a powerful tool for magnetic imaging with high spatial resolution. Different from conventional transmission electron microscopy, L-TEM utilizes the deflection of the electron beams due to Lorentz force to image the magnetic configurations of the specimen. Lorentz force ($F_L$) is well-known as $F_L = -e[E + (v \times B)]$, with $E$, $B$ being the electric and magnetic field in the specimen, respectively and $e$, $v$ being the electron charge and velocity, respectively. For electron
Figure 3.10: Simulations of Lorentz TEM image assuming different spin textures. (a) Simulated Lorentz TEM images of Néel-type magnetic skyrmions with spins pointing towards [a-(1)] or out from [a-(2)] the core direction. a-(3-4): sample tilted by $\phi = -30^\circ$, (a)-(5) and (a)-(6): no sample tilt $\phi = 0^\circ$, and a-(7-8): sample tilted by $\phi = +30^\circ$. $x$ axis is the rotation axis indicated by the black arrow and the rotation direction is illustrated by the red arrow. Magnetic contrast is only visible when the sample is tilted, but the configurations seen in [a-(1)] and [a-(2)] appear indistinguishable, as shown in the intensity line profiles seen in a-(9). (b) Simulated Lorentz TEM images of topologically-trivial bubbles with spins pointing to the left or to the right: b-(1) and b-(2) respectively. Sample tilt provides further contrast between the different spin configurations to be distinguished.
beams incoming perpendicular to the plane of the specimen with perpendicular magnetic anisotropy, the deflection of the electron beams due to Lorentz force is zero when \( v \) is parallel or antiparallel to \( B \) (out-of-plane magnetization) and maximized when \( v \) is perpendicular to \( B \) (in-plane magnetization). Therefore, usually, L-TEM is best for imaging in-plane magnetization and cannot image the out-of-plane component. However, to indirectly resolve partial information of the out-of-plane magnetization of the specimen using L-TEM, one can tilt the sample to introduce an in-plane component for imaging. Moreover, by introducing under-focus Fresnel imaging mode, domain walls, the transition between two uniformly magnetized regions, can be revealed as lines of black or white contrast depending on the transition direction of the spins within the domain walls.

In order to investigate Néel-type skyrmions in multilayers using L-TEM, the effects on the magnetic contrast with and without tilting the sample with respect to the incident electron beam was first simulated, and the results are shown in Figure 3.10 with three different tilting angles \( \phi = -30^\circ \), \( \phi = 0^\circ \), and \( \phi = +30^\circ \) [67]. The tilting axis is along the horizontal direction (black arrow) and the rotation direction is marked as the red arrow in the schematics. Néel-type skyrmions with opposite spin chirality are shown in Figure 3.10(a)-(1-2). Note that the sign of interfacial DMI determines the chirality spin structures. At zero tilting angle, the result shows no visible magnetic contrast. This result matches our expectations because the perpendicular magnetization is parallel or antiparallel to the electron beams’ velocity. For the tilted configurations, the introduced contrast is due to the in-plane projection of the perpendicular magnetization from regimes core area indicated as blue and outside area indicated as light brown, respectively. As a result, the black contrast is switched to white when the tilting angle is reversed. These simulations show that one could detect Néel-type skyrmions with L-TEM if the sample is tilted. The intensity plot of the Néel-type skyrmions with the opposite chiralities along the diagonal direction are very similar, as shown in Figure 3.10.

To distinguish the L-TEM images of skyrmion and trivial magnetic bubbles, simulation
of L-TEM images of magnetic bubbles with \( Q = 0 \) were also conducted and illustrated in Figure 3.10(a)-(1-2), corresponding to spin vectors that point towards either the right or left sides, respectively. The zero-tilt image shows a magnetic contrast at the domain wall location, with alternate black and white regimes, which is distinctively different from that of Néel-type skyrmions. The upper half of the trivial bubble has white on top of the black, and the same is repeated for the lower half of the bubble domain. Tilting of the sample also significantly affects the magnetic contrast, and introduces an asymmetry depending on the tilting direction, as shown in Figure 3.10(b)-(3-8). This magnetic contrast is like turbulence showing a swirling structure, very different from that of Néel-type. Again, the intensity along the diagonal direction is provided in Figure 3.10(b)-(9), from which one can distinguish two different cases of \( Q = 0 \) topologically trivial bubbles, with the spins pointing to either the right or left sides, respectively. Therefore, L-TEM can distinguish the magnetic configuration from Néel-type skyrmions to trivial magnetic bubbles.

A FEI Tecnai F20ST TEM/STEM system in Dr. Amanda K. Petford Long’s group at the Materials Science Division at ANL was used to take all the L-TEM images reported in this dissertation work.

### 3.5.3 Magneto-optical Kerr Effect Microscopy

Magneto-optical Kerr effect microscopy is a method in imaging magnetic domains based on magneto-optical Kerr effect (MOKE) as a contrast mechanism. When a beam of incident polarized light interacts with the magnetic material, the polarization state of the reflected light will change. The change in the polarization state or intensity is proportional to the magnetization in the material.

In fact, MOKE microscopy is an extension of a polarization contrast optical microscope and allows to probe the local magnetic properties of ferromagnetic materials [68]. To image the magnetic configuration in a sample, as shown in Figure 3.11, a polarized light passes through a beam splitter with half shining on the surface of a magnetic sample. Due to the
MOKE effect, the polarization of the reflected light is changed. After the original polarized light and the reflected polarized light go through the analyzer together, the change of the polarization of the reflected light will be illustrated as the magnetization of the sample collected by a CCD camera.

MOKE microscopy requires minimum sample preparation and does not require vacuum or sophisticated scanning mechanism. It can easily combine with an electromagnet and allow applying a large magnetic field to magnetize the sample. Comparing to PEEM and L-TEM, MOKE microscopy is a simpler instrument, does not require complex sample preparation, and allows measurements in a large external magnetic field. These advantages make MOKE microscopy the better choice in studying magnetic domains, especially the dynamics involving sending electric currents and changing external magnetic fields.

Due to the nature of an optical microscope, the resolution of MOKE microscopy has
the limitation of the wavelength of visible lights. It is very hard to image magnetic domains with size smaller than 1 \( \mu \text{m} \). L-TEM and PEEM as the electron microscopy, have much higher resolution due to the small wavelength of an electron. Therefore, when imaging magnetic domains in nm scale, L-TEM and PEEM microscopy are used. Note that, the element-specific feature of XMCD makes PEEM the best choice in distinguish the different magnetic components in a magnetic sample.

References of Chapter 3


CHAPTER 4
MAGNETIC SKYRMIONS IN [Pt/Co/HM]_n MULTILAYERS WITH INTERFACIAL DMI

4.1 Motivation and Experiments

The formation and stabilization of magnetic skyrmions without external magnetic fields are determined by the competition of the Heisenberg exchange interaction, the interfacial DMI, the dipolar interaction and magnetic anisotropy. To realize nano scale skyrmions at room temperature and zero field for technology applications, such as data storage, one needs to understand the effects of different interactions on stabilizing and tailoring skyrmions. This chapter presents the study of the effects of interfacial DMI and the dipolar interaction on the stabilizing and tailoring the size of skyrmion.

We specifically engineered multilayers Ta (2.0nm) /[Pt (1.5nm) /Co (1.0nm) /HM (1.0nm)]_n /Pt (2.0nm) (HM stands for heavy metal and n stands for the repetition number), as shown in Figure 4.1, and deposited these films on Si/SiO$_2$ substrates and transmission electron microscopy (TEM) grid substrates to directly image the stabilized magnetic configurations using magneto-optical Kerr effect (MOKE) microscopy and Lorentz-TEM (L-TEM). Multilayers were magnetized in an in-plane magnetic field between 0.6 Tesla to 1.2 Tesla before being imaged, which is an essential process for ensuring the existence of skyrmions at zero field [67].

All L-TEM images were taken in the under-focus Fresnel imaging mode, revealing domain walls as lines of black or white contrast. The size of the skyrmions at under-focus mode was calibrated using a lithographically-patterned microstructure. To image the field-dependent size evolution of magnetic skyrmions, \textit{in-situ} magnetizing is required, which is achieved by adjusting the current of the objective lens in L-TEM and adding an
4.2 Confirmation of Néel-type skyrmions at room temperature and zero field

Due to the strong spin-orbit coupling of Pt and HM atoms, together with the broken inversion symmetry at the interfaces, there should exist a substantial interfacial DMI which is necessary to stabilize the Néel-type skyrmions in $[\text{Pt}/\text{Co}/\text{HM}]_n$ multilayers. To distinguish the possible Néel-type skyrmions from topologically trivial bubble domains, L-TEM is used to identify the chiral Néel domain walls because the phase shift of bubble-like spin textures in the L-TEM mode show different features between Néel-type skyrmions and the topologically trivial bubbles, discussed in Chapter 3. Comparing the experimental results with the simulation results, we can determine whether the bubble domains in $[\text{Pt}/\text{Co}/\text{HM}]_n$ multilayers are Néel-type skyrmions.

Figure 4.2 shows the results of $[\text{Pt}/\text{Co}/\text{W}]_{n=8}$ multilayers. With the sample un-tilted, no observable magnetic contrast was seen in the image shown in Figure 4.2b, consistent...
Figure 4.2: Experimental identification of room-temperature Néel-type skyrmions in the absence of magnetic field in [Pt/Co/W]_{n=8} multilayers. a, L-TEM image of a $\phi = -30^\circ$ tilted sample with zoomed-in features in the blue box. The black and red arrows indicate the sample tilt axis and the rotation direction, respectively. The bubble-like and stripe-like features are magnetic contrast. Dark and bright contrast come from domain walls. The magnetic contrast of these domain walls is dark in the upper left corner and bright in the lower right corner. b, L-TEM image of an un-tilted sample. The round features come from the grains in the film but not magnetic contrast. c, L-TEM image of a $\phi = +30^\circ$ tilted sample with zoomed-in features in the blue box. The magnetic contrast of each feature is bright in the upper left corner and dark in the lower right corner, opposite from a. d, Intensity profile of slices, highlighted as dotted green and red lines, along the diagonal direction of the zoomed-in feature in blue boxes. e-g, Simulation of L-TEM images of a Néel-type bubble skyrmion when tilted at $\phi = -30^\circ$, 0 and $\phi = +30^\circ$, respectively. The center grey region corresponds to the uniformly magnetized core area of a Néel-type bubble skyrmion. The white and black edges are the Néel domain walls.
with the theoretical expectation of Néel-type skyrmions, as shown in Figure 4.2f. The contrast in Figure 4.2b resulted from the presence of grains in the film. When the sample was tilted by $\phi = -30^\circ$ away from the plane normal, randomly distributed bubble-like and stripe-like spin textures were clearly revealed, as shown in Figure 4.2a. For each individual feature, the upper left side appears black, and the lower right side appears white. This contrast reverses upon inverting the tilting angle to $\phi = +30^\circ$, as shown in Figure 4.2c. The switching of black and white contrast in the L-TEM images is consistent with the simulated results for Néel-type bubble skyrmions with different tilting angles shown in Figure 4.2e-g, suggesting that the magnetic structures are Néel-type skyrmions. Note that, compared to the simulation result in Figure 4.2e, the zoomed in skyrmions within the blue frame in Figure 4.2a seem to be rotated by about 45 degrees. This rotation is due to the unique setting and combination of the lens in the specific L-TEM we used, which cannot be changed through routine imaging process. Intensity profiles recorded along the diagonal direction of selected skyrmions further confirms the opposite direction of magnetization change from center to the boundary, as shown in Figure 4.2d. Therefore, the stabilization of Néel-type skyrmions in [Pt/Co/W]$_{n=8}$ multilayers at room temperature and zero field were confirmed using L-TEM. The size of spin textures varies spatially, which can likely be linked to the spatial variation of the local magnetic properties, including anisotropy, magnetization and DMI due to the natural presence of impurities, defects, grain orientations and film roughness.

### 4.3 The effect of interfacial DMI on skyrmions

Figure 4.3 show the L-TEM results of [Pt/Co/Mn]$_{n=8}$, [Pt/Co/Ir]$_{n=8}$ and [Pt/Co/Au]$_{n=8}$. No magnetic contrasts were observed when samples were not tilted. When samples were tilted by $\phi = +30^\circ$ degrees, we observed mainly stripe domains in [Pt/Co/Mn]$_{n=8}$ multilayers, as shown in Figure 4.3a, and Néel-type skyrmions in [Pt/Co/HM]$_{n=8}$ (HM = W, Ir, Au) multilayers, as shown in Figures 4.3b, c and d. Similar to Figure 4.2c, all of these $\phi = +30^\circ$
Figure 4.3: L-TEM imaging of +30° tilted [Pt/Co/HM]_{n=8} (HM=Mn, W, Ir, Au) multilayers. The black and red arrows indicate the sample tilt axis and the rotation direction, respectively. a, Stipe domains in [Pt/Co/Mn]_{n=8}. b, c and d, Néel-type skyrmions in [Pt/Co/W]_{n=8}, [Pt/Co/Ir]_{n=8} and [Pt/Co/Au]_{n=8} multilayers, respectively.
images show features of Néel domain walls with the upper left side appearing white and the lower right side appearing black. The average size of skyrmions in $[\text{Pt/Co/HM}]_{n=8}$ (HM = W, Ir, Au) multilayers is about 480 nm, 150 nm and 200 nm, respectively. The size (density) of skyrmions in $[\text{Pt/Co/HM}]_{n=8}$ (HM = Ir, Au) is much smaller (larger) than that in $[\text{Pt/Co/W}]_{n=8}$. Why does the changing in HM result in the change of skyrmion size?

When the heavy metal is tuned, the atomic number is different. The atomic number $Z$ of W, Mn, Ir and Au are 74, 25, 77, and 79, respectively. As discussed in Chapter 2 Equation (2.5), spin-orbit interaction $E_{\text{SOC}}$ is proportional to $Z^4$. Different heavy metals with different $Z$ induce different strength of spin-orbit interaction. Meanwhile, a large spin-orbit coupling of the non-magnetic atoms in the system is necessary for the interfacial DMI to exist [7, 8, 9]. Therefore, by selecting the heavy metal among W, Ir, Au, and Mn, we directly tune the spin-orbit interaction and thereby change the interfacial DMI in the system, which affects the size of the skyrmions.

To confirm the change of interfacial DMI in the system, we need to develop a method to calculate the strength of DMI in $[\text{Pt/Co/HM}]_{n=8}$ (HM=W, Au, Ir) multilayers. This can be accomplished by investigating the evolution of the skyrmion size as the external magnetic field changes by applying the scaling laws for an effective-medium model. In this model, the magnetic multilayers were considered as an effective thin film due to the coherency of the magnetization in each layer [12, 13, 69, 70] and the reduced (volume) self-energy $\hat{U}$ (that is normalized by $2 \pi^2 M_s^2 h^3$) of each individual, isolated, skyrmion can be expressed as follows:

$$\hat{U} = [N_B + H_a - 1] \left( \frac{d}{h} \right)^2 + \frac{4d}{M_s^2 h^2} \delta_{dw}, \quad (4.1)$$

where $M_s$ is the saturation magnetization, $d$ is the diameter of the skyrmion, $h$ is the effective thickness, $N_B = 1 + \frac{4}{3\pi} \cdot \frac{d}{h} \left\{1 - k^{-3} \left[ (1 - k^2) K + (2k^2 - 1) E \right] \right\}$ with $E$ and $K$ are being complex elliptical integrals of $k = \frac{d}{h} \left[ \left( \frac{d}{h} \right)^2 + 1 \right]^{-1/2}$. $H_a$ is the dimensionless external magnetic field $H_a = \frac{H}{M_s}$, and the domain wall surface energy density $\delta_{dw}$ is further
\[
\delta_{dw} = 4\sqrt{AK_{eff} - \pi |D_k|},
\]  

(4.2)

given by [71, 12, 13]

where \( A \) is the exchange stiffness, and \( K_{eff} \) is the effective perpendicular anisotropy energy density. With experimentally-determined parameters, the reduced energy \( \hat{U} \) as a function of skyrmion diameter \( d \) can be calculated for a given value of \( D_k \) and the results are shown in Figures 4.4, 4.5 and 4.6.

For \([\text{Pt}/\text{Co}/\text{W}]_n\) multilayers, as the external perpendicular magnetic field, \( H_z \), is gradually increased starting from zero, both the size and the density of skyrmions decrease monotonically, as shown in the Figures 4.4a-d. A minimum skyrmion size, \( d_0 = 220 \pm 10 \) nm, is seen, as shown in Figures 4.4e. Further increasing the magnetic field results in the annihilation of the skyrmions in the magnetic field range between 20.0 mT and 25.0 mT, rather than a continuous shrinking of size. After saturation, L-TEM imaging along the hysteresis loop by decreasing the magnetic field is also performed, which reveals labyrinthine stripe domains with widths of which are comparable with the diameter of skyrmions.

The DMI strength can be found by determining the value of \( D_k \), for which the experimentally determined skyrmion sizes correspond to the local minimum of \( \hat{U} \). In Figure 4.4f, the reduced energy \( \hat{U} \) of isolated magnetic skyrmions as a function of diameter \( d \) are plotted for different \( D_k \) at four applied fields of 25.0 mT, 21.0 mT, 12.1 mT and 7.6 mT. The three dashed/dotted curves in each graph are the theoretically calculated energy-diameter dependences for \( D_k = 1.3 \) mJ/m\(^2\), 1.5 mJ/m\(^2\), and 1.7 mJ/m\(^2\), with \( A = 10 \times 10^{-12} \) J/m, perpendicular anisotropy field \( H_k = 0.3 \) T, saturation magnetization \( M_s = 12 \times 10^6 \) A/m, and an effective anisotropy energy density \( K_{eff} = 6.62 \times 10^5 \) J/m\(^3\).

A stable skyrmion state is energetically favored at the energy minima. The experimentally observed skyrmion sizes, shown as vertical dashed lines, are close to the local energy minima at all fields when \( D_k = 1.5 \) mJ/m\(^2\), indicating this is the DMI strength of the system. When \( D_k = 1.3 \) mJ/m\(^2\) or 1.7 mJ/m\(^2\), the skyrmion sizes already do not coincide with the minima as well or only for a limited range of fields. Subsequently, the error in the
Figure 4.4: Calculation of DMI based on the field-dependence evolution of the skyrmion size in [Pt/Co/W]_{n=8}.  

**a-d**, L-TEM imaging at external magnetic fields of 7.6 mT, 21.0 mT, 25.0 mT and 29.0 mT, respectively. The increasing of perpendicular magnetic fields results in the shrinking/annihilation of skyrmions.  

**e**, The change of diameter of selected skyrmions, marked by green arrows in **a**, as a function of field.  

**f**, The reduced single skyrmion energy $\tilde{U}$ calculated for three different values of the DMI strength $D_k$ (1.3, 1.5 and 1.7 mJ/m$^2$) at applied magnetic fields of 25.0 mT, 21.0 mT, 12.1 mT, and 7.6 mT. The sizes of the individual skyrmions from **e** that were labeled as $S_{k1}$-$S_{k5}$, are shown at each field as vertical colored long dashed lines. A DMI value $D_k = 1.5$ mJ/m$^2$ (black thick dotted curves) is inferred, as at this value the experimental skyrmion sizes lie within the minimum energy valley for all applied field.
Figure 4.5: Calculation of DMI in [Pt/Co/Au]$_{n=4}$ multilayers based on the evolution of the Néel-type skyrmions sizes as a function of perpendicular magnetic fields. a-d MOKE images at external magnetic fields of 131 Oe, 169 Oe, 201 Oe and 211 Oe. The increasing of perpendicular magnetic fields results in the shrinking/annihilation of skyrmions. e, The change of diameter of selected skyrmions, marked by different colors of circles in a, as a function of field. f, the reduced single skyrmion energy $\hat{U}$ calculated for three different values of the DMI strength $D_k$ (1.9, 2.1 and 2.3 mJ/m$^2$) at applied magnetic fields of 211 Oe, 201 Oe, 169 Oe, and 131 Oe. The sizes of the individual skyrmions from e that correspond to the color of the circles in a-d, are shown at each field as vertical colored long dashed lines. A DMI value $D_k = 2.1$ mJ/m$^2$ (red short dotted curves) is inferred, as at this value the experimental skyrmion sizes lie within the minimum energy valley for all applied fields.
Figure 4.6: Calculation of DMI in [Pt/Co/Ir]_{n=4} multilayers based on the evolution of the Néel-type skyrmions sizes as a function of perpendicular magnetic fields. a-d, MOKE images at external magnetic fields of 0 Oe, 50 Oe, 60 Oe and 80 Oe. The increasing of perpendicular magnetic fields results in the shrinking/annihilation of skyrmions. e, The change of diameter of selected skyrmions, marked by different colors of circles, as a function of field. f, the reduced single skyrmion energy \( \hat{U} \) calculated for different values of the DMI strength \( D_k \) (2.1, 2.3 and 2.5 mJ/m\(^2\)) based on the effective medium model at applied magnetic fields of 80 Oe, 60 Oe, 50 Oe, and 40 Oe. A DMI value \( D_k = 2.3 \) mJ/m\(^2\) (black short dotted curves) is inferred, as at this value the experimental skyrmion sizes lie within the minimum energy valley for all applied fields.
determined value of $D_k$ is estimated to be at most 0.2 mJ/m$^2$. Note that the minima in the calculations are relatively shallow, which therefore results in the broad size distribution of the skyrmions in the presence of even moderate pinning due to materials imperfections. Furthermore, this may explain why the size evolution with field does not follow the ideal behavior expected from the calculated positions of the minima of the energy, but rather might be determined by a critical energy gradient in order to overcome local pinning. As shown in Figure 4.4f, the skyrmion diameter decreases following an increase of the external magnetic fields. At a magnetic field of 7.6 mT, the skyrmion diameter lies in a range of 200 nm to 700 nm. Experimentally, we have also observed a minimum diameter of value $d_0 = 220 \pm 10$ nm below which the skyrmion state vanishes, and following an increase in applied magnetic fields, the system evolves into a pure ferromagnetic state. Our calculation also captures this feature qualitatively. The less pronounced and subsequent disappearance of the local energy minimum in the calculation as the field increases suggests that the skyrmion phase is not energetically stable.

Similar experiments and calculations were performed in $[\text{Pt}/\text{Co}/\text{Au}]_{n=4}$ and $[\text{Pt}/\text{Co}/\text{Ir}]_{n=4}$ multilayers and the results are shown in Figures 4.5 and 4.6, respectively. Note that, the density of skyrmions in $[\text{Pt}/\text{Co}/\text{Au}]_{n=8}$ and $[\text{Pt}/\text{Co}/\text{Ir}]_{n=8}$ are high and the distance between skyrmions is smaller than the diameter of skyrmions, as in Figures 4.3d and f. To avoid the interaction between skyrmions and match the model created for individual, isolated skyrmion, we need to decrease the density of skyrmions and create larger distance between skyrmions. As shown in Figure 4.9, the density of skyrmion decreases as the repetition number $n$ decreases. Meanwhile, the interfacial DMI is a short distance exchange interaction, existing at the interface between Co and heavy metals, should remain the same in multilayers with different repetition number $n$, as discussed in Chapter 2. Therefore, we performed the experiments and calculation in $[\text{Pt}/\text{Co}/\text{Au}]_{n=4}$ and $[\text{Pt}/\text{Co}/\text{Ir}]_{n=4}$ multilayers to determine the DMI using MOKE microscopy, considering the size of the skyrmions in $[\text{Pt}/\text{Co}/\text{Au}]_{n=4}$ and $[\text{Pt}/\text{Co}/\text{Ir}]_{n=4}$ multilayers is large enough to be resolved by MOKE. The
<table>
<thead>
<tr>
<th>HM</th>
<th>A (J/m)</th>
<th>H_k (T)</th>
<th>M_s (A/m)</th>
<th>K_{eff} (10^5 J/m^3)</th>
<th>DMI strength (mJ/m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>W</td>
<td>10x10^{-12}</td>
<td>0.3</td>
<td>12x10^6</td>
<td>6.62</td>
<td>1.5± 0.2</td>
</tr>
<tr>
<td>Ir</td>
<td>10x10^{-12}</td>
<td>1.1</td>
<td>11x10^6</td>
<td>6.30</td>
<td>2.2± 0.2</td>
</tr>
<tr>
<td>Au</td>
<td>10x10^{-12}</td>
<td>1.2</td>
<td>11x10^6</td>
<td>6.00</td>
<td>2.1 ± 0.2</td>
</tr>
</tbody>
</table>

Figure 4.7: Parameters used in DMI calculation and the calculated DMI strength.

Figure 4.8: Field-dependent magnetization of [Pt/Co/HM]_n (HM = W, Mn, Ir, Au) with an external magnetic field applied along a out-of-plane direction and b in-plane direction.

parameters used in the calculation and the calculated DMI strength are listed in Figure 4.7. Exchange stiffness A is determined by the material Co. Perpendicular anisotropy field H_k, saturation magnetization M_s and an effective anisotropy energy density K_{eff} are extracted from field-dependent magnetization measurements, shown in Figure 4.8.

The stabilized skyrmions show different size, density and uniformity in [Pt/Co/HM]_{n=8} (HM= W, Au, Ir), where the magnitude of the interfacial DMI was estimated to be about D = 1.5 ± 0.2 mJ/m², D = 2.1 ± 0.2 mJ/m² and D = 2.2 ± 0.2 mJ/m² for [Pt/Co/W], [Pt/Co/Au] and [Pt/Co/Ir] interfaces, respectively. Therefore, it can be concluded that DMI
strength is different for multilayers with different heavy metals and affects the stabilization of magnetic skyrmions.

How does DMI affect the skyrmion size? According to Equation (4.2), the Néel domain wall width is determined by exchange stiffness $A$ of Co, effective anisotropy energy density $K_{\text{eff}}$ and the DMI strength. Therefore, domain wall surface energy density is affected by the DMI strength. The results in Figure 4.3 and the calculated DMI strength indicate that the larger the DMI strength is, the smaller the skyrmion size becomes. However, the correlation between DMI and other magnetic properties that might indirectly affect skyrmion stabilization remains unknown. DMI is related to the orbital motion of the electrons of the non-magnetic element, since it requires a strong spin-orbit coupling in the system. Recently, Belabbes et al. have revealed, through first-principle calculations, that the degree of orbital hybridization at the 3d-5d metal interfaces, such as Co-Pt, Co-W, Co-Ir, and Co-Au, directly controls the sign and the strength of the interfacial DMI [72, 73, 74]. Kim et al. claim the correlation of the DMI with Heisenberg exchange and orbital asphericity [75]. Park et al. correlate the DMI to the work function in metallic layers [76]. The changing of DMI relates to the variations in many other magnetic properties, resulting in the change of size and stabilization of magnetic skyrmions; however, the detailed direct correlation remains debatable.

4.4 The effect of the dipolar interaction on skyrmions

We also varied the repetition number $n = 3, 4, 5, 6, 8, 10, 12$ in [Pt/Co/Ir]$_n$ multilayers and observed Néel-type skyrmions. L-TEM or MOKE imaging of the Néel-type skyrmions with $n = 3, 5, 8, 10$ and 12 are shown in Figures 4.3a-e. As $n$ was small (3 or 5), the size of the skyrmions was large enough to be imaged by MOKE microscopy. When $n$ was larger than 5, L-TEM was used to image the smaller stabilized skyrmions. The size of the skyrmions decreases as $n$ increases, as shown in Figure 4.9f. Why does the changing in $n$ result in the change of skyrmion size?
Figure 4.9: L-TEM imaging of +30° tilted [Pt/Co/Ir]$_n$ ($n = 8, 10$ and $12$) multilayers and MOKE imaging of [Pt/Co/Ir]$_n$ ($n = 3, 5$). The black and red arrow indicate the sample tilt axis and the rotation direction, respectively. **a** and **b**, Néel-type skyrmions imaged by MOKE in [Pt/Co/Ir]$_n$ ($n = 3, 5$). In **a** and **b**, black contrast indicates the direction of magnetization is along -$z$ direction pointing into paper plane. **c**, **d** and **e**, Néel-type skyrmions imaged by L-TEM in [Pt/Co/Ir]$_n$ ($n = 8, 10, 12$) multilayers, respectively. **f**, The evolution of skyrmion size as repetition number $n$ in [Pt/Co/Ir]$_n$ multilayers.
As discussed in Chapter 2, any of the Heisenberg exchange, the DMI, the dipolar interaction and the anisotropy energy can affect the stabilization of skyrmions, when no external magnetic field exists. However, among them, only the dipolar interaction is a long-range interaction that could change as the thickness of the sample changes. The other three interactions are short-range interactions. When tuning the $n$, we would not expect the change of the Heisenberg exchange interaction, which is mostly related to the property of Co thin films, nor the interfacial DMI, which is related to the asymmetric interfaces, nor the anisotropy energy, which is dominated by the perpendicular magnetic anisotropy, nor the Zeeman energy, which does not even exist in this system due to the zero-field stabilization of these skyrmions [59].

To confirm the change of the dipolar interaction but not other interactions, we measured the out-of-plane and in-plane field-dependent magnetization hysteresis loops of [Pt/Co/Ir]$_n$ ($n = 1, 2, 3, 4, 5, 6, 8, 12$) multilayers and part of the results are shown in Figure 4.10. It is observed that when $n$ increases, the field-dependence hysteresis loop at the easy axis (out-of-plane) becomes more slanted with gradual changes towards saturation. In her dissertation, X.M. Cheng has pointed out that the larger the demagnetization factor is, the slanted the field-dependence hysteresis loop becomes [59]. Therefore, multilayer samples with larger repetition number $n$ have larger dipolar interaction (demagnetization interaction/stray field). It is also noticed that, as $n$ increases, the magnitude of the external in-plane field needed to saturate the sample is about the same for samples with different $n$. Figure 4.11 summarizes the magnetic properties of [Pt/Co/Ir]$_n$ ($n = 1, 2, 3, 4, 5, 6, 8, 12$) multilayers. The calculated saturation magnetization $M_s$, directly related to the exchange interaction, and effective anisotropy energy density $K_{eff}$, correspondence to the anisotropy energy, are also about the same for samples with different $n$. Therefore, if we assume the interfacial DMI remains the same since the Pt/Co and Co/Ir interfaces are the same for the samples with repetition number $n$, we can conclude that in [Pt/Co/Ir]$_n$ multilayers, the increase of dipolar interaction is the pronounced changes while DMI, saturation magneti-
How does the dipolar interaction affect skyrmion size? The size of the magnetic skyrmions observed in [Pt/Co/Ir]$_n$ multilayers is about 80 nm and does not decrease further as $n$ increases to larger than 8, shown in Figure 4.9f. Though the increase of dipolar interaction can result the decrease of skyrmion size, the effect stops at a certain point. As discussed in Chapter 2, the dipolar interaction favors breaking the large area of the magnetic domain and assists in forming magnetic flux-closure domains with smaller areas [58]. The larger the dipolar interaction is, the smaller the domain size is. Therefore, the dipolar interaction could affect the size of the uniformly magnetized core area of a bubble skyrmion. The size of the core area decreases as the dipolar interaction increases, resulting the decrease of the size of a skyrmion. Considering the observed Néel domain wall width in Pt/Co/MgO film is about 30 nm [77], we expect the smallest size of the magnetic skyrmions to be about 60 nm in a Pt/Co/MgO film, assuming minimum core area at the center of the magnetic skyrmions. The smallest size 80 nm observed in [Pt/Co/Ir]$_n$ could come purely from the width of domain walls in Co. Hence, it could be concluded that the dipolar interaction affects the size of the core area of the magnetic skyrmions. When the dipolar interaction is
| n | Hc⊥ (Oe) | Hk || (kOe) | Thickness(nm) | Ms (A/m) | Keff (J/m³) |
|---|---------|----------|--------------|----------|----------------|
| 1 | 100±10  | 11.0±0.2 | 1            | (1.06±0.03)*10⁶ | (6.2±0.5)*10⁵ |
| 2 | 100±10  | 11.4±0.2 | 2            | (1.06±0.03)*10⁶ | (6.2±0.7)*10⁵ |
| 3 | 150±10  | 11.8±0.2 | 3            | (1.06±0.03)*10⁶ | (6.8±0.7)*10⁵ |
| 4 | 150±10  | 11.4±0.2 | 4            | (1.06±0.03)*10⁶ | (6.2±0.7)*10⁵ |
| 5 | 200±10  | 11.4±0.2 | 5            | (1.02±0.03)*10⁶ | (5.9±0.3)*10⁵ |
| 6 | 100±10  | 11.4±0.2 | 6            | (1.10±0.03)*10⁶ | (6.5±0.3)*10⁵ |
| 8 | 150±10  | 11.4±0.2 | 8            | (1.06±0.03)*10⁶ | (6.2±0.3)*10⁵ |
| 12| 150±10  | 11.8±0.2 | 12           | (1.15±0.03)*10⁶ | (6.5±0.5)*10⁵ |

Figure 4.11: Magnetic properties of [Pt/Co/Ir]ₙ (n = 1, 2, 3, 4, 5, 6, 8, 12) multilayers.

increased to a specific value, the size of the core area does not decrease anymore, and the total size of the magnetic skyrmions depends on the width of the Néel-type chiral domain walls.

4.5 Conclusions

The stabilization of Néel-type skyrmions was observed and confirmed by L-TEM imaging and MOKE microscopy in the (Pt/Co/HM)ₙ (HM = W, Ir and Au, n = 3, 4, 5, 8, 10 and 12) multilayers, at room temperature in the absence of an external field. The effect of interfacial DMI and the dipolar interaction on skyrmion stabilization were investigated in [Pt/Co/HM]ₙ (HM = W, Mn, Ir and Au, n = 3, 4, 5, 8, 10, and 12) multilayers by tuning the heavy metal (HM) and repetition number n, respectively.

The magnitude of the interfacial DMI changes as HM changes in multilayers. Based on the evolution of the skyrmion size as a function of the perpendicular field, we estimated the strength of DMI to be about $D_k = 1.5 \pm 0.2$ mJ/m² for [Pt/Co/W] interface. $D_k$ can be increased to $D_k = 2.1 \pm 0.2$ mJ/m² and $D_k = 2.2 \pm 0.2$ mJ/m² by changing W to Au and Ir. The size of the stabilized skyrmions decreases as the interfacial DMI increases. Our
results suggest that L-TEM and MOKE imaging combined with analytical calculation can provide valuable quantitative information about the interfacial DMI strengths, which can be helpful for optimizing skyrmion materials.

The dipolar interaction was tuned by varying the repetition number \( n \) in \([\text{Pt/Co/Ir}]_n\) multilayers. Comparing the hysteresis loops of samples with different \( n \), we confirmed the increases of dipolar interaction as \( n \) increases. Considering the unaffected saturation magnetization, effective anisotropy and interfacial DMI, we could attribute the decreases of skyrmion size from a few \( \mu \text{m} \) to about 100 nm to the increase of dipolar interaction through changing \( n \) from 3 to 8. The size of skyrmion does not decrease any further after \( n=8 \), suggesting that the dipolar interaction mainly affects the size of the uniformly magnetized core area of a bubble skyrmion.

Our results show that skyrmion formation is sensitive to the relative magnitudes of different effects and represent an important step towards understanding the conditions necessary for stabilizing room temperature magnetic skyrmions. Some of the results in this chapter were published as "Quantifying chiral exchange interaction for Néel-type skyrmions via Lorentz transmission electron microscopy" in Physics Review B in collaboration with researchers from Argonne National Laboratory and the University of New Hampshire.

References of Chapter 4


CHAPTER 5
DIRECT OBSERVATION OF SKYRMION HALL EFFECT

5.1 Motivation and Experiments

The study of current-driven skyrmion motion is an important step towards the understanding of the current-induced spin dynamics and the application of skyrmions in data storage devices [21, 12]. This chapter presents the investigation of current-driven skyrmion dynamics and the first experimental observation of skyrmion Hall effect, which refers to the phenomenon that when driven by an electric current, magnetic skyrmions deviate from the current direction towards the edge of the devices. Skyrmion Hall effect is so named because its analogous to the ordinary Hall effect. Figure 5.1a shows the ordinary Hall effect, known for centuries, in which electrons move towards the sample edge as a result of the Lorentz force when there exists an external magnetic field perpendicular to the device surface and the current direction. In the skyrmion Hall effect, as shown in Figure 5.1b, skyrmion topological charge, rather than electrical charge, causes skyrmions to travel towards the edge.

Hall bar devices, as discussed earlier in Chapter 3, with three different dimensions were patterned on Ta/CoFeB/TaOx films for studying current-driven skyrmion dynamics. A Hall bar with dimensions of 100 μm in width and 500 μm in length was used to study the current-dependent skyrmion motion. A narrower device of 60 μm in width and 500 μm in length was used to observe the skyrmion accumulation along the edge of the device. Another hall bar of 80 μm in width and 100 μm in length was used to probe the saturation of the skyrmion Hall angle, the angle between applied current density direction and skyrmion motion direction. This shorter device enables larger current density because the current density is inversely proportional to the length of the device, the material resistance, fixed
Figure 5.1: Illustration of ordinary Hall effect and skyrmion Hall effect. $J$ is the current. $j_e$ denotes the electron density. The negative sign means the electron motion is along negative $x$ direction from right to left. 

**a**, Ordinary Hall effect of an electron. The red arrow indicates the moving trajectory of an electron when there exists a magnetic field ($H$) orthogonal to the sample surface and the current direction. 

**b**, Skyrmion Hall effect of a skyrmion with topological charge $Q = -1$. The red arrow refers to the deviation of an skyrmion towards the edge of device from electron motion direction. Notice that an external magnetic field is not necessary for skyrmion Hall effect.
by material Ta and CoFeB, and proportional to the voltage applied to the device. The pulse
generator used in this work has the capability of applying pulses with 0-45 V and the range
of current density achieved by this shorter device is $0.1 \times 10^6 \text{ A} \cdot \text{cm}^{-2} < j_e < 9.0 \times 10^6$
$\text{A} \cdot \text{cm}^{-2}$. The current density is calculated based on $j_e = \frac{V}{Rwt}$, with $V$ being the applied
voltage, $R$ being the measured resistance of device, $w$ being the width of the device and
t being total thickness of 5 nm Ta and 1.1 nm CoFeB. The direction of current was also
switched to probe the effect of current direction on skyrmion Hall angle.

Moreover, skyrmion with different topological charges $Q = +1$ or $Q = -1$ and different
sizes were stabilized to investigate the effect of topological charge’s sign and skyrmion
size on skyrmion motion. The switching of the topological charge’s sign was achieved by
reversing the direction of the external magnetic fields. The modification of the skyrmion
size was achieved by tuning the magnitude of the external magnetic fields between 4.6 to
5.4 Oe.

This work is a collaborative work among researchers from Argonne National Labora-
tory, Chinese University of Hong Kong, Universities of California, Los Angeles and Bryn
Mawr College. My contributions include photolithography Hall bar devices and Magneto-
optical Kerr effect microscopy imaging and data analysis for the results in Figures 5.3, 5.5
and 5.6, as well as the derivation of equation (5.2). This work was published in Nature

5.2 Observation of skyrmion transverse motion

For a Hall bar device with the dimensions of 100 $\mu$m in width and 500 $\mu$m in length,
upon applying currents, a few skyrmions formed underneath the electrodes. Those skyrmions
were driven by the current into the area of interest, shown in Figure 5.2. As shown in Fig-
ure 5.2, when skyrmions move in the interior of the Hall bar devices, the trajectory follows
approximately a straight line that has an angle with the electron motion direction. This
phenomenon indicates that skyrmions experience not only the force along but transverse to
Figure 5.2: Magneto-optical Kerr effect (MOKE) imaging of current driven skyrmion motion that deviates from the current direction towards the device edge. The blue area is substrate. The black area is the device. The dark contrast indicates the magnetization in this area is along $-z$ direction, the same direction as the external magnetic field of $-5.2$ Oe. The dimensions of the Hall bar device are $100 \mu m$ in width and $500 \mu m$ in length, but the area shown in the figure is about $100 \mu m \times 160 \mu m$. The red arrow on top of the device refers to electron motion direction. The magnitude of electron current density $j_e$ equals to $6.0 \times 10^6$ A·cm$^{-2}$. The negative sign of $-j_e$ means the electron motion is along negative $x$ direction from right to left. The white contrast of the skyrmions suggests that the magnetization of the core area is along $+z$ direction and the topological charge of the skyrmion is $+1$. A few $Q = +1$ skyrmions moved approximately along a straight line, marked as a dashed grey line for eye guidance, forming an angle with the current direction.[30]
Figure 5.3: Accumulation of skyrmions at the device edge. The dimensions of device is 60 \( \mu \text{m} \) in width and 500 \( \mu \text{m} \) in length, but the area shown in the image is about 60 \( \mu \text{m} \times 140 \mu \text{m} \). The red arrow refers to electron motion direction. The negative sign of \(-j_e\) means the electron motion is along negative \( x \) direction from right to left. 

(a) Demonstration of skyrmion (\( Q = -1 \)) accumulation at the edge of the device. This is done by repetitively applying 50 pulsed currents of 50 \( \mu \text{s} \) duration at a frequency of 1 Hz with a current density \( j_e = 6 \times 10^6 \text{ A/cm}^2 \) and an applied field of +5.4 Oe along +\( z \) direction. 

(b) Reversing the magnetic field from positive (+5.4 Oe in Figure 5.3a) to negative (-5.2 Oe in Figure 5.3b) leads to the accumulation of skyrmions with positive topological charge \( Q = +1 \) at the opposite edge.[30] 

The electron motion direction, which is similar to the ordinary Hall effect.

To draw a closer link between the ordinary Hall effect from electric charge and the Hall effect from topological charge, we demonstrated experimentally the accumulation of skyrmions at the edge of device. This was done in a narrow device of 60 \( \mu \text{m} \) in width and 500 \( \mu \text{m} \) in length. By applying 50 pulsed currents of 50 \( \mu \text{s} \) duration at a frequency of 1 Hz with a current density \( j_e = 6 \times 10^6 \text{ A/cm}^2 \), the accumulation of \( Q = -1 \) skyrmions is observed at the lower edge of the device, as shown in Figure 5.3a. Again, reversing the magnetic field from positive (+5.4 Oe in Figure 5.3a) to negative (-5.2 Oe in Figure 5.3b) leads to the reversal of topological charge, and the accumulation of \( Q = +1 \) skyrmions at the upper edge, shown in Figure 5.3b. This observation resembles the charge accumulation in ordinary Hall effect.
5.3 Topological Magnus force and Thiele equation

In analogues to Lorentz force which causes the ordinary Hall effect, there should exist a force that makes the skyrmion move transversely. By considering an isolated Néel skyrmion as a point-like rigid particle, the translational motion can be described by a modified Thiele equation [24, 25, 26, 27, 28]

\[ G \times v - \alpha D \cdot v + 4\pi B \cdot j_{hm} = 0, \quad (5.1) \]

where \( G = (0, 0, -4\pi Q) \) is the gyromagnetic coupling vector with \( Q \) being the topological charge, \( v \) is the skyrmion drift velocity along both \( x \) and \( y \) axes, \( \alpha \) is the magnetic damping coefficient, \( D \) is the dissipative force tensor, and \( j_{hm} \) is the electrical current density flowing in the heavy metal layer. The first term in equation (5.1) is the topological Magnus force that results in the transverse (gyrotropic) motion of skyrmions with respect to the driving current [25, 27, 28, 29]. This term acts equivalently to the Lorentz force for electric charge, and thus gives rise to a Hall-like behavior of magnetic skyrmions [78]. The second term is the dissipative force that is linked to the intrinsic magnetic damping of a moving magnetic skyrmion, and the third term is the driving force from the spin Hall spin torque. We note that equation (5.1) does not include possible pinning effects that may impede skyrmion motion due to the presence of material imperfections [79, 80, 81], nor does it include the possibility of exciting internal degrees of freedom of the skyrmion. Such internal degrees of freedom may modify the dynamics and dissipation of the driven skyrmion [82, 83].

Upon applying a spatially homogeneous current along the \( x \) direction \( j_{hm} = (j_x, 0) \), the resultant velocity along the \( x \) and \( y \) axes can be calculated as \( v_x = \frac{-\alpha D}{Q^2 + \alpha^2 D^2} B_0 j_x \), \( v_y = \frac{Q}{Q^2 + \alpha^2 D^2} B_0 j_x \), respectively [27, 28]. Here \( B_0 \) is a constant that can be estimated based on the detailed spin configuration. This leads to the following expression for the
ratio of in-plane velocity components to be written as:

\[ \frac{v_y}{v_x} = \frac{-Q}{\alpha D}, \]  

(5.2)

where \( \alpha \approx 0.02 \) is the damping parameter for the magnetic bilayer involved in this study [21]. The dissipative tensor, describing the effect of the dissipative force on the moving magnetic skyrmion, is given by

\[ \mathcal{D} = D_{xx} = D_{yy} = \left( \frac{1}{4\pi} \right) \int (\partial \mathbf{m} / \partial x) \cdot (\partial \mathbf{m} / \partial x) \, dx \, dy \]

and \( D_{xy} = D_{yx} = 0 \). Assuming a Néel-type skyrmion with a linear change of the spin profile along the radial direction inside the domain wall (the angle \( \theta \) with respect to the \( z \) axis going from \( \pi \) inside the skyrmion to 0 outside of the skyrmion), the dissipative tensor can be analytically expressed as:

\[ \mathcal{D} = \pi^2 d / 8 \gamma_{\text{dw}}. \]  

(5.3)

Thus \( \mathcal{D} \) depends on the domain wall width \( \gamma_{\text{dw}} \) and is proportional to the skyrmion diameter \( d \). Hence, small Néel skyrmions \( v_y/v_x \gg 1 \) dynamics show large skyrmion Hall angle close to ±90°, resulting in mostly transverse motion, while larger skyrmions move with smaller skyrmion Hall angle and their perpendicular motion can be less pronounced with \( v_y/v_x \approx 1 \). In either case, skyrmions move towards the edge of the device.

5.4 Current-dependent skyrmion dynamics

To investigate the current-dependent skyrmion dynamics, we studied the skyrmion transverse motion of an isolated skyrmion by changing driven current density for both \( Q = -1 \) and \( Q = +1 \) scenarios in a Hall bar device with the dimensions of 100 \( \mu \text{m} \) in width and 500 \( \mu \text{m} \) in length. Note that we studied a single skyrmion that is isolated from other skyrmions to avoid complications due to skyrmion-skyrmion interactions.

MOKE images are shown in Figures 5.4a-f for \( Q = -1 \) skyrmions and in Figures 5.4g-l for \( Q = +1 \) skyrmions, respectively. These two experiments were performed by applying
Figure 5.4: MOKE imaging of current-dependent skyrmion dynamics. All experiments were done by using 50 µs pulsed currents. The red arrow refers to electron motion direction. The plus sign of $+j_e$ indicates the electron motion is along positive $x$ direction from left to right. a-e, Snapshots of $Q = -1$ skyrmion motion captured after applying successive current pulses of amplitude $j_e = 1.3 \times 10^6$ A·cm$^{-2}$ and external perpendicular magnetic field $H_\perp = +4.8$ Oe. f, Summary of the skyrmion trajectory from a-e, showing no net transverse motion along the $y$ direction. g-k, Snapshots of $Q = +1$ skyrmion motion at $j_e = 1.3 \times 10^6$ A·cm$^{-2}$ and magnetic field $H_\perp = -5.2$ Oe. l, Stochastic trajectory from g-k, again showing no net transverse motion. m-q, Snapshots of $Q = -1$ skyrmion motion at $j_e = 2.8 \times 10^6$ A·cm$^{-2}$ and magnetic field $H_\perp = +5.4$ Oe. r, Summary of the trajectory from m-q. Nearly straight and diagonal trajectory indicates the presence of transverse motion along the $+y$ direction. The size of skyrmion shrinks slightly as compared to a-e due to the larger perpendicular magnetic fields. Two other skyrmions that moved into the frame, marked with green circles in m and o, were not studied. s-w, Snapshots of $Q = +1$ skyrmion motion at $j_e = +2.8 \times 10^6$ A·cm$^{-2}$ and magnetic field $H_\perp = -5.2$ Oe. x, Summary of the trajectory from s-w. Again, there is a nearly straight and diagonal trajectory. However, the slope is opposite, indicating that the presence of transverse motion is along the $-y$ direction and opposite to r.[30]
pulsed electron currents of amplitude \(j_e = +1.2 \times 10^6 \text{ A} \cdot \text{cm}^{-2}\) and with a duration of 50 \(\mu\text{s}\). By comparing the trajectories shown in Figure 5.4f for the \(Q = -1\) skyrmion and Figure 5.4i for the \(Q = +1\) skyrmion, it is clear that the observed stochastic motion does not have transverse components, which is consistent with the observation by Jiang \textit{et al} where skyrmions move along the device edge without a traverse component with current density \(j_e < +1.0 \times 10^6 \text{ A} \cdot \text{cm}^{-2}\) [21].

After increasing the current density to \(j_e = 2.8 \times 10^6 \text{ A} \cdot \text{cm}^{-2}\), it is observed that the direction of motion develops a well-defined transverse component, which is exemplified by a straight and diagonal trajectory. Figures 5.4m-r correspond to a \(Q = -1\) skyrmion, and Figures 5.4s-x to a \(Q = +1\) skyrmion. It is also noticed that the motions of the skyrmions with opposite topological charge \(Q\) also have opposite sign of slopes, as shown in Figures 5.4r and x. The opposite sign of slopes in Figures 5.4r and x are consistent with the opposite sign of the topological Magnus force, that consequently gives rise to opposite directions for the transverse motion, as in equation 5.2.

By performing similar measurements in Figures 5.4m-r but gradually varying the current density \(0.1 \times 10^6 \text{ A} \cdot \text{cm}^{-2} < j_e < 6.2 \times 10^6 \text{ A} \cdot \text{cm}^{-2}\), we systematically study the current-dependent transverse motion of a single skyrmion stabilized with an external magnetic field +5.2 Oe. The results of current-dependent velocity along \(x\) axis, current-dependent velocity along \(y\) axis and current-dependent skyrmion Hall angle are shown as Figure 5.5. The current-velocity relationship is shown in Figure 5.5a, indicating a monotonic increase of the average velocity as a function of current density. The average velocity \((\bar{v})\) is defined as \(\bar{v} = \mathcal{L}/(N \cdot \Delta t)\), where \(\mathcal{L}\) is the total displacement, \(N\) is the number of pulses, and \(\Delta t\) is the duration of pulse. The number of pulses was typically chosen to be \(N > 10\) to minimize the uncertainty due to the stochastic motion of skyrmion in the creep motion regime. For a fixed pulse duration of 50 \(\mu\text{s}\), there is a threshold depinning current density \(j_e = (0.6 \pm 0.1) \times 10^6 \text{ A} \cdot \text{cm}^{-2}\), below which skyrmions remain stationary, shown as light blue regime in Figure 5.5a. Above this threshold depinning current two features
Figure 5.5: Current-dependence transverse motion of a single skyrmion. **a**, The average skyrmion velocity ($\bar{v}$) as a function of electron current density $j_e$. The light blue region corresponds to the skyrmion-pinning regime, where $j_e < (0.6 \pm 0.1) \times 10^6$ A·cm$^{-2}$. The beige region corresponds to the regime of stochastic motion without net transverse motion, where $(0.6 \pm 0.1) \times 10^6$ A·cm$^{-2} < j_e < 1.5 \times 10^6$ A·cm$^{-2}$. **b**, Evolution of the skyrmion Hall angle $\varphi_{sk}$ and the ratio between the transverse and longitudinal velocities of the skyrmion $v_y/v_x$. The green region corresponds to the regime without net transverse motion, where $j_e < 1.5 \times 10^6$ A·cm$^{-2}$. When $j_e > 1.5 \times 10^6$ A·cm$^{-2}$, both $\varphi_{sk}$ and $v_y/v_x$ are monotonically increasing as a function of current density.[30]

were observed: (I) stochastic migration of skyrmions following the electron current direction when $j_e < 1.5 \times 10^6$ A·cm$^{-2}$, shown as beige regime in Figure 5.5a. (II) Motion of skyrmions with a well-defined transverse velocity when the current density $j_e < 1.5 \times 10^6$ A·cm$^{-2}$, shown as light green regime in Figure 5.5b. For example, the velocity is estimated to be $\bar{v} \approx 0.75 \pm 0.02$ m/s at a current density $j_e = 6.2 \times 10^6$ A·cm$^{-2}$. It is also noticed that the threshold depinning current density evolves as a function of pulse duration. More specifically, for shorter pulses, larger amplitudes are required to result in skyrmion motion, indicating a thermally assisted depinning process of magnetic skyrmions from local pinning sites due to disorder.

The transverse motion is shown in Figure 5.5b. The deviation of skyrmion motion with respect to the applied current direction $+x$ can be quantified by a skyrmion Hall angle $\varphi_{sk} = \tan^{-1}(v_y/v_x)$. With increasing current density to $j_e > 1.5 \times 10^6$ A·cm$^{-2}$, the ratio
of \((v_y/v_x)\), shown as blue symbols, and consequently the skyrmion Hall angle \(\varphi_{sk}\), shown as dark red symbols, increase monotonically. The skyrmion Hall angle can be as large as \(\varphi_{sk} \approx 16^\circ\) with a ratio of \((v_y/v_x) \approx 0.28\) at the maximum current density \(j_e = 6.2 \times 10^6\) A·cm\(^{-2}\) (limited by the present instrument). Given the roughly linear dependence on the current density without indication of saturation, it is expected that the skyrmion Hall angle \(\varphi_{sk}\) can be even larger for even higher current densities. However, the electrical resistance of the first type of devices prevented application of higher current densities. We addressed this issue with slightly modified devices, as discussed further in the next section.

This current density dependence is inconsistent with the simple theoretical prediction given in equation (5.2), which suggests a constant value of \((v_y/v_x)\) and is independent of the driving current. On the other hand, the magnitude obtained at the highest achievable current density is approximately in the range of what can be expected theoretically. While for most theoretical studies, small skyrmions with diameters of 10 nm are considered, leading to a small dissipative term and \((v_y/v_x) > 1\), the skyrmions imaged here are significantly larger (\(\approx 1,000\) nm). For skyrmions of diameter \(\approx 1,000 \pm 300\) nm with a fixed domain wall widths \(\gamma_{dw} = 21\) nm, the value of the dissipative term is estimated to be around 60 and increases proportionally with the area of the skyrmion. The error bar of 300 nm arises from the optical diffraction limit. This suggests a skyrmion Hall angle \(\varphi_{sk}\) around 40\(^\circ\) as an upper limit, which is compatible with the observed values of \((v_y/v_x)\) being less than 0.3 for current density \(j_e = 6.2 \times 10^6\) A·cm\(^{-2}\). A further increase of the current density could thus lead to higher values of \((v_y/v_x)\) and the saturation of skyrmion Hall angle.

One possible reason for the apparent discrepancy between our experimental observation of the current-dependent increasing value of \((v_y/v_x)\) and the simple prediction of equation (5.2) is the presence of pinning that affects the skyrmion motion [79, 80, 81, 84]. Such pinning may originate from random disorder/defects in the sputtered films, which is consistent with the experimentally observed threshold depinning of skyrmions and stochastic motion at low driving currents. Detailed theoretical investigation of the dynamics of skyrmions
interacting with randomly distributed disorder/defects has shown a significant reduction of the skyrmion Hall angle $\varphi_{sk}$, as well as complex skyrmion trajectories [79, 80, 81, 84]. Specifically, the skyrmion Hall angle is minimized around the depinning threshold and increases monotonically with the driving current, due to side-jump scattering of skyrmions from the scattering potentials. This is reminiscent of our experimental observations in the absence of interactions between multiple skyrmions. Experimentally for $j_e < 1.5 \times 10^6$ A·cm$^{-2}$, skyrmions escape from the pinning potential and exhibiting a hopping-like motion along the driving direction with a zero skyrmion Hall angle. In the strong-driving regime $j_e > 1.5 \times 10^6$ A·cm$^{-2}$, increasing the driving force increases monotonically the skyrmion Hall angle $\varphi_{sk}$. In fact, a recent study of the dependence of skyrmion Hall angle on the driving force in the presence of random defects reproduced the experimentally observed behavior and suggested the saturation of the skyrmion Hall angle in the strong-driving regime [81].

5.5 Skyrmion Hall angle diagram

To probe the saturation of skyrmion Hall angle and summarize the factors affecting skyrmion Hall angle, a skyrmion Hall angle diagram is explored. The results are shown in Figure 5.6. The dimensions of the device are 80 $\mu$m in width and 100 $\mu$m in length. When the applied current density is larger than $8 \times 10^6$ A·cm$^{-2}$, indeed a saturation of the skyrmion Hall angle is observed, shown in Figure 5.6. For regime I ($+j_e$, -Q) we present data for the positive electron current density $+j_e$ and positive perpendicular magnetic fields -Q. For $H_\perp = +5.4$ Oe, a saturation of skyrmion Hall angle $\varphi_{sk} \approx 32 \pm 2^\circ$ is observed on a skyrmion of diameter $d = 800\pm300$ nm, and $\varphi_{sk} \approx 28 \pm 2^\circ$ is observed for $H_\perp = +4.8$ Oe on a skyrmion with a larger diameter $d = 1,100\pm300$ nm. This trend agrees with the dependence of dissipative tensor $D$ on $d$ given by equation (5.3). The values are just below the expected range of values based on equations (5.2) and (5.3), which are $47 \pm 11^\circ$ and $38 \pm 8^\circ$ for $d = 800$ nm and 1,100 nm, respectively, using the estimated domain wall width.
Figure 5.6: Skyrmion Hall effect diagram. Diagram of the skyrmion Hall angle as a function of current density/sign of topological charge in a modified device of dimensions 80 µm (width) × 100 µm (length), obtained by tracking the motion of several skyrmions. In the low-current-density regime, the skyrmion Hall angle $\varphi_{sk}$ exhibits a linear dependence similar to that shown in b. A further increase of current density $j_e > 8 \times 10^6$ A·cm$^{-2}$ results in the saturation of the skyrmion Hall angle. By alternating the sign of the driving electron current density ($\pm j_e$) and the sign of topological charge ($\pm Q$), a phase diagram for the four different regimes was determined. Namely, for negative topological charge (under positive magnetic fields), regime I ($+j_e$, -Q) with positive $\varphi_{sk}$ and regime III ($-j_e$, -Q) with negative $\varphi_{sk}$ were identified by changing the polarity of the electron current. For skyrmions with positive topological charge (under negative magnetic fields) a positive $\varphi_{sk}$ in regime II ($-j_e$, +Q), and negative $\varphi_{sk}$ in regime IV ($+j_e$, +Q) were detected. The decrease of skyrmion Hall angle from $|\varphi_{sk}| \approx 32 \pm 2^\circ$ to $|\varphi_{sk}| \approx 28 \pm 2^\circ$ is also demonstrated by increasing the skyrmion diameter from $d = 800 \pm 300$ nm (+5.4 Oe-5.2 Oe) to $d = 1100\pm300$ nm (+4.8 Oe-4.6 Oe).[30]
\( \gamma_{dw} \) of 21 nm, and taking only the uncertainties in the skyrmion diameter into account. The quantitative agreement is reasonable considering the uncertainty in the exchange stiffness in thin films [85, 10, 86] which impacts the estimated value of the domain wall width, and the fact that equation (5.1) is derived assuming point-like rigid skyrmions.

It is noted that the size of skyrmions does not change significantly as a function of perpendicular fields due to the strong contribution from the dipolar interaction in the present system. Above +5.4 Oe, skyrmion bubbles collapse, whereas below +4.8 Oe, skyrmion bubbles transform into stripe domains. Nevertheless, by varying the strength of magnetic field and hence the size of skyrmion bubbles, we observed a size-dependent skyrmion Hall angle.

A negative electron current direction \(-j_e\) reverses the direction of skyrmion motion that leads to a negative saturation skyrmion Hall angle, shown in regime III \((-j_e, -Q)\). In the presence of a negative perpendicular magnetic field with a positive topological charge +Q, such a trend is reversed, as summarized in the regime II \((-j_e, +Q)\), and regime IV \((+j_e, +Q)\), respectively. This is consistent with the opposite topological Magnus force.

### 5.6 Conclusion

In summary, we have observed the transverse motion of skyrmions deviating from the current direction towards the edge of the devices. We also observed the accumulation of skyrmions with opposite topological charge at opposite side of the edges in analogues to the charge accumulation in the ordinary Hall effect. By changing the sign of the topological charge, and the sign of the electric current, we have revealed a strong similarity between the ordinary Hall effect of the electronic charge and the skyrmion Hall effect due to the topological charge. Combining the theoretic derivation based on the Thiele equation, we attribute our observation to the topological Magnus force together with random pinning potentials in the materials, where the competition between pinning potentials and the skyrmion driving force leads to a significant current-dependence of the skyrmion Hall effect.
angle. For instance, below a certain current density, the skyrmions show a hopping-like motion with a zero skyrmion Hall angle. At higher current densities, a stronger driving force allows skyrmions to overcome pinning potentials gradually, and eventually leads to the saturation of the skyrmion Hall angle. Our observations also indicate that the topological charges of magnetic skyrmions, in combination with the current-induced torque, can be potentially integrated for realizing novel functionalities, such as topological sorting.

References of Chapter 5


CHAPTER 6
PERSISTENCE OF CHIRAL DOMAIN WALLS IN
ANTIFERROMAGNETICALLY-COUPLED BUBBLE SKYRMION PAIRS
DURING SPIN REORIENTATION TRANSITION

6.1 Motivation and experiments

Conventional ferromagnetic skyrmions with topological charge \( Q = \pm 1 \) can be stabilized at room temperature and zero field in multilayers with the interfacial Dzyaloshinskii-Moriya interaction (DMI) and efficiently driven by a current, presented in Chapters 4 and 5, respectively, giving rise to the promising applications in data storage. However, ferromagnetic skyrmions have some intrinsic issues in terms of the applications in race-track memory and one of them is the skyrmion Hall effect (SkHE), causing a drift of skyrmions towards the device edge and their possible subsequent annihilation, as discussed in Chapter 5. Recently, antiferromagnetically-coupled (AFM-coupled) bubble skyrmions, a bi-layer pair of AFM-coupled bubble skyrmions with \( Q = 0 \), as illustrated in Figure 6.1 has attracted intense research interest. Compared to their ferromagnet counterparts, AFM-coupled skyrmion pairs have faster mobility and suppressed SkHE because of the reduced net magnetization and angular magnetic moment, predicted by theory\[87, 88, 17\] and then observed in experiments by Woo \textit{et al} \[86, 18\] and Caretta \textit{et al} \[89\]. However, both Woo’s and Caretta’s systems include alloy, which does not have a defined interface, complicating the study of the antiferromagnetic exchange interaction between Co and Gd. Also, these AFM-coupled bubble skyrmions can only be stabilized with an external magnetic field. Moreover, these AFM-coupled skyrmion pairs have reduced but non-zero net angular magnetic moment which gives only reduced but not cancelled SkHE.

We specifically created and studied a simple and well-controlled [Co/Gd/Pt]_n multi-
Figure 6.1: Illustration of an AFM-coupled bubble skyrmion pair, as a perspective view (at the top) and a view of the spin configuration along radial direction (at the bottom). Arrows represent the atomic spins. Color difference corresponds to different magnetization along the $z$ direction as indicated in the color scale bar. The topological charge $Q$ of the top (bottom) skyrmion is $-1$ ($+1$) and therefore the total $Q = 0$. Spins in the top skyrmion are one-on-one AFM-coupled to the spins in the bottom skyrmion, as shown by the opposite colors in the color scale.
layer system with defined Co/Pd interfaces and many tunable parameters, such as thickness and repetition number [90, 91]. We also investigated the AFM-coupled skyrmions at different temperatures to realize zero net magnetization or zero net angular magnetic moment and study the temperature-dependent stabilization, considering the temperature-dependent magnetization of Gd below room temperature.

A [Co/Gd/Pt]ₙ multilayer film has a similar structure as the [Pt/Co/HM] multilayers, which can host bubble skyrmions at room temperature and zero field, but AFM-coupled Co/Gd itself. To study the AFM-coupled skyrmion pairs, Ta (5 nm) /Pt (4 nm) /[Co (0.5 nm) /Gd (1 nm) /Pt (1 nm)]ₙ=10 /Al (2 nm) multilayers were engineered.

Photo emission electron microscopy (PEEM) was used to image the skyrmions at Gd layer and Co layer while temperature was decreased from room temperature to 30 K and subsequently increased to 55 K. Multilayers were magnetized in an in-plane magnetic field of 0.6 Tesla before imaging, which is an essential process for ensuring the existence of skyrmions at zero field [67]. The energy of x-ray was tuned to the Gd M₅ peak (1188.8 eV) and the Co L₃ peak (778.5 eV) for element-specific imaging. To quantitatively determine the temperatures where total magnetization or angular magnetic moment is zero, x-ray magnetic circular dichroism spectroscopy (XMCD) in combination of sum rules [64, 65, 66] were used to calculate the magnetic moment of Gd and Co at different temperatures without an external magnetic field. Measurements were performed while temperature was decreased from room temperature to 10 K and increased back to room temperature. At each temperature, XMCD spectra at the Co L₂,₃ edges and the Gd M₄,₅ edges were measured with an x-ray incident angle of 20° or 70° away from the sample plane to characterize the almost "in-plane" and "out-of-plane" magnetization. The number of holes (nₜ) nₜ₋ₐₗ₉ₒ = 2.5 for 3d metal Co, and nₜ₋ₐₗ₉ₒ = 7 for 4f metal Gd were used for sum rules calculations, explained in detail in Chapter 3.
Figure 6.2: PEEM images of AFM-coupled bubble skyrmion pairs at room temperature and zero field. 

**a**, Schematic of PEEM geometry. The yellow arrow indicates the x-ray propagation direction \( \mathbf{k} \) with an incident angle of 30° degree. 

**b** and **c**, PEEM images acquired at the Co L\(_3\) and Gd M\(_5\) absorption edges at room temperature without external fields. The contrast of bright and dark corresponds to magnetization oriented along \(-z\) and \(+z\) directions, respectively. The field of view is 20 \(\mu m\). Note that the bright areas in Co layer and the dark areas in Gd layer are the core area of the bubble skyrmions with uniformly magnetized spins in out-of-plane direction. The Néel domain wall is beyond the imaging resolution.
6.2 AFM-coupled bubble skyrmion pairs at room temperature and zero field

PEEM images of stabilized skyrmions in \([\text{Co/Gd/Pt}]_{n=10}\) multilayers at room temperature and zero field are shown in Figure 6.2. The skyrmions have the same shapes in Co and Gd layers but with the opposite grey level contrast. The bright (dark) areas in Figure 6.2b correspond to the dark (bright) areas in Figure 6.2c, indicating that the magnetization of Gd is in the opposite direction than that of Co. Therefore, skyrmions in Co and Gd layers are AFM-coupled. AFM-coupled skyrmion pairs are formed in \([\text{Co/Gd/Pt}]_{n=10}\) multilayers at room temperature and zero field. The different shape and size of the bubble skyrmions are likely due to the spatial variation of the local magnetic properties, including anisotropy, magnetization and DMI due to the natural presence of impurities, defects, grain orientations and film roughness.

6.3 Spin reorientation of the core area of the AFM-coupled bubble skyrmion pairs

We further investigate the effect of temperature on the AFM-coupled bubble skyrmions. During cooling down from room temperature, the skyrmions remain the same without showing obvious changes till 55 K, then become more and more faded at 50 K and 48 K and finally disappear at 45 K and below, as shown in the images at the top within the blue frame in Figure 6.3. When temperature is increased back, the disappeared skyrmions recover, as shown in the images at the bottom within the red frame in Figure 6.3. These core area of the AFM-coupled bubble skyrmion pairs remain stable over a large range of temperature from room temperature to 55 K without an external field, disappear at below 45 K, and recover themselves when temperature is increased back to above 50 K.

Figures 6.3a-d are the temperature-dependent magnetic moments of Co and Gd atoms (\(m_{\text{Co}}\) and \(m_{\text{Gd}}\)), measured with the x-ray beam incoming at 20 and 70 degrees. Because XMCD probes the magnetization along the x-ray prorogation direction, the measured magnetic moment, shown in Figures 6.3a-b, with the 20-degree incident angle can reflect the
Figure 6.3: Observation of bubble skyrmion core area disappearance and recovery during spin reorientation at zero field in Gd layer. Scale bar in the PEEM image is 5 \( \mu \)m. PEEM images with blue (red) background are the images taken at different temperatures as temperature decreases (increases). All images are taken at Gd M\(_5\) edge. \( \mathbf{a-d} \), XMCD results of calculated magnetic moment per Co and Gd atom along x-ray propagation direction at different temperatures and zero field. Each data point is the calculated magnetic moment per atom based on XMCD spectra and sum rules. The lines are for eye guidance. The open and solid symbols are the results when x-ray is incident at an angle of 20 and 70 degrees, respectively. Green and red symbols in \( \mathbf{a} \) and \( \mathbf{c} \) are the results of Gd moment during cooling down and warming up, respectively. Orange and purple symbols in \( \mathbf{b} \) and \( \mathbf{d} \) are the results of Co moment during cooling down and warming up, respectively. \( \mathbf{a} \) and \( \mathbf{b} \) are the results when x-ray is incident at 20 degrees, therefore are the magnetic moment projected to almost in-plane direction. \( \mathbf{c} \) and \( \mathbf{d} \) are the results when x-ray is incident at 70 degrees, therefore are the magnetic moment projected to almost out-of-plane direction. The curving arrows in \( \mathbf{a-d} \) indicates the temperature changing direction of different curves with different colors.
almost in-plane ("in-plane") component of the magnetization while the results in Figures 6.3c-d mainly indicate the almost out-of-plane ("out-of-plane") component of the magnetization. It is noticed that, at all the temperatures and for both "in-plane" and "out-of-plane" components, $m_{Co}$ is always AFM-coupled to $m_{Gd}$, indicated by an opposite sign. At 300 K, the "in-plane" $m_{Co}$ and $m_{Gd}$ are smaller than $0.2 \, \mu_B$ (Figures 6.3a-b) while the "out-of-plane" $m_{Co}$ and $m_{Gd}$ are a lot larger with $m_{Gd} \approx 0.3 \, \mu_B$ and $m_{Co} \approx 0.6 \, \mu_B$ (Figures 6.3c-d).

As temperature decreases to about 60 K, $m_{Co}$ remains unchanged (Figures 6.3b,d) but $m_{Gd}$ keeps increasing (Figures 6.3a,c). This temperature-dependent $m_{Co}$ and $m_{Gd}$ between 60 K and 300 K is because this temperature range is way below the Curie Temperature ($T_C$) of bulk Co (about 1300 K) and right below that of bulk Gd (about 300 K) for [92]. The "out-of-plane" $m_{Gd}$ increases from about 0.3 to 0.8 $\mu_B$ (green curve in Figure 6.3c) and $m_{Co}$ remains stable at about -0.6 $\mu_B$ (orange curve in Figure 6.3d). At about 100 K, $m_{Gd}$ is about the same magnitude but opposite sign as $m_{Co}$ not only in "out-of-plane" direction but in the "in-plane" direction. The SkHE is expected to be completely cancelled at the angular momentum compensation temperature ($T_A$), at which angular momentum of the Co atoms equals to the that of the Gd atoms ($A_{Co} = A_{Gd}$) [93]. $A_{Co} = \frac{m_{Co}}{\gamma_{Co}}$ and $A_{Gd} = \frac{m_{Gd}}{\gamma_{Gd}}$ with gyroscopic ratio $\gamma_{Co} = g_{Co}\mu_B/\hbar$, $\gamma_{Gd} = g_{Gd}\mu_B/\hbar$ and $g$ factors $g_{Co} \approx 2.2$ and $g_{Gd} \approx 2.0$ [93]. Therefore, $T_A$ is the temperature where $\frac{m_{Gd}}{g_{Gd}}/\frac{m_{Co}}{g_{Co}} \approx 1$, which is about 120 K in [Co/Gd/Pt]$_{n=10}$ multilayers. Considering that AFM-coupled bubble skyrmions remain stable above 55 K, we can conclude that AFM-coupled bubble skyrmions with zero net angular moment are stabilized in [Co/Gd/Pt]$_{n=10}$ multilayers at $T_A \approx 120$ K. The cancellation of SkHE should be observed with future current-driven investigation.

Below 60 K, the "in-plane" $m_{Co}$ (Figure 6.3b) starts to increase while the "out-of-plane" $m_{Co}$ begins to decrease (Figure 6.3d). The same thing happens to $m_{Gd}$, suggesting that the magnetization of both Co and Gd start to rotate from the original out-of-plane to in-plane direction as temperature decreases. This spin reorientation starts at about 60 K, then becomes more and more pronounced as temperature keeps decreasing to about 20 K
and finally remains fully in-plane below 20 K. Note that the "out-of-plane" $m_{\text{Gd}}$ increases (Figure 6.3b) below 50 K. This is the effect of the combination of the increasing of the total magnitude of Gd moment as background trend and the rotating of the magnetic moment away from the original out-of-plane direction. As temperature decreases, the magnetization of Gd rotates away from out-of-plane direction but its magnitude increases simultaneously, resulting the "out-of-plane" component of the increased but more away magnetization still being larger than the "out-of-plane" $m_{\text{Gd}}$ at higher temperatures.

Due to the spin reorientation, the magnetic moment of Co is about 0.4 $\mu_B$ in both "in-plane" and "out-of-plane" directions (Figures 6.3b,d) at 50 K, where the core of the bubble skyrmions start to fade out. As the magnetization becomes more and more "in-plane" below 50 K, the core of the bubble skyrmions disappear. Therefore, the disappearance of the core of the bubble skyrmions is due to the spin reorientation form the original out-of-plane direction to in-plane. As temperature increases back, the direction of magnetization changes from in-plane to original out-of-plane and the core area of bubble skyrmions recover.

The spin configuration of a skyrmion is a result of magnetic energy minimization at the corresponding temperature [88, 17]. In $[\text{Co/Gd/Pt}]_{n=10}$ multilayers, this energy minimization mainly includes the competition among the interfacial DM interaction at Gd/Co, Co/Pt, Pt/Gd interfaces [93], short-range exchange interaction among adjacent Gd-Gd, Co-Co and Co-Gd atoms [92], the long-range dipolar interaction and the magnetic anisotropic anisotropy energy [94, 95]. Above 60 K, the perpendicular magnetic anisotropy (PMA) is strong [94]. The magnetization direction of Co atoms favors out-of-plane direction. With the existence of a large dipolar interaction, to minimize the total energy, the uniform out-of-plane magnetization is broken into magnetic bubble domains with magnetization pointing up or down. The domain wall width is mainly determined by the magnetocrystalline anisotropy energy and the exchange energy of Co atoms. Due to the strong net interfacial DMI, the domain wall favors the energy minimum Néel chiral domain wall. Therefore, the skyrmion state is the magnetic state of the Co layer with a local energy minimum. Since
room temperature is close to the $T_C$ of Gd, the Heisenberg exchange interaction between Gd atoms is weak, compared to the AFM exchange interaction between the Co and Gd atoms. As a result, the magnetization of Gd at the interface is aligned to out-of-plane direction but opposite to that of the Co atoms. However, the magnitude of the AFM exchange interaction decays very fast as the distance between Gd and Co atoms become larger. Therefore, Gd atoms that are not next to Co atoms is mainly affected by the exchange interaction and magneto-crystalline anisotropy energy of the Gd atoms, favoring magnetization direction randomly away from out-of-plane, causing the net magnetization of Gd smaller from the saturation magnetization. As temperature decreases, the exchange interaction between Gd atoms becomes stronger and all the Gd atoms become more and more aligned. Therefore, those Gd atoms that are far away from Co atoms become more aligned to the out-of-plane direction, resulting the increasing of the average magnetization of Gd.

On the other hand, due to the increase of the exchange interaction between the Gd atoms as temperature decreases, at a certain temperature, the exchange energy becomes so large that the in-plane magnetization is more favored in terms of energy minimization in the Gd layer. This certain temperature is considered as spin reorientation temperature ($T_R$), which is around 60 K for the $[\text{Co/Gd/Pt}]_{n=10}$ multilayers. In this new state of local energy minimization, the Gd atoms give up the out-of-plane magnetization and start to align randomly towards in-plane. The changes in the direction of magnetization towards in-plane direction are controlled by the exchange interaction and magneto-crystalline anisotropy energy of Gd. Due to the very strong AFM exchange coupling between the Co and Gd atoms, PMA surrenders and Co atoms are forced to be aligned in the opposite direction than that of the Gd atoms towards the film plane. The magnetization becomes more and more towards in-plane and finally, completely falls into in-plane alignment at about 20 K where the in-plane magnetic moment of Co stops increasing (Figure 6.3b). When temperature increases back, the spin reorientation process reverses.
6.4 Persistence of chiral domain walls

To investigate the change of bubble skyrmions during spin reorientation, we focus on analyzing one specific bubble skyrmion. Very importantly and interestingly, the persistence of the chiral domain walls during the temperature changes was observed in the PEEM results, as shown in Figure 6.4. As temperature decreases to 50 K, the uniformly magnetized core area of this bubble skyrmion starts to break into multiple smaller domains. The bubble skyrmion is still observable at this temperature because the multiple domains within the core area still share similar contrasts with slight differences. As temperature keeps decreasing, the core area is not obvious any more, replaced by small areas with dark and bright contrasts. The same thing happens to the uniformly magnetized domain outside the core area. Based on XMCD results, we know at these temperatures the magnetization is aligned more towards in-plane direction. During spin reorientation, without seeing the uniformly magnetized core areas of bubble skyrmions, one can still see the domain wall of this bubble skyrmion in the PEEM images, highlighted by the dashed red arrows. We believe the domain wall should be a Néel domain walls based on the interfacial DMI existing in this system, although experimentally imaging is beyond the PEEM resolution.

To further investigate the persistence of the domain wall and show the evidence for chiral Néel domain walls of bubble skyrmions, micromagnetic simulations were performed and the results are shown in Figures 6.5 and 6.6. Figure 6.5a shows the simulated spin configuration at 160 K, where out-of-plane is the preferred magnetization direction. The magnetization of the white and black areas are along +z and -z direction, respectively. The rainbow colors at the boundary of the bubble skyrmions indicate magnetization along different in-plane directions. The out-of-plane spins rotate to the in-plane directions at 30 K, shown in Figure 6.5b, with different colors representing different in-plane directions. Figure 6.5c shows the comparison of the spin configuration at 160 K (before spin reorientation) and 30 K (after spin reorientation). From this comparison, we can tell that the uniformly
Figure 6.4: Observation of the persisted chiral domain walls by PEEM imaging. Scale bar in the top left image is 1 $\mu$m. The images with blue background in the top two rows are the PEEM images of one skyrmion taken at Gd M$_5$ and Co L$_3$ edges at various temperatures during cooling down from room temperature to 33 K. Only some of the results at the representative temperatures are shown here. The blue arrow from left to right indicates the temperature cooling direction. The PEEM images with red background at the bottom row are the images taken at Gd M$_5$ during warming up from right to left as indicated by the red arrow. The dotted red arrows in all the images surround the bubble skyrmion and highlight the wall area of the bubble skyrmions.
Figure 6.5: Micromagnetic simulations: persistence of in-plane magnetization at the wall area. 

- **a**: Magnetic domain pattern at 160 K in Gd layer. 
- **b**: Magnetization configuration at 30 K in Gd layer. 
- **c**: The overlapping of 40% transparency of the area within dotted red lines and the area within the black dotted lines in **b**. Note that the highlighted area in **b** is the corresponded area of the highlighted area in **a** at 30 K. The small triangles indicate the local magnetization direction. The in-plane magnetization at the wall area (boundary) of the bubble in **a** and the magnetic configuration at the same position in **b** are very similar. 
- **d**: is the same as **a**. 
- **e**: is the z component of **b**. 
- **f**: Magnetic domain pattern at 160 K in Gd layer after going through spin reorientation. 
- **g**: The overlapping of **e** and 40% transparency of **d**. 
- **h**: The overlapping of **e** and 40% transparency of **f**.
magnetized core area of the bubble skyrmion breaks into smaller domains with different in-plane magnetization directions, indicated by the different colors and the triangles pointing at different directions. However, the domain wall areas of the bubble skyrmions at both 160 K and 30 K have almost the same chiral Néel domain walls configurations. That is to say, the original chiral Néel domain walls remain unchanged during the spin reorientation.

We further compare the out-of-plane magnetization and the results are shown in Figure 6.5d-h. Figure 6.5g shows the comparison of the spin configuration at 160 K (before spin reorientation) and 30 K (after spin reorientation). The original Néel domain walls of the big white bubble skyrmion highlighted in Figure 6.5a are in between a dark purple line and a yellow line with only a few obvious points, which suggests the expansion of Néel domain walls into a wider wall with the magnetization changes from $M_z = -1$ (shown as dark purple) to $M_z = +1$ (shown as yellow) in the $z$ component. The narrow boundary of a bubble skyrmion become a larger ring-like magnetic configuration. In Figure 6.5h, the Néel domain wall of the recovered bubble skyrmion is still in between a yellow and purple line, indicates that as the temperature warms back to 160 K, the wider walls in between $M_z = -1$ (shown as dark purple) to $M_z = +1$ (shown as yellow) shrink to the Néel type domain wall of the recovered bubble skyrmion.

As we know, the short-range exchange interactions between neighboring Gd atoms changes when temperatures changes, affecting the effective anisotropy energy and total magnetization. To minimize the local energy, the magnetization of Gd and Co changes towards in-plane (out-of-plane) when temperature decreases (increases) and the system enters different local energy minimal wells. However, no matter how the uniformly magnetized domain area changes, the chiral Néel domain walls formed originally at room temperature remain stable and are not affected by the magnetization changes caused by temperature changes. During the energy changes affected by temperatures, the energy barrier of topologically preserved chiral domain walls remains unbreakable, which is understandable considering the spins rotate to in-plane direction without any preferred directions at zero-field.
That is to say, there are two factors contributing to the persistence of chiral domain walls. One of them is the topological protection, which tends to maintain the original chiral domain walls and the other is the absence of other preferential direction, such as an external magnetic field. Due to the persisted chiral Néel domain walls, the magnetic skyrmions recover back to its original places as temperature increases back. The magnetic configuration goes back to the minimum energy state of bubble skyrmions again.

Figure 6.6 shows the calculation of the skyrmion topological charge at both 160 K before and after going through spin reorientation. The topological charge remains constant $Q=1$ even tough the shape of the skyrmions become slightly different. The colors at the wall areas are very similar for one-on-one comparison of the $x$, $y$ and $z$ before and after spin reorientations (Figures 6.6a, d and 6.6b, e and 6.6c, f).

Micromagnetic simulations were also performed with different intensity of DM interaction to investigate the effect of DMI on spin reorientation and Néel domain walls. The results are shown in Figure 6.7. If DMI = 0, the out-of-plane magnetization still becomes in-plane at 30K. After warmed back, not only the domain wall are not chiral domain wall, the domain patterns do not look like the original ones at all. When we increase the DMI value to 0.5 $mJ/m^2$, the system has Néel-type domain wall but the domain patterns do not recover after being warmed back. However, when we increase the DMI value to 2.5 $mJ/m^2$, the domain patterns recover to its original ones after going through spin reorientation. Therefore, we can conclude that the interfacial DMI plays a very important role in the recovery of magnetic bubble skyrmions after spin reorientation.

6.5 Conclusions

In this chapter, we report the results of zero-field AFM-coupled bubble skyrmions pairs at room temperature in [Co/Gd/Pt]$_{n=10}$ multilayers, the study of the stability of AFM-coupled bubble skyrmions pairs between room temperature and 33 K and the investigation of reasons behind the recovery of AFM-coupled bubble skyrmions after spin reorientation.
Figure 6.6: Micromagnetic simulations: persistence of topological charge during spin re-orientation. 

- **a-c**, The $x$, $y$ and $z$ component of the skyrmion bubble at 160 K in Gd layer before going through spin reorientation, respectively. 
- **d-f**, The $x$, $y$ and $z$ component of the recovered skyrmion bubble after spin reorientation in Gd layer. The calculated topological charge is 1 for both skyrmion bubbles before and after going through spin reorientation even though the shape is slightly different.
Figure 6.7: Micromagnetic simulations: the effect of DMI when $K_{eff} = 4.9 \times 10^5$ J/m$^3$. First row: relaxed magnetic domain patterns at 160K. Second row: relaxed magnetic domain pattern after cool down to 30 K. Bottom row: relaxed magnetic domain patterns at 160 K after warm up from 30 K. Orange and green boxes are for Co and Gd respectively. Three DMI values were investigated and the left two, middle two and right two columns of Co and Gd are the results with the DMI = 0, DMI = 0.5 and 2.5 mJ/m$^2$ respectively.
The AFM-coupled Co and Gd skyrmion pairs are stable over a large temperature range from about 300 K to 55 K, including the compensation temperatures $T_m=100$ K and $T_A=120$ K. At 55 K, the magnetization of the core area of the skyrmion pairs starts to rotate from the original out-of-plane direction to in-plane. This spin reorientation transition becomes more pronounced when the temperature is decreased further to 33 K. However, the domain wall regions are preserved during this spin reorientation transition, and after the temperature is increased back to above 50 K, AFM-coupled bubble skyrmions are recovered to their original states before the spin reorientation transition. The observed temperature-dependent spin configuration changes in the [Co/Gd/Pt] multilayers can be attributed to the competition among the ferromagnetic exchange coupling within the Gd spins and Co spins, antiferromagnetic exchange coupling between the interfacial Gd and Co spin, and the interfacial DMI. The recovery of the skyrmions can be attributed to the persisted chiral domain walls due to the strong topological protection by DMI and the absent of other preferential in-plane direction. Micromagnetic simulations suggest that 1) the in-plane component of the Néel domain wall is persisted during spin reorientation, 2) the Néel domain could expand the shrink during spin reorientation, 3) the topological charge is persisted during spin reorientation and 4) a strong DMI is necessary for the skymion memory phenomenon. Our studies not only achieved the zero-field stabilized AFM-coupled bubble skyrmion pairs but demonstrate that the domain walls of the skyrmions are topologically preserved during the spin reorientation due to the temperature change which can be the reason for the recovery of skyrmion states. These results help us understand the energy competition in AFM-coupled skyrmions, open a possibility for the application of antiferromagnetic skyrmions in information encryption and decoding, as well as the race-track memory.

**References of Chapter 6**

[18] L. Caretta, M. Mann, F. Büttner, K. Ueda, B. Pfau, C. M. Günther, P. Hessing, A.
Churikova, C. Klose, M. Schneider, D. Engel, C. Marcus, D. Bono, K. Bagschik, S.


[88] M. Stier, W. Häusler, T. Posske, G. Gurski, and M. Thorwart, Physical Review Let-


CHAPTER 7
CONCLUSIONS

In this dissertation, the author reports the investigation of magnetic skyrmions in multilayers with the interfacial Dzyaloshinskii-Moriya interaction (DMI). First, [Pt/Co/HM]_n (HM = W, Mn, Ir, Au, n = 1, 3, 5, 8, 10, 12) multilayers were designed and fabricated to study the effect of the interfacial DMI, dipolar interaction and their interplay with other interactions on the stabilization of magnetic skyrmions. Second, current-driven skyrmion motion was studied in the Ta/CoFeB/TaO_x tri-layer. Finally, to cancel the undesirable skyrmion Hall effect in the current-driven skyrmion motion, Antiferromagnetically-coupled (AFM-coupled) bubble skyrmion pairs at room temperature were realized in [Co/Gd/Pt]_10 multilayers without the application of a magnetic field. The stability of the AFM-coupled bubble skyrmion and the chiral domain walls were investigated between 33 K and 300 K.

Multilayers with the interfacial DMI including [Pt/Co/HM]_n (HM = W, Mn, Ir, Au, n = 1, 3, 5, 8, 10, 12), Ta/CoFeB/TaO_x and [Co/Gd/Pt]_10 were successfully fabricated by the DC magnetron sputtering method. X-ray small angle reflectivity analyses confirmed the deposited multilayer structures as designed. Hall bar devices for current-driven studies were patterned using photolithography. The magnetic properties of the multilayer films were measured using magnetometry and x-ray magnetic circular dichroism spectroscopy (XMCD). Magnetic skyrmions as well as their field-dependence, temperature-dependence, and current-driven motion were directly imaged by synchrotron x-ray photoemission electron microscopy (PEEM), Lorentz-transmission electron microscopy (L-TEM), and magneto-optical Kerr effect microscope (MOKE).

The formation and stabilization of magnetic skyrmions without external magnetic fields are determined by the competition of the Heisenberg exchange interaction, dipolar interaction, magnetic anisotropy, and interfacial DMI. In [Pt/Co/HM]_n (HM = W, Mn, Ir, Au,
$n = 1, 3, 5, 8, 10, 12$) multilayers, DMI and dipolar interaction were tuned by changing the heavy metal (HM) and repetition number $n$, respectively. The size of the stabilized skyrmions decreased as DMI or dipolar interaction increased. Due to the strong spin-orbit coupling in Pt and HM, together with the broken inversion symmetry, there existed a substantial interfacial DMI that was necessary to stabilize the Néel-type skyrmions. Indeed, the existence of Néel-type skyrmions was confirmed by L-TEM imaging in the $(Pt/Co/W)_8$ multilayers, at room temperature in the absence of field. The magnitude of the interfacial DMI was estimated to be about $D = 1.5 \pm 0.2$ mJ/m$^2$ based on the evolution of the skyrmion size as a function of the perpendicular field. $D$ can be increased to $D = 2.1 \pm 0.2$ mJ/m$^2$ and $D = 2.2 \pm 0.2$ mJ/m$^2$ by changing W to Au and Ir. No skyrmions can be stabilized in $(Pt/Co/Mn)_n$ multilayers. The decrease of skyrmion size from a few $\mu$m to about 100 nm was realized by increasing $n$ from 3 to 8.

Current-driven skyrmion motion was investigated in Ta/CoFeB/TaO$_x$ multilayer and the skyrmion Hall effect was observed. When driven by an electric field, ferromagnetic skyrmions deviated from the current direction and moved towards the device edge. The transverse motion can be quantized by the skyrmion Hall angle, the angle between the current direction and the skyrmion motion direction. Combining the experimental results and the theoretic derivation based on the Thiele equation, we attribute the skyrmion Hall effect to the topological Magnus force and random pinning potentials in the materials. A significant current/velocity dependence of the skyrmion Hall angle was observed. For instance, below a specific current density, the skyrmions show a hopping-like motion with a zero skyrmion Hall angle. At higher current densities, a stronger driving force allows skyrmions to overcome pinning potentials gradually and eventually leads to the saturation of the skyrmion Hall angle. It is also observed that skyrmions, with opposite topological charges, accumulate at the opposite side of the edges, in analogs to the charge accumulation in the ordinary Hall effect. By changing the sign of the topological charge $Q$ and electric current, a substantial similarity between the conventional Hall effect of the elec-
tronic charge and the Hall effect due to the topological charge has also been revealed.

To prevent the skyrmions from moving towards the device edge, where they could annihilate, AFM-coupled skyrmion pairs were realized and investigated. \([\text{Co/Gd/Pt}]_{10}\) multilayers were engineered to host these AFM-coupled skyrmion pairs, a bi-layer pair of AFM-coupled skyrmions with \(Q = 0\). PEEM imaging reveals that the AFM-coupled Co and Gd skyrmion pairs are stable over a broad temperature range from about 300 K to 55 K. At 50 K, the magnetization of the core area of the skyrmion pairs starts to rotate from the original out-of-plane direction to in-plane. This spin reorientation transition (SRT) becomes more pronounced when the temperature is decreased further to 33 K. However, the domain wall regions are preserved during this SRT. After the temperature is increased back to above 50 K, the same skyrmions are recovered to their identical state before the SRT, as if the skyrmions have memory. The recovery of the skyrmions can be attributed to the persisted chiral domain walls due to the strong topological protection by the interfacial DMI. Micromagnetic simulations suggest that 1) the in-plane component of the Néel domain wall is persisted through spin reorientation, 2) the Néel domain could expand the shrink during spin reorientation, 3) the topological charge is persisted through spin reorientation and 4) a strong DMI is necessary for the skyrmion memory phenomenon. The observed temperature-dependent spin configuration changes in the \([\text{Co/Gd/Pt}]_{10}\) multilayers can be attributed to the competition among the ferromagnetic exchange coupling within the Gd spins and Co spins, antiferromagnetic exchange coupling between the interfacial Gd and Co spin, and the interfacial DMI.

This dissertation revealed many interesting properties and phenomena of magnetic skyrmions in multilayers with the interfacial DMI. First, it shows that skyrmion formation is sensitive to the relative magnitudes of these effects and represents an important step towards understanding the conditions necessary for stabilizing room-temperature magnetic skyrmions. Then, the observed skyrmion Hall effect during current-driven motion not only provides fundamental physics, guides the race-track memory design but also indicates that
the topological charges of magnetic skyrmions can be potentially integrated for realizing novel functionalities, such as topological sorting. Finally, AFM-coupled skyrmion pairs were successfully realized as the solution to cancel the undesired skyrmion Hall effect for the application of skyrmion race-track memory. The recovery of bubble skyrmions after SRT can be potentially used for information encryption and decoding.
List of Publications


Manuscripts under preparation


"Magnetic exchange interactions between Fe$^{3+}$ and R$^{3+}$ in epitaxial h-RFeO$_3$ (R = Yb, Ho) thin films" Xiao Wang, Zhuyun Xiao, Yaohua Liu, Kishan Sinha, Xiaoshan Xu, Wenbin Wang, David Keavney, and X. M. Cheng.
Appendices
A magnetic skyrmion in multilayers (a planar geometry) is characterized by a topological charge, that is, the skyrmion winding number. The topological charge is given by

$$Q = \frac{1}{4\pi} \int d^2 r \rho_{\text{sky}}(r)$$

where the topological charge density \( \rho_{\text{sky}}(r) \) is given by

$$\rho_{\text{sky}}(r) = \frac{1}{4\pi} m(r) \cdot (\partial_x m(r) \times \partial_y m(r))$$

with the unit vector in the direction of the local magnetization. Thus, the topological charge of the magnetic skyrmion is

$$Q = \frac{1}{4\pi} \int m(r) \cdot (\partial_x m(r) \times \partial_y m(r)) d^2 r$$

which is also referred to as the Winding number. It counts how many times \( m(r) \) is wrapped around the unit sphere as the coordinates \((x, y)\) span the whole planar space.

A point of the planar space is parameterized as \( x = r \cos \varphi, y = r \sin \varphi \). By applying the mapping \( r = 0 \) as \( z \to -1, r = 1 \) as \( z = 0, r \to \infty \) as \( z \to 1 \), and \( \lim_{r \to \infty} m(x, y) = \lim_{z \to 1} m(z, \phi) \), we consider the compactification of the planar space to a sphere in the \( xyz \)-space parameterized by \( x = \sqrt{1 - z^2} \cos \phi(\varphi) = \sqrt{1 - \cos^2 \theta} \cos \phi, y = \sqrt{1 - z^2} \sin \phi(\varphi) = \sqrt{1 - \cos^2 \theta} \sin \phi, \) and \( z = \cos \theta \). Here \( z \) is defined by \( r = \frac{1+z}{1-z} = \frac{1+\cos \theta}{1-\cos \theta} \).

Hence, we can write

$$m(r) = m(\theta, \phi) = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$$

By substituting equation (A.2) into equation (A.1), we obtain

$$Q = \frac{1}{4\pi} \int_0^\pi \sin \theta d\theta \int_0^{2\pi} d\phi = -\frac{1}{4\pi} \left[ \cos \theta \right]_0^{2\pi} \left[ \phi \right]_0^{2\pi} = -\frac{1}{2} \left[ \cos \theta \right]_0^{2\pi}$$

(A.3)
Thus, when the spins at $r \to \infty$ point towards the $+z$ direction while the spin at $r = 0$ points towards the $-z$ direction, $[\cos \theta]^0_\pi = 2$, resulting in the negative skyrmion number $Q = -1$. In the same way, when the spins at $r \to \infty$ point towards the $-z$ direction while the spin at $r = 0$ points towards the $+z$ direction, $[\cos \theta]^0_\pi = -2$, resulting in the positive skyrmion number $Q = +1$. 
The magnetic spin moment and orbital moment of an atom can be determined from the x-ray magnetic circular dichroism spectroscopy results according to sum rules. The equations for 3$d$ metals based on the $L$ edge absorption and 5$d$ metals based on the $M$ edge absorption are shown in the figures below.
For 3d metals

\[
\begin{align*}
    m_L &= 2n_h \int_{L_3}^{L_2} (\mu^+ - \mu^-) dE \\
    m_L &= 2n_h \int_{L_3}^{L_2} (\mu^+ + \mu^- + \mu^0) dE \\
    m_S + m_T &= 3n_h \int_{L_3}^{L_2} (\mu^+ + \mu^- + \mu^0) dE \\
    \text{Spin} \\
    \text{Orbital} \\
    \text{with } \mu^0 = \frac{\mu^+ + \mu^-}{2} \text{ good for metals}
\end{align*}
\]

\[
\begin{align*}
    m_L &= \frac{4}{3} n_h \int_{L_3}^{L_2} (\mu^+ + \mu^-) dE \\
    m_L &= \frac{4}{3} n_h \int_{L_3}^{L_2} (\mu^+ + \mu^-) dE \\
    m_S + m_T &= 2n_h \int_{L_3}^{L_2} (\mu^+ + \mu^-) dE \\
    \text{Spin} \\
    \text{Orbital} \\
    \text{with } \mu^0 = \frac{\mu^+ + \mu^-}{2} \text{ good for metals}
\end{align*}
\]

Figure B.1: Sum rules for 3d metals
For 5d metals

\[
\begin{align*}
 m_L &= -3n_{h} \frac{\int_{M_4+M_5} (\mu^+ - \mu^-) dE}{\int_{M_4+M_5} (\mu^+ + \mu^- + \mu^0) dE} \quad \text{Orbital} \\
 m_S + m_T &= -3n_{h} \frac{\int_{M_5} (\mu^+ - \mu^-) dE - 3 \int_{M_4} (\mu^+ - \mu^-) dE}{2 \int_{M_4+M_5} (\mu^+ + \mu^- + \mu^0) dE} \quad \text{Spin} \\
 \text{with } \mu^0 &= \frac{\mu^+ + \mu^-}{2} \quad \text{good for metals}
\end{align*}
\]

\[
\begin{align*}
 m_L &= -2n_{h} \frac{\int_{M_4+M_5} (\mu^+ - \mu^-) dE}{\int_{M_4+M_5} (\mu^+ + \mu^-) dE} \quad \text{Orbital} \\
 m_S + m_T &= -n_{h} \frac{\int_{M_5} (\mu^+ - \mu^-) dE - 3 \int_{M_4} (\mu^+ - \mu^-) dE}{\int_{M_4+M_5} (\mu^+ + \mu^-) dE} \quad \text{Spin}
\end{align*}
\]

Figure B.2: Sum rules for 5d metals
VITA

Xiao Wang was born on July 23rd, 1989 in Sheyang, Yancheng Province, the People’s Republic of China. In 2008, she attended Nanjing Normal University and majored in Physics. During her undergraduate study, she was awarded various honors, including the China Governmental Scholarship for Undergraduate Students and Nanjing Normal University President Scholarship. She graduated in 2012 with a B.S. degree. In September 2012, she joined the Ph.D. program in the department of Physics at Bryn Mawr College and started to work with Professor Xuemei May Cheng in Spintronics and Nanomaterials Lab. She received Dean’s Fellowship and Marguerite N. Farley Fellowship from Bryn Mawr College. She worked at Materials Science Division at Argonne National Laboratory as a visiting graduate student for two and a half years starting from February 2016. She has published over ten refereed articles during her Ph.D. study.