Magnetotransport studies of heterostructures based on transition metals and their oxides

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**MAGNETOTRANSPORT STUDIES OF
HETEROSTRUCTURES BASED ON TRANSITION
METALS AND THEIR OXIDES**

HUANG KE

SCHOOL OF PHYSICAL AND MATHEMATICAL SCIENCES

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HETEROSTRUCTURES BASED ON TRANSITION
METALS AND THEIR OXIDES**

HUANG KE

SCHOOL OF PHYSICAL AND MATHEMATICAL SCIENCES

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

2021

Statement of Originality

I hereby certify that the work embodied in this thesis is the result of original research done by me except where otherwise stated in this thesis. The thesis work has not been submitted for a degree or professional qualification to any other university or institution. I declare that this thesis is written by myself and is free of plagiarism and of sufficient grammatical clarity to be examined. I confirm that the investigations were conducted in accord with the ethics policies and integrity standards of Nanyang Technological University and that the research data are presented honestly and without prejudice.

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Authorship Attribution Statement

This thesis contains material from 3 papers published in the following peer-reviewed journal(s) and 1 paper in submission in which I am listed as an author.

Part of Chapter 3 is in submission as M. Ma,#, **K. Huang**,#, Y. Li, S. Li, Q. Feng, C. C. I. Ang, T. Jin, Y. Lu, Q. Lu, W. S. Lew, F. Ma, X. Renshaw Wang. (# equal contribution). Nano-engineering the evolution of skyrmion crystal in synthetic antiferromagnets.

The contributions of the co-authors are as follows:

- Prof. X. Renshaw Wang and Prof. F. Ma designed and guided this project.
- M. Ma conducted all the MOKE measurements.
- I conducted all the magnetotransport measurements.
- I analyzed all the experimental results and prepared the manuscript.
- Q. Feng conducted the MFM measurements.
- S. Li prepared the samples.
- All authors contributed to the discussion of this project and edited the manuscript.

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The contributions of the co-authors are as follows:

- Prof. F. Ma designed and guided this project.
- Y. Li conducted the MOKE measurements.
- Dr. S. Li prepared the samples.
- Q. Feng conducted the MFM measurements.

- Prof. X. Renshaw Wang guided the magnetotransport measurements.
- I conducted all the magnetotransport measurements and analyzed the magnetotransport results.
- All authors contributed to the discussion of this project and edited the manuscript.

Part of Chapter 4 is published as **K. Huang**[#], T. Wang[#], M. Jin, L. Wu, J. F. Wang, S. Li, D. Qi, S. Cheng, Y. Li, J. Chen, X. He, C. Li, S. J. Pennycook, and X. Renshaw Wang. ([#] equal contribution). Bipolar Conduction and Giant Positive Magnetoresistance in Doped Metallic Titanium Oxide Heterostructures, *Advanced Materials Interfaces* 8, 2002147 (2021). DOI: 10.1002/admi.202002147.

The contributions of the co-authors are as follows:

- Prof. X. Renshaw Wang designed and guided this project.
- I conducted all the transport measurements and prepared the manuscript drafts.
- Dr. T. Wang prepared the samples and conducted the chemical composition analysis.
- Prof. C. Li conducted the STEM and EELS measurements.
- All authors contributed to the discussion of this project and edited the manuscript.

Part of Chapter 5 is published as **K. Huang**[#], L. Wu[#], M. Wang, N. Swain, M. Motapothula, Y. Luo, K. Han, M. Chen, C. Ye, A. J. Yang, H. Xu, D. Qi, A. T. N'Diaye, C. Panagopoulos, D. Primetzhofer, L. Shen, P. Sengupta, J. Ma, Z. Feng, C.W. Nan, X. Renshaw Wang. ([#] equal contribution). Tailoring magnetic order via atomically stacking 3d/5d electrons to achieve high-performance spintronic devices. *Applied Physics Reviews* 7, 011401 (2020). DOI:10.1063/1.5124373

The contributions of the co-authors of this paper are as follows:

Abstract

To contribute to the next-generation electronic and spintronic devices, systematic investigations on emerging magnetotransport phenomena are in demand. Among the various condensed matter systems which might nurture these phenomena, the heterostructures based on the transition elements are intriguing. This thesis includes studies of three representative systems, namely (i) transition metal nanostructures, (ii) transition metal oxide heterostructures, and (iii) transition metal oxide superlattices. This thesis focuses on discovering novel longitudinal and transverse magnetoresistance at transition metal (oxides) based heterointerfaces and revealing their underlying charge transport mechanisms.

In the first system, we propose and demonstrate that the spin textures in synthetic antiferromagnetic Pt/Co-based nanostructure are controllable via structural engineering. Primarily through modulating the thickness of the bottom layers in the nanostructure, the transition from skyrmion bubble to antiferromagnetic or ferromagnetic spin textures could be artificially switched. More importantly, the nano-engineered structure arouses an asynchronous annihilation of skyrmion bubbles. By utilizing this asynchronicity, we realized multiple non-volatile resistance states.

In the second system, we achieve the modulation of charge carriers in $\text{TiO}_x\text{D}_y/\text{SrTiO}_3$ heterostructures from electron to bipolar (electron and hole) conduction. The heterostructures are fabricated by treating Ti film with deuterium (D) doping and surface/interface oxidation. The electron mobility of the D-doped sample is 10 times as large as that of the undoped one. More importantly, the emerging holes exhibit high mobility comparable to that of the electrons. Furthermore, we also observe a giant magnetoresistance (MR), more than 900% at 6 T, which intrinsically is induced by this bipolar conduction.

In the last system, we report the modulation of the strength and symmetry of the magnetic orders in $\text{LaMn}^{3+}\text{O}_3/\text{SrIr}^{4+}\text{O}_3$ superlattices. Different from the well-studied $\text{Mn}^{4+}/\text{Ir}^{4+}$ superlattices, the $\text{Mn}^{3+}/\text{Ir}^{4+}$ structure provides a stronger interfacial effect together with a unique polar discontinuity. An evolution of ferromagnetism is demonstrated and explained by playing with the competition of polar catastrophe and 3d/5d electronic coupling. Furthermore, the symmetry

of anisotropic magnetoresistance undergoes a transition from four-fold to two-fold, which is triggered by a gradually enhanced Rashba spin-orbit interaction.

This thesis validates that magnetotransport measurements can be used to investigate the unique electrical and magnetic properties. The systematic studies on transition metals and oxides demonstrate the feasibility of revealing emerging phenomena via building transition metal element-based heterostructures. Significantly, these emerging interfacial magnetotransport phenomena enable insights into future spintronic and electronic applications.

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List of Symbols and Abbreviations

Ψ	Digamma function
ε	Energy
ε_F	Fermi energy
λ_F	Fermi wavelength
τ	Relaxation time
μ	Mobility
μ_s	Electron dipole moment
μ_B	Bohr magnetron
σ	Conductivity
σ_{xx}	Longitudinal conductivity
σ_{xy}	Transverse conductivity
ρ	Resistivity
ρ_{xx}	Longitudinal resistivity
ρ_{xy}	Transverse resistivity
$\hbar$	Reduced Planck constant
ω_c	Cyclotron frequency
A	Surface area of a condensed matter system
a	Acceleration
B	Magnetic flux density
B_c	Coercivity
B_ϕ	Effective magnetic flux density converted from l_ϕ
B_s	Saturation magnetic flux density
B_{so}	Effective magnetic flux density induced by spin-orbit interaction
d_c	Cyclotron diameter
e	Elemental charge
$\mathbf{e}$	Easy axis direction of a ferromagnet
E	Electric field strength
F	Force
G	Conductance
G_0	Conductance quantum
H	Hamiltonian, total energy operator
I	Current
j	Current density
$\mathbf{k}$	Wavevector
$\mathbf{k}_F$	Fermi wavevector
L	Length of a condensed matter system
L_e	Orbit angular momentum
l	Length of the conduction channel
l_ϕ	Average length of an electron that itinerates without a phase change
l_{mfp}	Mean free path
l_{so}	Average length converted from B_{so}
M	Magnetization
M_s	Saturation magnetization
N	Total number of charge carriers

n	Charge carrier density
n_{1D}	one-dimensional charge carrier density
n_{2D}	two-dimensional charge carrier density
n_{3D}	three-dimensional charge carrier density
P	Possibility
p	Momentum
p_F	Fermi momentum
R	Resistance
R_0	Resistance quantum
R_s	Sheet resistance
R_{xy}	Transverse resistance
r	Position vector
S	Local angular momentum
s	Itinerant spin angular momentum
T	Temperature
t	Thickness of the conduction channel
V	Volume of a condensed matter system
v	Velocity
v_d	Drift velocity
v_F	Fermi velocity
w	Width of the conduction channel
x	x-direction unit vector
y	y-direction unit vector
z	z-direction unit vector
2DEG	Two-dimensional electron gas
AHE	Anomalous Hall effect
AMR	Anisotropic magnetoresistance
AST	Antiferromagnetic spin texture
CPU	Central processing unit
CFST	Canted ferromagnetic spin texture
DRAM	Dynamic random-accessed memory
ERDA	Elastic recoil detection analysis
FST	Ferromagnetic spin texture
HDD	Hard-disk drive
IP	In-plane
LMO	LaMnO ₃
MFM	Magnetic force microscopy
MG	Magnetoconductance
MR	Magnetoresistance
MOSFET	Metal-oxide-semiconductor field-effect transistor
OOP	Out-of-plane
RBS	Rutherford backscattering
SCIP	Standard commands for programmable instruments
SIO	SrIrO ₃
SOI	Spin-orbit interaction
SRAM	Static random-accessed memory
SSD	Solid-state drive

STT MRAM accessed memory	Spin-transfer-torque-based magnetoresistivity random-
VDP	van der Pauw
VTI	Variable temperature insert
WL	Weak localization
WAL	Weak antilocalization
XAS	X-ray absorption spectroscopy
XRD	X-ray diffraction
XMCD	X-ray magnetic circular dichroism

Chapter 1

Introduction

1.1 Research background and motivation

1.1.1 Research background

Boosted by research and industrial developments, smart electronic devices have gradually sneaked into our daily lives within the first 20 years of the 21st century [1]. Generally, a smart device is mainly composited with electronic components that exhibit binary states, *i.e.*, “0” and “1” [2]. The binary states allow an electronic component to imitate the functionalities of the human brain, such as calculation and memorization. Physically, the binary states are achieved by switching the resistance (R) of a system with two experimentally distinguishable values [2]. The switching speed, the energy required for the switching, and the volume of the condensed matter system are three typical parameters of these electronic components [3,4]. Different electronic devices have different requirements for these parameters. For example, in a modern personal computer, there are four functional modules, *i.e.*, Processor, Cache, Memory, and Storage [5]. From the Storage to the Processor, a higher R switching speed and a smaller volume are required. Accordingly, more significant energy consumption is unavoidable from the Storage to the Processor (Figure 1.1).

Intrinsically, R reflects the motions of charge carriers. In a condensed matter system, charge carriers include electrons and holes. The motion of charge carriers is referred to as charge transport. The study on controlling the charge transport for applications is called electronics. Metal-oxide-semiconductor field-effect transistor (MOSFET) is one of the most famous electronic applications [6]. It allows users to turn on/off the carrier motion via a gate voltage. Nowadays, the MOSFET-based electronic devices, such as central processing unit (CPU), static random-accessed memory (SRAM), dynamic random-accessed memory (DRAM), and solid-state drive (SSD), have been widely used to build the four modules of computers (Figure 1.1) [5]. In recent years, an increasing number of researchers start to focus on tuning the spin of charge carriers to achieve similar purposes as the electronics [7–10]. This spin-based electronics is referred to as spintronics. Up to now, spintronics has also bred valuable products, such as hard

disk drive (HDD) [11] and spin-transfer-torque-based magnetoresistivity random-accessed memory (STT MRAM) [10] for Storage and Memory modules, respectively (Figure 1.1). Especially, STT MRAM becomes a strong competitor to the traditional transistor-based DRAM due to its lower energy consumption [5].

Figure 1.1. Four functional modules in a modern computer [5].

To improve the performance of an electronic device, the most straightforward route is to increase the density of electronic components on the device. One example is the transistor density on commercially obtainable CPUs (Figure 1.2) [12]. From 1970 to 2020, this density almost follows an exponential increase. This remarkable achievement demonstrates the thriving modern semiconductor industry. However, two potential crises associated with this “density strategy” limit the future development of the semiconductor industry. Firstly, the skyrocketing of the density is achieved by shrinking the size of an electronic component. When any electronic component reaches the atomic scale, the tunnelling of charge carriers cannot be ignored. The original R may shift due to this tunnelling. Therefore, the expected “0” state might be misinterpreted as a “1” state, leading to the malfunction of the electronic component. If we keep

shrinking the size of electronic components, the cost to compensate for these malfunctions will be unaffordable. Secondly, the energy consumption rises together with the density of electrical components. Both the energy consumption itself and the accompanied heat dissipation are far from negligible.

Figure 1.2 Evolution of the transistor density on commercially obtainable CPUs [12].

1.1.2 Research motivation

New devices have been constantly proposed and tested to bypass the shortcomings of traditional devices. For example, STT-MRAM has become a strong competitor to the traditional transistor-based DRAM due to its lower energy consumption and faster switching speed [5]. While the current transistor-based devices are still the most widely used, is it too early to be devoted to a new framework? The answer would be “NO”. Figures 1.3a and 1.3b show the developments of MOSFET and STT MRAM, respectively. After the ideas were originally proposed, both of them underwent decades of studies before they were invented in the laboratory. It required several extra years to push the laboratory unit to the market. Behind every product on the market, years of theoretical, experimental, and industrial efforts are in demand. Emerging magnetotransport phenomena are the catalysts for the evolution of these devices. For example, the

discovery of giant magnetoresistance (GMR) became the milestone of STT-MRAM [7]. Therefore, if we are aiming at the devices working in the second half of the 21st century, more magnetotransport studies are in demand. Motivated by the development of modern electronic and spintronic devices, this thesis presents systematic studies on the emergent magnetotransport phenomena of transition metals and their oxides. The potential applications based on these phenomena are discussed.

Figure 1.3. The developing histories of MOSFET (a) [6,13,14] and STT MRAM (b) [7,15–18].

1.2 Literature review of charge transport

1.2.1 Overview of transport phenomena

When charge carriers itinerate, their trajectories may be deviated by the surroundings, such as defects and phonons. This deviation is referred to as scattering [19]. Depending on whether the charge carriers are or are not scattered, charge transport can be classified into ballistic or diffusive transport, respectively. Figure 1.4 shows an overview of ballistic and diffusive transport. The theory of ballistic transport is relatively simple because charge carriers itinerate independently. The theory of diffusive transport is much more complex. Different types of scattering centres have different contributions to transport and should be treated differently. Generally, three different types of treatments are utilized to analyze diffusive transport. The classical treatment is entirely based on classical

physics, with the scattering deemed as a collision. This treatment well explains the electron-electron and electron-impurity scatterings. Semiclassical treatment considers the correction from quantum effect and is usually used in the electron-phonon scattering or spin-dependent scattering. Quantum treatment is entirely based on quantum mechanics, with the electron motion explained as wave propagation. This treatment is used to analyze quantum phenomena such as localization. From classical to quantum, the physical model captures more details of charge transport while, at the same time, complicates the analysis of electrical phenomena.

Figure 1.4 Overview of ballistic and diffusive transport.

Charge transport can be easily disturbed. The magnetic field is one of the major disturbances. Charge transport in a magnetic field is named magnetotransport [20]. Each charge transport mechanism corresponds to a specific magnetotransport phenomenon. Therefore, magnetotransport serves as a tool to identify the underlying charge transport mechanism of a condensed matter system. A famous example is that the anomalous Hall effect (AHE) becomes an identification of the scattering induced by ferromagnetism [21].

The following sections introduce the basic concepts in charge transport studies and review both ballistic and diffusive transport. Especially, the magnetotransport of a condensed matter system with bipolar conduction, ferromagnetism, strong spin-orbit interaction (SOI) will be detailly elaborated.

1.2.2 Basic concepts

Before conducting the literature reviews, I start by elaborating on relevant physics concepts.

(1) Charge transport and band theory

In the viewpoint of quantum physics, a condensed matter system possesses different states. Each state is unique and can be determined by two parameters, *i.e.*, energy (ε) and wavevector ($\mathbf{k}$) [22]. A group of states with continuous ε and $\mathbf{k}$ forms an energy band. If we plot all the states in the graph, we obtain an ε - $\mathbf{k}$ diagram. The ε - $\mathbf{k}$ diagram is also referred to as band structure. ε - $\mathbf{k}$ diagram exists even if there are no electrons in a condensed matter system (Figure 1.5a). If we add electrons to a condensed matter system whose temperature (T) = 0 K, electrons will preferably occupy the states with lower ε . Each state allows two electrons with opposite spin directions. The momentum ($\mathbf{p}$) of an electron in a state ($\varepsilon, \mathbf{k}$) is calculated with $\mathbf{p} = \hbar\mathbf{k}$. When there are no empty states whose ε are lower than any occupied state, the condensed matter system is in its ground state. Experimentally, the ground state is obtained by cooling the condensed matter system. When T rises, the thermal energy allows electrons to occupy states with higher ε , leaving the lower- ε states empty. In this case, the condensed matter system is in an excited state.

The energy required to add an extra electron into a condensed matter system is the Fermi level (ε_F). In a metal (Figure 1.5b), the occupied band with the highest ε is only partially filled. ε_F crosses this band and is equal to the highest electron energy. The $\mathbf{k}$ and $\mathbf{p}$ associated with ε_F are the Fermi wavevector ($\mathbf{k}_F$) and Fermi momentum ($\mathbf{p}_F$), respectively. In an insulator (Figure 1.5c), all the bands with electrons occupied are full while all the other bands are empty. Strictly speaking, insulating only exists when the condensed matter system is in the ground state. In an insulator, ε_F sits in the middle of two adjacent bands and is not related to the energy of any electron. The fully occupied band below ε_F is called the valance band, while the empty band above ε_F is called the conduction band. The energy difference between the minimum ε of conduction and the maximum ε of valance band is called the bandgap. An insulator has a relatively large bandgap, which cannot be easily crossed by the electrons in the valance band. Note that in a metal, the ε_F does not separate two bands. The band crossed by ε_F could be considered as an overlapping of the conduction band and valance band.

The bandgap of a semiconductor is relatively smaller and could be crossed by the excited electrons from the valance band (Figure 1.5d). In a semiconductor, both the electrons in the conduction band and the holes (absence of electrons) in the valance band could contribute to the conduction. External modifications, such as chemical doping, allow a semiconductor to be pure electron conduction (Figure 1.5e) or hole condition (Figure 1.5f) by adding electrons to the conduction band or removing electrons from the valance band, respectively. Usually, un-doped semiconductors are called intrinsic semiconductors. Doped semiconductors with more electron or hole conduction are called n-type or p-type semiconductors.

Figure 1.5 Schematics of ϵ - k diagram of (a) a condensed matter system with no electron, (b) a metal, (c) an insulator, (d) a semiconductor, (e) an n-type semiconductor, (f) a p-type semiconductor. The dashed and bold lines represent the empty and occupied states in a band, respectively. The red dot line represents the Fermi level.

Even if the electrons in the conduction band are itinerant, it may not lead to a current (I). For example, in a metal film, electrons itinerate with p_F in the film plane randomly, leading to the $I = 0$. If we plot the occupied states along one axis of the condensed matter system, all these occupied states should form a square area (Figure 1.6a, bottom). Electrons in this area do not possess neat motion. However, if we connect the source and drain electrodes to the condensed

matter system and apply a potential difference (ΔV) (Figure 1.6b), the ε_F of the two electrodes splits. The electrons in the condensed matter system, whose energies are in between the ε_F of the two electrodes, obtain a neat motion. Accordingly, a current is generated.

Figure 1.6 Occupied states plotted along an axis of a condensed matter system when a ΔV is not (a) and is (b) applied.

(2) Quantitative characterization of the charge transport

In a condensed matter system, the average length of charge carriers that freely iternate is defined as the mean free path (l_{mfp}) [20]. The average time duration for this free iternation is defined as relaxation time (τ) [20]. l_{mfp} and τ are connected by the electron velocity (v) through $l_{mfp} = v \cdot \tau$. In a metal, v is equal to the Fermi velocity (v_F). The neat velocity obtained by charge carriers is defined as drift velocity (v_d) [20]. When no electric field is applied, $v \neq 0$ but $v_d = 0$. When an electric field with strength, E , is applied, charge carriers are forced to low potential energy end, leading to $v_d \neq 0$. Charge carrier mobility (μ) is defined as $\mu = v_d/E$ [20]. Usually, v_d is linear to E . Therefore, μ is independent of the E .

These physical quantities describe the motion capability of charge carriers from different perspectives. τ , l_{mfp} and v are defined from the condensed matter system itself, while μ and v_d are defined based on an applied electric field. However, they are intrinsically related. v_d can be modelled with Newton's second law, *i.e.*, $v_d = a \cdot \tau$, where a is the average acceleration of the charge carriers. When an electric field is applied to the condensed matter system, but no magnetic field is applied, a can be calculated with $a = eE/m$, where m is the effective mass of the charge carrier. In this thesis, e is the absolute value of the elemental charge. Therefore, we can link the v_d and τ with

$$v_d = \frac{eE\tau}{m}. \quad 1.1$$

Plugging the definition, $\mu = v_d/E$ into Equation 1.1, we obtain the relation between μ and τ ,

$$\mu = \frac{e\tau}{m}. \quad 1.2$$

The conductivity (σ) is related not only to the motion capability but also to the charge carrier density (n). We first define the total number of charge carriers as N . The three-dimensional (n_{3d}), two-dimensional (n_{2d}), and one-dimensional carrier density (n_{1d}) can be calculated with $n_{3d} = N/V$, $n_{2d} = N/A$, and $n_{3d} = N/L$, respectively. V , A , and L are the volume, surface area, length of the condensed matter system, respectively. Usually, n_{3d} is simplified as n . σ can be calculated via

$$\sigma = en\mu. \quad 1.3$$

When the magnetic field is not applied, resistivity (ρ) can be calculated via

$$\rho = \frac{1}{en\mu}. \quad 1.4$$

Equation 1.4 allows us to predict the temperature-dependent resistivity (ρ - T). When we cool an undoped semiconductor to its ground state, electrons in the conduction band drop to the unfilled states in the valance band. The n decreases down to 0. Therefore, we would observe an increasing ρ of a semiconductor when T drops. When T approaches 0 K, ρ approaches positive infinity. Differently, if we cool metals, the change of n may not be significant enough to dominate the ρ - T . Contribution from μ should be considered. We will explain the details in Chapter 1.24.

In a metal, the analytical expression between l_{mfp} and μ is obtainable. The definition of p_F gives $p_F = mv_F = \hbar k_F$. Plugging this equation and Equation 1.2 into $l_{mfp} = v_F \tau$, we obtain,

$$l_{mfp} = \frac{\hbar\mu}{e} k_F \quad 1.5$$

Equation 1.5 is a general method to calculate the l_{mfp} in a metal. This equation can be simplified depending on how we model the k_F . If we use parabolic band approximation and periodic boundary conditions, we can link k_F with n [23].

$$n_{1D} = \frac{2k_F}{\pi} \quad 1.6$$

$$n_{2D} = \frac{k_F^2}{2\pi} \quad 1.7$$

Plugging Equation 1.6 and 1.7 into 1.5, we can estimate l_{mfp} based on n and μ ,

$$l_{mfp} = \frac{\hbar\pi}{2e}\mu n_{1D} \quad 1.8$$

$$l_{mfp} = \frac{\sqrt{2\pi}\hbar}{e}\mu\sqrt{n_{2D}} \quad 1.9$$

Equations 1.6 and 1.7 only provide the estimation of the l_{mfp} in metal. The real condensed matter system may not follow the exact rules.

1.2.3 Ballistic transport

(1) Overview of ballistic transport

Ballistic transport is the charge transport where the charge carriers are not scattered. Quantitatively, ballistic transport appears when $l_{mfp} > L$. In ballistic transport, the charge carriers are usually electrons. The condensed matter system with ballistic transport is sometimes called the ballistic conductor [24,25]. Each conduction channel in a ballistic conductor contributes to a constant amount of conductance (G). This constant conductance (G_0) is $2e^2/h$, predicted by either the Landauer formula or the Boltzmann equation. When the magnetic field is not applied, a conduction channel contains two degenerated electrons with spin up and down. The total G can be calculated by summarizing all conduction channels, *i.e.*, $G = N_c G_0$, where N_c is the number of channels. Accordingly, the resistance per channel is $R_0 = 1/G_0 = h/2e^2 \approx 12.9 \text{ k}\Omega$ and the total $R = R_0/N_c$. Note that R of ballistic transport is the contact resistance that appears when the charge carriers are transferred between the contact and the condensed matter system. The condensed matter system itself exhibits no resistance because it does not scatter electrons.

l_{mfp} is a critical parameter to judge whether the charge transport in a condensed matter system is ballistic. Figure 1.7 summarizes the l_{mfp} of some condensed matter systems that are intensively studied recently. Note that two-dimensional electron gas (2DEG) is not a certain type of material. Instead, it is often referred to as the electrons that mainly conduct at the interfaces of heterostructures. These electrons usually exhibit extremely large μ and l_{mfp} [26]. Up until now, the resolution of the patterned condensed matter system has hit 10 nm using modern extreme ultraviolet lithography (EUV) [27]. Therefore, any condensed matter system whose l_{mfp} is larger than 10 nm can potentially be a system with ballistic transport.

Figure 1.7 Experimentally measured l_{mfp} of different condensed matter systems. TMD and 2DEG are abbreviations of transition metal dichalcogenide and two-dimensional electron gas.

Quantized conductance ($G = NG_0$) is the characteristic of ballistic transport. However, the transport measurement cannot capture this quantization easily. The reason is that the R , the physics quantity we measure, is inverse proportional to N ($R = 1/(NG_0)$). Only when N is small enough (usually less than 10), the change of N can induce a measurable change of R . One might wonder how to reduce the number of charge carriers in a ballistic conductor. The most widely used method nowadays is to limit the motion of charge carriers to 1D. This method was first proposed in the 2DEG at the GaAs/AlGaAs interface with $T < 1$ K [40,42]. Figure 1.8a shows the schematic configuration. A split gate is placed on top of the 2DEG condensed matter system. When a negative gate voltage is applied, a depletion region forms under the gate and forbids the electrons from passing through. Electrons are only allowed to penetrate between the gap of two gates. The gap is less than 200 nanometers wide, which is comparable to the Fermi wavelength λ_F (42 nm [4]) of the itinerant electrons. Therefore, the charge transport near the gap is quasi-1D. Only a digit number of electrons with a specific wavelength can pass through the gap, and the conductance becomes quantized (Figure 1.8b). This small gap is later termed quantum point contact. Similar quantizations via quantum point contact are also observed in the 2DEG condensed matter system, such as InAs/AlSb [43,44] and LaAlO₃/SrTiO₃ [41,45].

Apart from the quasi-1D transport in quantum point contact, natural 1D system, *e.g.*, metal wires [46–49] and carbon nanotube [50,51], and etched 1D system, *e.g.*, graphene [52,53] and 2DEG [54], also exhibit quantized conductance.

Figure 1.8. Conductance quantization. (a) Schematic of quantum point contact in 2DEG system. (b) The first reported conductance quantization, measured with a quantum point contact [40].

(2) Ballistic transport in a magnetic field

If the ballistic transport is confined in 1D, the conductance will be quantized with step G_0 ($2e^2/h$). The “2” in G_0 represents two degenerated states with opposite spin directions. When a magnetic field is applied, these two states will be split due to the Zeeman effect [55], demonstrated in Figure 1.9a. The electron in each split state contributes only $0.5G_0$ to the total conductance. Therefore, as the magnetic field increases, the step of the quantized conductance decreases from G_0 to $0.5G_0$. Figure 1.9b shows the $0.5G_0$ step of GaAs/AlGaAs at 80 mK [56]. Similar quantization has been reported in 2DEG systems [41,57–59] and 1D wires [60,61].

If the ballistic transport is not confined in 1D, charge carriers in magnetic flux density (B), exhibit a cyclotron motion with a cyclotron diameter (d_c) = $2mv_F/eB$ and cyclotron frequency (ω_c) = eB/m [62–64]. When a current is applied, the cyclotron motion bends the trajectory of the electrons along the current direction. A special magnetotransport phenomenon, magnetic focusing, occurs when electrons are guided to a specific position by the magnetic field. Magnetic focusing in a condensed matter system is achieved via a modified quantum point contact. Figure 1.9c shows the first configuration that reports the magnetic focusing in GaAs/AlGaAs [64]. It is a combination of two adjacent quantum point contacts used as electrons injector and detector. Two pairs of electrodes are

used in this configuration to apply current and measure ΔV , respectively. The more electrons the detector detects, the larger the measured signal. The injector and detector have 20 and 80 nm widths, respectively. The distance between the injector and detector (L_{id}) is 1.5 μm [64]. When an I and a magnetic field are applied, electrons pass through the injector and bend toward the detector side. Majorities of electrons will be focused into the detector when $L_{id} = K \cdot d_c$, where K is any positive integer. $L = K \cdot d_c$ can be rearranged into

$$\frac{K}{B} = \frac{Le}{2m v_F}. \quad 1.10$$

If we measure the field-dependent potential difference (ΔV - B), we can observe ΔV peaks with $1/B$ periodicity (Figure 1.9d) [64].

Figure 1.9. Ballistic transport in a magnetic field. (a) Degenerated states split by a magnetic field. The electron in each split state contributes half G_0 to the total conductance (b) Conductance of 2DEG quantum point contact as a function of gate voltage with B ranging from 0 to 8 T [56]. (c) Modified quantum point contact setup with four contacts. The left two contacts are used to apply a constant current, while the right two contacts are used to measure the potential difference. Usually, the distance between the bottom or top two contacts is at the micrometre level [64]. (d) The first theoretical calculated (top) and experimental observation (bottom) of magnetic focusing in 2DEG [64].

1.2.4 Diffusive transport

(1) Overview of diffusive transport

Diffusive transport is the charge transport when charge carriers are scattered ($l_{mfp} < L$). Different scattering centre affects the charge transport differently. Therefore, we should treat scattering specifically based on the type of scattering centres. In a condensed matter system, common scattering centres include impurities, dislocations, and phonons. These scattering centres affect the τ of the condensed matter system and thereby affects the μ via Equation 1.2, $\mu = e\tau/m$. Table 1 summarizes the μ and τ in different types of scattering. As shown in the table, μ is negatively correlated with T when charge carriers are scattering from phonons or dislocations. Therefore, if we cool a condensed matter system to its ground state, the phonon and dislocation scattering gradually decreases and finally disappears. In most metals, the dominating scattering is the phonon scattering. Therefore, we usually observe a positive correlation between R and T in metal.

When more than one type of scattering mechanism affects the charge transport, the total relaxation time could be estimated with

$$\frac{1}{\tau} = \sum_i \frac{1}{\tau_i} \quad [19], \quad 1.11$$

where τ_i is the relaxation time of each type of scattering. Longitudinal conductivity (σ_{xx}) can be calculated once μ is known via $\sigma_{xx} = en\mu$.

Scattering type	Relaxation time (τ)	Mobility (μ)	Character
Ionized impurity scattering [19]	$\frac{1}{\tau} = \frac{e^4 NL(2k\lambda_D)}{16\pi(2m^*)^{0.5}\epsilon_0^2\epsilon_s^2 E^{1.5}}$	$\mu = \frac{64 \sqrt{\pi\epsilon_0^2\epsilon_s^2(2k_B T)^{1.5}}}{N e^3 m^* \ln\left(\frac{12m^* k_B^2 T^2 \epsilon_0 \epsilon_s}{e^2 \hbar^2 n'}\right)}$	$\mu \propto T^{1.5}$
Neutral impurity scattering [19]	$\frac{1}{\tau} = \frac{10\epsilon_0\epsilon_s N \hbar^3}{\pi^2 m^{*2} q^2}$	$\mu = \frac{\pi^2 m^* e^3}{10\epsilon_0\epsilon_s N \hbar^3}$	μ is T independent
Acoustic phono scattering	$\frac{1}{\tau} = \frac{m_n^{*1.5} k_B T E_{c1}^2 (2E)^{0.5}}{\pi \hbar^4 c_1}$	$\mu = \left(\frac{2 \sqrt{2\pi} \hbar^4 c_1}{3m_n^{*1.5} m_c^* k_B^{1.5} E_{c1}^2}\right) T^{-1.5}$	$\mu \propto T^{-1.5}$
Piezoelectric Scattering [19]	$\tau = \frac{2^{1.5} \pi \hbar^2 \epsilon_0 \epsilon_s}{m^{*0.5} e^2 K^2 k_B T} E^{0.5}$	$\mu = \frac{16 \sqrt{2\pi} \hbar^2 \epsilon_0 \epsilon_s}{3m^{*1.5} e K^2 (k_B T)^2} E^{0.5}$	$\mu \propto T^{-0.5}$
Optical phonon scattering [65]	$\frac{1}{\tau} = \left(\frac{m_n^{*1.5}}{\tau_0}\right) W \theta_D T^{0.5} \left[\left(\epsilon_0 + 1\right) \left(\epsilon_0 - \frac{\theta_D}{T}\right)^{0.5} + \left(\epsilon_0 + \frac{\theta_D}{T}\right)^{0.5} \right]$	$\mu = \frac{8e\epsilon_0}{2\sqrt{2\pi} m k_B \theta_D^{0.5}} \left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0}\right)^{-1} \left(\frac{m_0}{m^*}\right)^{0.5} \chi(z_0) \frac{e^{z_0} - 1}{z_0^{0.5}}$	$\mu \propto T^{-1.5} \sim -2.5$
Dislocation scattering [19]	$\tau = \frac{3}{8N_d R v}$	$\mu = \left(\frac{3q}{8N_d R}\right) \frac{1}{(3m^* k_B T)^{0.5}}$	$\mu \propto T^{-0.5}$

Table 1. Summary of temperature-dependent μ and τ in different types of scattering. Some example cases are listed.

(2) Diffusive transport in a magnetic field

The most classical way to treat diffusive transport in a magnetic field is the Drude model [66]. It assumes charge carriers lose all $\mathbf{p}$ once they are scattered. Therefore, after time dt , the average momentum of the charge carriers would be $\mathbf{p} + d\mathbf{p}$, calculated with

$$\mathbf{p} + d\mathbf{p} = \left(1 - \frac{dt}{\tau}\right) (\mathbf{p} + \mathbf{F}dt) + \frac{dt}{\tau} \cdot \mathbf{0}. \quad 1.12$$

$\mathbf{F}$ and dt/τ are the force that acts on the carrier and the possibility that this carrier is scattered, respectively. $\mathbf{F}$ can be expressed as a sum of electric field force and Lorentz force, i.e., $\mathbf{F} = e(\mathbf{E} + \mathbf{v} \times \mathbf{B})$. Assume the condensed matter system is in a steady state, $d\mathbf{p}/dt = 0$, we get

$$\mathbf{E} = -\left(\mathbf{v} \times \mathbf{B} + \frac{\mathbf{p}}{e\tau}\right). \quad 1.13$$

By replacing $\mathbf{v}$ and $\mathbf{p}$ with the current density ($\mathbf{j}$) through the definitions $\mathbf{j} = -en\mathbf{v}$ and $\mathbf{p} = m\mathbf{v}$, we get

$$\mathbf{E} = \left(\frac{1}{ne}\mathbf{j} \times \mathbf{B} + \frac{m}{n\tau e^2}\mathbf{j}\right). \quad 1.14$$

Assume the carrier moves in the xy -plane and $\mathbf{B}$ is along the z -direction. Equation 1.14 can be rewritten in the following matrix form with the longitudinal (ρ_{xx}) and transverse (ρ_{xy}) resistivity

$$\mathbf{E} = \rho \mathbf{j} = \begin{pmatrix} \rho_{xx} & \rho_{yx} \\ \rho_{xy} & \rho_{yy} \end{pmatrix} \mathbf{j}. \quad 1.15$$

Therefore, ρ_{xx} and ρ_{xy} are given by

$$\rho_{xx} = \frac{m}{n\tau e^2}, \quad 1.16$$

$$\rho_{xy} = \frac{B}{ne}. \quad 1.17$$

The σ_{xx} and transverse conductivity (σ_{xy}) can be expressed as

$$\sigma_{xx} = \frac{\rho_{xx}}{\rho_{xx}^2 + \rho_{xy}^2} = \frac{n\tau e^2}{m} \frac{1}{1 + \omega_c^2 \tau^2}, \quad 1.18$$

$$\sigma_{xy} = \frac{\rho_{xy}}{\rho_{xx}^2 + \rho_{xy}^2} = \frac{n\tau e^2}{m} \frac{\omega_c^2 \tau}{1 + \omega_c^2 \tau^2}. \quad 1.19$$

The Drude model succeeds in explaining the linear relation between the transverse resistance (R_{xy}) and B [67]. However, it claims that R_{xx} is independent of B , which is inconsistent with most materials. The defect of the Drude model is that it assumes all charge carriers are from the same band. Therefore, all carriers have the same μ . However, there might be charge carriers from different bands contributing to the transport simultaneously. If we consider charge carriers from

two bands, the total conductivity is the sum of the conductivities of from each band, *i.e.*, $\sigma_{xx} = \sigma_{xx1} + \sigma_{xx2}$, $\sigma_{xy} = \sigma_{xy1} + \sigma_{xy2}$ [23]. The resistivity matrix should be modified as

$$\rho = \begin{pmatrix} \rho_{xx} & -\rho_{xy} \\ \rho_{xy} & \rho_{xx} \end{pmatrix} = \begin{pmatrix} \sigma_{xx1} + \sigma_{xx2} & -(\sigma_{xy1} + \sigma_{xy2}) \\ \sigma_{xy1} + \sigma_{xy2} & \sigma_{xx1} + \sigma_{xx2} \end{pmatrix}^{-1}. \quad 1.20$$

Plugging Equation 1.18 and 1.19 into 1.20, we obtain

$$\rho_{xx} = \frac{n_1\mu_1 + n_2\mu_2 + \mu_1\mu_2(n_1\mu_2 + n_2\mu_1)B^2}{e[(n_1\mu_1 + n_2\mu_2)^2 + (\mu_1\mu_2B)^2(n_2 + n_1)^2]}, \quad 1.21$$

$$\rho_{xy} = \frac{(n_1\mu_1^2 + n_2\mu_2^2) + (\mu_1\mu_2B)^2(n_2 + n_1)}{e[(n_1\mu_1 + n_2\mu_2)^2 + (\mu_1\mu_2B)^2(n_2 + n_1)^2]} B. \quad 1.22$$

Equation 1.21 shows that if we consider the contribution from more than one band, the R_{xx} could exhibit a quadratic relation with B , which is widely observed in semiconductors and metals [19,20,23]. Equation 1.22 shows that the R_{xy} is slightly nonlinear to B when two bands are considered. Note that, when we fit Equation 1.21 or Equation 1.22, n and μ of electrons should be negative while those of holes should be positive.

The Drude model after two-band correction is still imperfect because it is based on classical physics. More corrections are required in situations when the diffusive transport must be treated with quantum mechanics. These situations include, but are not limited to, a system with inhomogeneity (disorder), a system where the wave-like character of electrons can not be ignored, and a system with correlated degrees of freedom. Usually, only the correction to the R_{xx} is required. Correction to R_{xy} will be considered when the condensed matter system is magnetic. To eliminate the field-independent contribution, R_{xx} is often converted into magnetoresistance (MR) with via

$$\text{MR} = \frac{R_{xx}(B) - R_{xx}(B=0)}{R_{xx}(B=0)}. \quad 1.23$$

In some studies, $\Delta G = G(B) - G(B=0)$ is used instead of MR. Table 2 summarizes experimentally proved MR that disobey the B^2 relation and their corresponding mechanisms.

Mechanism	Description	Character
Electron-electron scattering [68,69]	$\text{MR} = R_0 \left(\frac{-e^2}{2\pi^2 \hbar} \right) [1 - (\omega_c \tau_0)^2] (1 - F) \ln \frac{T}{T_0}$	$\text{MR} \propto -B^2$
Inhomogeneous carrier density [70,71]	$\text{MR} = \frac{k N_i}{\pi n^2 e c} B$	$\text{MR} \propto B$
Weak localization [68,72]	$\Delta G = \frac{e^2}{\pi \hbar} \left\{ \psi \left(\frac{1}{2} + \frac{B_\phi}{B} \right) - \ln \frac{B_\phi}{B} \right\}$	Giant negative MR near zero fields.
Weak antilocalization [72-74]	$\Delta G = \frac{e^2}{\pi \hbar} \left\{ \psi \left(\frac{1}{2} + \frac{B_{so}}{B} \right) + \frac{1}{2} \psi \left(\frac{1}{2} + 2 \frac{B_{so}}{B} \right) - \ln \frac{B_{so}}{B} - \frac{1}{2} \ln \frac{2B_{so}}{B} \right\}$	Giant positive MR near zero fields.

Table 2. Summary of mechanisms of MR deviated from the quadratic rule.

(3) Magnetoresistance of a condensed matter system with spin-orbit interaction

Spin, orbit and charge are degrees of freedom when electrons move in free space. However, in a condensed matter system, these degrees of freedom could interact with each other and arouse emerging phenomena (Figure 1.10a). SOI is one of the most investigated interactions because it promises great potentials in spintronics applications [7]. Figure 1.10b demonstrates the SOI of an electron, with spin ($\mathbf{s}$), itinerating in an electric field. The orbit angular momentum of electron ($\mathbf{L}_e$) can be calculated from $\mathbf{p}$ and position vector ($\mathbf{r}$) via $\mathbf{L}_e = \mathbf{r} \times \mathbf{p}$. The electron dipole moment ($\boldsymbol{\mu}_s$) can be calculated with $\boldsymbol{\mu}_s = \mu_B \mathbf{s}$, where μ_B is the Bohr magneton. The relative motion between electron and electric field generates an effective magnetic flux density ($\mathbf{B}_{so}$) on the spins. According to relativity, $\mathbf{B}_{so}$ can be calculated with

$$\mathbf{B}_{so} = -\frac{1}{mc^2} \mathbf{p} \times \mathbf{E}. \quad 1.24$$

The energy correction induced by the SOI can be calculated with

$$\Delta \epsilon_{so} = -\boldsymbol{\mu}_s \cdot \mathbf{B}_{so} = \frac{e\mu_B}{2mc^2} \mathbf{s} \cdot (\mathbf{p} \times \mathbf{E}). \quad 1.25$$

Similar to the magnetic field, SOI also splits the degenerated states with spin up and down.

Figure 1.10. Interactions between degrees of freedom (a) and schematic of SOI (b).

According to Equation 1.24, $\mathbf{E}$ is critical for the existence of SOI. In a condensed matter system, $\mathbf{E}$ comes from the atomic nucleus and the asymmetric interface. In an atom, the electric field near the valence electrons is given by $\mathbf{E} =$

$\frac{1}{4\pi\epsilon_0} \frac{Q}{r^2}$, where Q is the effective charge of the nucleus after considering the influences of the inner electrons. Equation 1.25 could be rearranged into

$$\Delta\epsilon_{so} = \frac{e}{4\pi\epsilon_0 m^2 c^2} \frac{Q}{r^3} \mathbf{s} \cdot \mathbf{L}. \quad 1.26$$

Q , r , and L are atomic size dependent. Usually, a larger atomic size leads to a larger SOI.

Another intensively investigated case is the SOI of interfacial itinerant electrons induced by the electric field at the asymmetric interface [75–77]. This SOI was first proposed by Rashba and was later named Rashba SOI [78]. Figure 1.11a shows the schematic of Rashba SOI. Usually, the direction of the electric field is defined as z -direction, and the interface is defined as the xy -plane. $\mathbf{B}_{so}$, according to Equation 1.24, is always in the xy -plane and perpendicular to $\mathbf{p}$. When the SOI is sufficiently strong, $\mathbf{B}_{so}$ could be large enough to align the $\mathbf{s}$ along the direction of $\mathbf{B}_{so}$. The perpendicularly locking of $\mathbf{s}$ and $\mathbf{p}$ by strong SOI is called spin-momentum locking. With a constant direction of $\mathbf{E}$ throughout the heterostructure, Equation 1.25 can be rearranged into

$$\Delta\epsilon_{so} = v_0 \mathbf{z} \cdot (\mathbf{p} \times \mathbf{s}), \quad 1.27$$

where $\mathbf{z}$ is the unit vector in the z -direction, and v_0 is a coefficient. According to this equation, the energy splitting is determined by p and s and their relative directions. Figure 1.11b shows the side view of the ϵ - k diagram with Rashba SOI. Two states with the same $\mathbf{p}$ direction but opposite $\mathbf{s}$ directions or two states with the same $\mathbf{s}$ direction but opposite $\mathbf{p}$ directions exhibit different energy levels. However, two states with both $\mathbf{s}$ and $\mathbf{p}$ directions opposite exhibit the same energy level (Figure 1.11b top). Figure 1.11c shows the top view of the Fermi surface. For any given $\mathbf{k}_F$, Rashba SOI could force the $\mathbf{s}$ pointing perpendicular to $\mathbf{k}_F$, leading to a circular spin texture at the Fermi level.

Figure 1.11. Demonstration of Rashba SOI. (a) Schematic of interfacial electron affected by Rashba SOI. (b) Side view (top) and top view (bottom) of ϵ - k diagram after Rashba splitting.

Since the SOI does not couple charge directly, one might wonder how SOI affects charge transport? SOI affects charge transport by changing the phase coherence of the electrons. Classical scattering theory treats electrons as pure particles [79]. If we assume the diffusion of an electron is random and limited in 2D, the possibility (P) of finding an electron at a position $\mathbf{r}$ and time t follows

$$P(\mathbf{r}, t) = \frac{1}{4\pi Dt} e^{-\frac{r^2}{4Dt}}, \quad 1.28$$

where D is a diffusion coefficient (see Figure 1.12a black line [79]). Since the scattering is random, the electron could be backscattered and return its initial position ($\mathbf{r}_0 = 0$). Figure 1.12b shows one of the paths that the electron is backscattered and returns $\mathbf{r}_0$. According to Equation 1.28, the possibility to find an electron at time t and $\mathbf{r}_0$ is given by $P(r=0) = 1/(4\pi Dt)$. However, in quantum mechanics, electrons exhibit a wave-like character. With the same frequency and phase, two electrons propagate on the opposite path could constructively interfere with each other when they both return $\mathbf{r}_0$ (Figure 1.12c). The constructive interference doubles the possibility of finding an electron at $\mathbf{r}_0$ (Figure 1.12a, blue dash line), leading to the localization of electrons [79]. The localization, which arouses the constructive interference of backscattered electrons, is termed weak localization (WL). WL in the condensed matter system was initially proposed in 1980 [80]. However, electron localization aroused by constructive interference could date back to 1927, when the famous electron double slits experiment was conducted [81]. According to the definition, WL occurs when the condensed matter system exhibit diffusive transport and the electrons retain their phases

after scatterings. Mathematically, these two requirements can be expressed as $l_{mfp} < L$ and $l_{mfp} \ll l_\phi$, where l_ϕ is the average length of an electron that itinerates without a phase change. When a relatively strong SOI joins, the electron's spin becomes path-dependent (see Figure 1.12d). Two electrons will exhibit different s directions if they return r_0 along with the opposite directions. Together with the difference in s , the energy levels of these two electrons are split, leading to the decoherence of electron waves at r_0 . The possibility of finding an electron at r_0 is half the classical one (Figure 1.12a, red dot line) because of the destructive interference [79]. The delocalization of electrons, aroused by the destructive interference of backscattered electrons, is termed weak antilocalization (WAL). Since WAL is induced by SOI, measuring WAL has become a general method to probe the strength of SOI [82–86].

Figure 1.12. Schematic of WL and WAL (a) The possibility distribution of finding an electron at position r and time t [79]. The black line is the classic distribution with electrons considered as pure particles. The blue dash and red dot line are the possibility distributions near r_0 when WL and WAL join, respectively. (b-d) Schematic of electron diffusive scattering. The classic scattering (b), scattering with WL (c), and scattering with WAL. The grey arrows represent the electron paths. The black arrows represent the directions of s . The coloured circles represent the scattering centre.

To identify the WL/WAL, one needs to measure the OOP MR. Qualitatively, a magnetic field, which passes through the closed path of an electron, adds a phase shift to the electron wave [87]. The phase shift reverses the sign when the electron follows an opposite path. In a condensed matter system with strong WL, a magnetic field destroys the phase coherence of the two electrons with opposite paths, resulting in a negative MR (Figure 1.13a). In a condensed matter system with strong WAL, a magnetic field cancels the phase shift induced WAL, resulting in a positive MR (Figure 1.13b). Between these two limits, the MR of a condensed matter system could undergo a transition from positive to negative (Figure 1.13c). Figure 1.13d shows the ΔG of 9.2 nm Mg film covered with coverage of Au measured at 4.6 K [82]. In this system, Au/Mg interface is used to generating SOI. When the coverage of Au increases from 0% to 16%, we can observe a transition from positive ΔG to negative ΔG , indicating a transition from WL to WAL.

Figure 1.13. Magnetotransport of condensed matter system with WL or WAL. (a-c) Schematic MRs of a condensed matter system with WL (a), WAL (b), and WL + WAL (c). (d) ΔG of Mg film covered by Au film [82]. When the coverage of Au increases, SOI becomes more significant. Therefore, MR shifts from WL dominated to WAL dominated.

MR induced by WL and WAL may coexist in the same condensed matter system. Therefore, we cannot avoid comparing the strength of WL and WAL quantitatively. However, the physical quantity that characterizes the WL is l_ϕ while the quantity that characterizes the WAL is the B_{SO} . Directly compare two

quantities with different dimensions is impossible. Therefore, l_φ and B_{so} are usually switched to B_φ or l_{so} for comparison via [73]

$$B_i = \frac{\hbar}{4el_i^2} (i = \text{SO}, \varphi). \quad 1.29$$

When $B_{so} > B_\varphi$, we would observe an MR with more WAL contribution than WL contribution, and vice versa. Quantitatively, the correction to the magnetotransport induced by WAL (SOI) can be expressed as a product of G_0 ($2e^2/h$) and a function of B_{so} and B_φ ,

$$\Delta G = G_0 \cdot f(B_\varphi, B_{so}). \quad 1.30$$

Different models treat $f(B_\varphi, B_{so})$ differently [72–74,82]. The most widely used one is the Hikami-Larkin-Nagaoka (HLN) equation [73]

$$\Delta G = \frac{e^2}{\pi h} \left\{ \Psi \left(\frac{1}{2} + \frac{B_\varphi}{B} + \frac{B_{so}}{B} \right) + \frac{1}{2} \Psi \left(\frac{1}{2} + \frac{B_\varphi}{B} + 2 \frac{B_{so}}{B} \right) - \frac{1}{2} \Psi \left(\frac{1}{2} + \frac{B_\varphi}{B} \right) - \ln \frac{B_\varphi + B_{so}}{B} - \frac{1}{2} \ln \frac{B_\varphi + 2B_{so}}{B} + \frac{1}{2} \ln \frac{B_\varphi}{B} \right\}, \quad 1.31$$

where Ψ is the digamma function.

(4) Magnetotransport of a ferromagnet

Before we review the magnetotransport in a ferromagnet, basic concepts of magnetism should be elaborated. In a condensed matter system, apart from the itinerant electrons, there are local electrons attracted to the nucleus. The spin and orbit of all the local electrons around a nucleus contribute to a local angular momentum ($\mathbf{S}$). Each $\mathbf{S}$ exhibits a magnetic moment. All the local magnetic moments of a condensed matter system sum into the magnetization ($\mathbf{M}$). Local angular momentum is often called the local spin. Local spins could interact with each other. The total Hamiltonian of the interaction is given by

$$H = -\sum_{i \neq j} J \mathbf{S}_i \cdot \mathbf{S}_j, \quad 1.32$$

where J is the Heisenberg exchange coefficient. i and j in Equation 1.32 represent any two adjacent positions where $\mathbf{S} \neq \mathbf{0}$. The interaction is unchanged even when two local spins swap positions. Therefore, this interaction is also called exchange interaction. Thermodynamically, local spins prefer the orientations that lead to the lowest Hamiltonian. Therefore, for a ferromagnet ($J > 0$), local spins tend to be parallel. For an antiferromagnet ($J < 0$), adjacent spins prefer antiparallel orientations. If we take the magnetic anisotropy and magnetic field into consideration, the Hamiltonian would be

$$H = -\sum_{i \neq j} J \mathbf{S}_i \cdot \mathbf{S}_j - k_e \sum_i (\mathbf{S}_i \cdot \mathbf{e})^2 - \mu_B \sum_i \mathbf{B} \cdot \mathbf{S}_i, \quad 1.33$$

where k_e is a positive constant and $\mathbf{e}$ is a unit vector along the easy axis of the ferromagnet. The third term shows the contribution of the magnetic field, which tends to align the local spins along the field direction. The second term shows the contribution of magnetic anisotropy. It is called anisotropy because the local spins prefer the orientation of $\mathbf{e}$ or $-\mathbf{e}$. Figure 1.14 shows the simulated M - B of a ferromagnetic cube with Mumax3 [88]. Note that the parameters are made up to illustrate the influence of each contribution in Equation 1.33. The three orthogonal axes of the cube are defined as x, y, z , as shown in Figure 1.14a inset. The three components of M along x, y , and z are M_x, M_y , and M_z , respectively. The result is normalized by the saturation magnetization (M_s). M - B curves of a ferromagnet exhibit hysteresis, which means that M does not decrease to 0 when B decreases to 0. If we want to decrease M to zero, a non-zero field along the opposite direction needs to be applied. The absolute value of this extra field is the coercivity (B_c). Figure 1.14a and b compare the M - B hysteresis with $k_e = 0$ and varied $\mathbf{B}$ directions. Both curves are identical, indicating that ferromagnetism is isotropic. Figure 1.14a, c, and d demonstrate the influence from the easy axis (anisotropy). When $\mathbf{e} \parallel \mathbf{B}$ (Figure 1.14 c), M - B hysteresis along the $\mathbf{B}$ direction exhibits a steeper transition near B_c compared with those in Figure 1.14a, and vice versa (Figure 1.14d). Equation 1.32 can be further enriched by considering other contributions, such as the Dzyaloshinsky-Moriya (DM) interaction [89,90]. We will explain the details in Chapter 3.

Figure 1.14. Simulated normalized M_z hysteresis loop with different parameters. (a, b) normalized M_z - B hysteresis loop of an isotropic ferromagnet, with $\mathbf{B} // \mathbf{z}$ (a) and $\mathbf{B} // \mathbf{x}$ (b). (c, d) normalized M_z - B hysteresis loop of ferromagnets, with $\mathbf{e} // \mathbf{B} // \mathbf{z}$ (c), and $\mathbf{e} // \mathbf{z}$ while $\mathbf{B} // (0, 1, 1)$ (d).

In a ferromagnet without strong SOI, magnetic fields and local spins are two major influences on the itinerant spins. The interaction between itinerant and local spins is called the Kondo effect [91]. The Hamiltonian of the Kondo effect is given by

$$H = -J_K \sum_i \mathbf{S}_i \cdot \mathbf{s}_i, \quad 1.34$$

where J_k is a coefficient. $\mathbf{S}_i$ and $\mathbf{s}_i$ are local and itinerant spins, respectively. According to the Hamiltonian, the local spin tend to align the itinerant spins. In a condensed matter system with random local spins ($\mathbf{M} = \mathbf{0}$), influence from a magnetic field on the itinerant spins is partially negated by the Kondo effect. In a condensed matter system with parallel local spins and magnetic field ($\mathbf{M} // \mathbf{B}$), such as a saturated ferromagnet, both the Kondo effect and magnetic field tends to polarize the itinerant spins along the same direction. Therefore, the majority of the itinerant spins will be parallel.

In a condensed matter system, itinerant electrons will be scattered at the position where local spins are not zero. When the relative orientation of local and itinerant spins becomes larger, there will be a larger possibility that electrons are backscattered [92]. A larger backscattering possibility leads to a larger resistance of a condensed matter system. Figure 1.15a shows the schematics. Since the spins determine this scattering, this type of scattering is called spin-dependent scattering. The spin-dependent scattering of a ferromagnet will arouse a hysteresis of the R_{xx} . To illustrate the R_{xx} hysteresis, we assume that $\mathbf{B} \parallel z$ and consider the M_z only. In the region where M_z is saturated, all local spins and most itinerant spins are polarized. Since the spin-dependent scattering is unchanged in this field region, R_{xx} is a constant (Figure 1.15b, c). Local spins get randomized at $B = -B_c$ (Figure 1.15b) or $B = +B_c$ (Figure 1.15c). At these two field points, the backscattering possibility induced by the spin-dependent scattering is the largest, leading to a sharp R_{xx} peak. Once B crosses $-B_c$ (or $+B_c$) and M_z is saturated, R_{xx} drops back to constant.

Figure 1.15. R_{xx} hysteresis induced by spin-dependent scattering in a ferromagnet. (a) Schematic of spin-dependent scattering (b,c) R_{xx} - B and M_z - B with z -direction magnetic field sweeping from positive to negative (b) and from negative to positive (c)

Unlike the intuitive mechanism of R_{xx} in a ferromagnet, R_{xy} in a ferromagnet is much more complex. Hall discovered the ordinary R_{xy} which is linear to the magnetic field, in 1879 [67]. Two years later, Hall discovered that the R_{xy} of ferromagnetic Fe was several times greater than that of Au at the low magnetic field but deviated from the linearity when the field increases [93]. This phenomenon was later called the AHE. Both discoveries were astonishing at that time, given that “electron” was discovered almost 18 years afterwards [94].

Nowadays, the linearly B -dependent R_{xy} is called the Hall resistance or ordinary Hall resistance (R_{xy}^{OHE}). The strengthened R_{xy} in the ferromagnet is called the anomalous Hall resistance (R_{xy}^{AHE}) [21].

The underlying mechanism that bridges the AHE and ferromagnetism undergoes a half-century debate mainly between three mechanisms, proposed by J. Smit, L. Berger, and J. M. Luttinger [21]. Nowadays, all three mechanisms are considered valid and may coexist in a condensed matter system. The mechanisms proposed by Smit and Berger are later addressed as skew scattering [95,96] and side jump [97], respectively. The original theory proposed by Luttinger [98] is nowadays reworked with topology and becomes the third mechanism, the Berry phase [99,100]. Figure 1.16 shows the schematics of the mechanisms. All three mechanisms agree that AHE arises from the transverse biased motion of polarized itinerant spins. This biased motion switches direction once the itinerant spins are flipped. In a condensed matter system, when M_z changes, the majority of the itinerant spins are flipped accordingly, leading to the change of R_{xy}^{AHE} . An phenomenological relation between the R_{xy} , B , and M_z is

$$R_{xy} = R_{xy}^{OHE} + R_{xy}^{AHE} = R_H B + R_M M_z, \quad 1.35$$

where R_H and R_M are normal and anomalous Hall coefficients, respectively.

Equation 1.35 is agreed in the original papers of three mechanisms [95–98]. However, the three mechanisms differ in explaining the transverse biased motion. Figure 1.16a shows the schematic of the mechanism proposed by Smit [95,96]. When a scattering centre with SOI scatters itinerant electrons, a B_{so} acts on the electrons. Because of the relativistic nature of SOI, B_{so} will switch direction once the scattering direction is changed. If the spins of electrons are random, the possibilities of scattering to the left or right are the same. If the spins are polarized, the scattering possibility will be larger along the direction that leads to lower electron energy. This asymmetric scattering due to SOI is called skew scattering [95,96]. Berger followed the method of skew scattering but modelled electrons as wave packets instead of plane waves [97]. When the wave packets are scattered by the SOI centre, apart from the time-dependent transverse motion (skew scattering), there is also a constant transverse shift. This constant shift is called the side jump. Figure 1.16b shows the schematic. The side jump of an electron can be analogous to the dislocation of an object after the collision.

Nowadays, the AHE in ferromagnet is usually explained by skew scattering or side jump. The skew scattering mechanism predicts, without a magnetic field, $R_{xy} \propto R_{xx}$ while side jump mechanism predicts $R_{xy} \propto R_{xx}^2$. If a ferromagnet exhibits $R_{xy} \propto R_{xx}^a$, where $1 < a < 2$, both mechanisms contribute to the AHE. Apart from the mechanism based on scatterings, Luttinger proposes that the sum of the velocities transverse to the $\mathbf{E}$ -direction of all electrons in a ferromagnetic conductor was not zero even without any magnetic field applied [98]. Since his calculation is independent of scattering, this mechanism is referred to as the intrinsic mechanism. However, Luttinger failed to name the source of this transverse velocity. Until recently, the driving force for this intrinsic AHE is revealed to be the phase shift of electron when its $\mathbf{p}$ undergoes a closed path in the k -space. This phase shift is called the Berry phase. Nowadays, the mechanism based on the Berry phase is mainly used to explain the emerging AHE in non-ferromagnet [101–103].

Figure 1.16. Schematic of AHE mechanism. (a) Skew scattering (b) Side jump (c) Intrinsic mechanism. Both skew scattering and side jump attribute the AHE to the SOI of the scattering centre, while the intrinsic mechanism thinks the AHE is independent of scattering.

1.3 Experiment design

Figure 1.17 summarizes the commonly used techniques for investigating emerging phenomena. One of the most widely used techniques is interfacial engineering, which is to engineer the physical properties of a condensed matter system via the interface [104]. Generally, interfacial engineering is realized by build heterostructure. The interface region of the heterostructure inherits the properties from both layers and offers a playground for the interlayer interaction. For example, when a ferromagnet and a material with strong SOI form a heterostructure, the itinerant spins in the SOI layer could flip or even reorder the local spins in the ferromagnet [7]. Another advantage of building heterostructure

is that the interface region may exhibit a unique electronic structure. For example, the interface region between two insulators, *i.e.*, LaAlO_3 (LAO) and SrTiO_3 (STO), shows unexpected high conductivity [105]. Other techniques could further boost interfacial engineering. For example, the stacking of two atomically thin graphene flakes with a “magic” angle difference arouses an emerging superconducting [106].

Figure 1.17. Techniques used for materials' modifications [102,105,107–111].

Intrinsically, emerging transport phenomena arise from special electronic structures. Therefore, a condensed matter system with transition metal elements is an ideal candidate for investigation. Transition elements are those elements whose d-electrons have the largest energy. Compared with non-transition metal, d-electrons allow transition metals to exhibit properties such as ferromagnetism. When a transition metal is oxidized, it can lose multiple d-electrons, leading to its various oxidation states and electronic structures [75,110]. Figure 1.18 listed

the characteristics of transition metal oxide. Simple transition metal oxides contain only one type of metal element. These oxides do not have a strict stoichiometric ratio. Therefore, they exhibit a broad range of conductivity, from metallic to insulating. More importantly, when an oxygen ion bridges two transition metal ions, the d electrons of metal could interact with the p electron of oxygen and generate unique phenomena, such as double electron exchange. Complex transition metal oxides, contain at least two metal elements. The crystal structures of complex transition metal oxides, such as perovskite, split the degenerated d orbits and create more complex electronic structures. Both transition metals and their oxide exhibit appealing electrical and magnetic properties [75,110]. Therefore, to study the emerging magnetotransport phenomena, I plan to focus on the transition metals and their oxides-based heterostructure.

Figure 1.18. Major advantages of different condensed matter systems based on transition metal.

1.4 Organization of the thesis

This thesis is organized as follows:

In Chapter 1, I first broadly introduce modern electronics and spintronics. Then I give an overview of ballistic and diffusive transport in a condensed matter system. The diffusive transports of condensed matter systems with strong SOI and ferromagnetism are detailly elaborated. Finally, I conclude my research motivation and explain why I would choose heterostructures based on the transition metal and their oxide for my Ph.D. works.

In Chapter 2, I first detailly explain the methods used to conduct magnetotransport measurements. Then I elaborate the afterwards the methods used for post-experiment data processing.

In Chapter 3, I present the investigations on the skyrmion bubbles in the synthetic antiferromagnetic nanostructure. The evolution of spin textures is characterized via field-dependent transverse resistance (R_{xy} - B) and magnetic force microscopy (MFM) measurements. Then I achieved non-volatile multiple resistance states based on the skyrmion bubbles.

In Chapter 4, I present the investigations on the bipolar conduction (coexistence of electron and hole) in D-doped TiO_x . The mobility of electrons is 10 times larger after doping. Significantly, this bipolar conduction leads to a giant MR, 900% at 6 T.

In Chapter 5, I present the investigations on the ferromagnetism in $\text{LaMnO}_3/\text{SrIrO}_3$ (LMO/SIO) superlattices. The magnetic order shows a strong interface dependence. Through tuning the interfacial effect, we modulate the magnetization, easy axis, and AMR symmetry of the superlattices.

In Chapter 6, I summarize the projects and provide an outlook.

Chapter 2

Methodology

2.1 Magnetotransport measurements

Magnetotransport is the charge transport under a magnetic field. Properties of materials regarding multiple degrees of freedom, such as the carrier density, ferromagnetism, and SOI could be revealed through magnetotransport. This section introduces the experimental setup for a high-accuracy magnetotransport measurement.

2.1.1 Four-probe techniques

R_{xx} and R_{xy} should be measured with the four-probe technique. This technique requires two probes, denoted as I+ and I-, to pass the I and another two probes, denoted as V+ and V-, to measure the ΔV . Depending on the shape of the sample and electrode, the four probes should make contact with the sample or electrode following either Hall bar geometry or van der Pauw (VDP) geometry [112].

The four-probe technique with a Hall bar geometry is used when the sample or electrode is etched or deposited in a Hall bar shape. Figure 2.1a shows the schematics of the Hall-bar-shape electrodes and samples. The connections of the four probes for R_{xx} and R_{xy} measurement are marked in red and blue, respectively. R_{xx} and R_{xy} can be calculated with R_{xx} or $R_{xy} = \Delta V/I$.

Figure 2.1. Schematic connection of four-probe methods. (a) Hall bar geometry for cases when the sample or electrode has a Hall bar shape. (b) VDP geometry for a square film on the substrate.

Another widely used four-probe geometry is the VDP geometry, where four probes directly contact the corners of the sample deposited on the square

substrate. Figure 2.1b shows the schematic connections. To measure the R_{xy} of a film, I+ and I- are connected to the two ends of the film diagonal, while V+ and V- are connected across the other diagonal. R_{xy} is calculated with $R_{xy} = \Delta V/I$. To measure the sheet resistance (R_s) of a film, I+ and I- are connected across a side of the film while V+ and V- are connected on the opposite side. R_s is calculated with $R_s = (\pi/\ln^2)/(\Delta V/I)$. Chapter 2.2 explains how to convert R_s into R_{xx} .

Figure 2.2 shows the local current that flows along I+, I-, V+, and V- when we use VDP geometry to measure R_s . When all four probes are close enough to the corners, current passes from V+ to V- linearly. Therefore, ΔV is intrinsically the signal generated from a strip of the sample, framed with yellow dashes in Figure 2.2

Figure 2.2. Schematic of the current that passes through V+ and V- in a VDP geometry. The yellow dashes frame the area that generates the ΔV signal.

2.1.2 Real-time signal processing

The environmental influences add unwanted signals, such as noise and offset, during the measurements. Properly preprocessing the sample and the environment can reduce unwanted signals but cannot remove them [113]. Therefore, effective methods for real-time signal processing, which means process the signal during the measurement, are in demand. Here we model the signal with equation $\Delta V_{raw} = \Delta V + \Delta V_{noise} + \Delta V_{drift} + \Delta V_c$, where ΔV_{raw} , ΔV , ΔV_{noise} , ΔV_{drift} and ΔV_c represent the unprocessed signal, the desired potential difference, noises, drift, and constant offset, respectively.

ΔV_{noise} is an unknown function of time. However, we can average multiple measurements to reduce the ΔV_{noise} [113].

ΔV_c usually arises when the contact between the probe and sample is imperfect. It is independent of the current direction. Therefore, we can cancel the ΔV_c by applying a square wave with equal positive and negative current magnitude (Figure 2.3a) [113]. Figure 2.3b shows the ΔV_{raw} after the square wave is applied. The other influences are not included in Figure 2.3b to simplify the elaborations. R be calculated with

$$R = \frac{1}{2} \left(\frac{\Delta V + \Delta V_c}{-I} + \frac{-\Delta V + \Delta V_c}{-I} \right) = \frac{\Delta V}{I} \quad 2.1$$

ΔV_{drift} is usually induced by a gradually changing environment, such as the vibration of temperature. Similar to the ΔV_c , ΔV_{drift} is also invariant to the current direction. Therefore, we can implement a similar strategy used for ΔV_c cancellation [113]. Figure 2.3c shows the ΔV_{raw} composited with ΔV and ΔV_{drift} . When the square wave we applied has a sufficiently short period, the ΔV_{drift} is approximately linear to time. ΔV_{drift} within half the period will be a constant, dV . If we measure the ΔV_{raw} once when the current is switched to positive and measure the ΔV_{raw} twice when the current is switched to negative. R can be calculated by averaging the twice measurements at two positive current regions and twice measurements at negative current regions.

$$R = \frac{1}{4} \left(\frac{\Delta V}{I} + 2 \frac{-\Delta V + dV}{-I} + \frac{\Delta V + 3dV}{I} \right) = \frac{\Delta V}{I} \quad 2.2$$

Figure 2.3 Illustration of the real-time signal processing. (a) The applied current, a square wave with equal positive and negative current magnitude. (b) Signal

with ΔV and ΔV_c . (c) Signal with ΔV and ΔV_{drift} . dV is the ΔV_{drift} within half the period [113].

2.2 Conversion between resistance, sheet resistance, and resistivity

R , R_s and ρ are linked by the thickness (t), width (w) and length (l) of the conduction channel with equation

$$\rho = R_s \cdot t = R \frac{t \cdot w}{l} \quad 2.3$$

Usually, the t , w and l of the conduction channel are the same as those of the condensed matter system. However, in heterostructures with interfacial conduction, the t of the conduction channel is not equal to the sample thickness. Therefore, to compare the conductivity of these interfacial conduction samples, we should calculate the R_s instead of ρ .

2.3 A general method for fitting

Physical quantities, such as μ and B_{so} , cannot be directly measured. We can only obtain their values via fittings. Here I introduce the fitting method used in this thesis. We use a function with two parameters, denoted as a and b , as an example. The function can be expressed as $Y = f(a, b, X)$, where X and Y are independent and controlled variables, respectively. In a magnetotransport study, X is usually magnetic field strength or temperature, while Y is the conductivity or resistivity. Assume we obtain a series of X and Y via measurements, denoted as $(x_1, x_2 \dots x_n)$ and $(y_1, y_2 \dots y_n)$. The purpose of the fitting is to obtain the desired a and b . Assume the value of a and b are within a range $[a_1, a_k]$ and $[b_1, b_k]$. We can replace these continuous ranges with discrete vectors, *i.e.*, $(a_1, a_2, a_3 \dots a_k)$ and $(b_1, b_2, b_3 \dots b_k)$. The total number of possible a and b combinations is k^2 . We can calculate the expected Y values for each pair of a and b , (a_i, b_i) based on the $Y = f(a_i, b_i, X)$ and $(x_1, x_2 \dots x_n)$. The pair of a and b that provides the minimum standard deviation of the expected and measured Y values is the best fit.

2.4 Controlling instruments with LabVIEW

In this thesis, variable sample temperature and high magnetic fields are achieved by the superconducting magnet system, TeslatronPT [114]. Figure 2.4 shows the schematic of the dynamic temperature of TeslatronPT. TeslatronPT stabilizes the sample temperature via a dynamic equilibrium between heating and

cooling. Samples are placed on the sample probe, whose temperature sensor is located near the samples. The temperature of the sample probe is stabilized by heating from its heater and cooling from a variable temperature insert (VTI). The heat exchange between the sample probe and VTI is achieved by static gas. The temperature of VTI is stabilized by heating from its heater and cooling from a cold head. Unlike VTI, the cold head has a relatively constant temperature, 3.2 K. A cycling gas achieves the cold head and VTI heat exchange. Because we can control the pressure of the cycling gas, the temperature of VTI could be lower than 3.2 K.

Figure 2.4. Dynamic temperature equilibrium of TeslatronPT [114].

Currently, there is no open-source software for the control of TeslatronPT. Therefore, we need to build programs to control it based on the driver of TeslatronPT [115]. We choose LabVIEW [116] as the front-end and back-end programming language. Generally, the block diagram of a LabVIEW program can be separated into four steps, *i.e.*, instrument connection, instrument configuration, action, and connection close. Figure 2.5a, b shows the block diagram of the program for stabilizing and reading the sample temperature, respectively. Three parameters, *i.e.*, sample probe temperature, VTI temperature, and pressure of cycling helium, should be set during the instrument configuration

step. Note that to control the pressure of the cycling helium, the string command used for the “instrument connection” step should be “DBX:P1:TEMP” instead of “DBX:P1:PRES” in the TeslatronPT manual [114]. Because the pressure unit in our TeslotronPT is list No.3 among all the units. Therefore, we should use “DB3:P1:TEMP” command (Figure 2.5a).

Figure 2.5. Block diagram of the LabVIEW program for setting (a) and reading (b) the temperature of the sample probe. The yellow, green, blue and pink areas correspond to the instrument connection, instrument configuration, action, and connection close, respectively.

Similarly, two different programs should be used to set (Figure 2.6a) and read (Figure 2.6b) the strength of the magnetic field. Note that to set the strength of the magnetic field, we need a “Ramp to set” action during the “action” step (Figure 2.6a).

Figure 2.6. Block diagram of the LabVIEW program for setting (a) and reading (b) the magnitude of the magnetic field. The yellow, green, blue and pink areas correspond to the instrument connection, instrument configuration, action, and connection close, respectively.

In this thesis, R is measured with Keithley 6221 [113] and Keithley 2182A [113], which are used to generate current and measure the ΔV , respectively. Tektronix releases an official software for R measurement using Keithley 6221 and 2182A [117]. However, this software is not compatible with TeslatronPT. Therefore, we choose to build an R measurement program based on the driver of these instruments [118]. Figure 2.7 shows the block diagram of our program. The I- of the Keithley 6221 is not grounded by default. Therefore, we need to set it to the ground state manually during the “instrument configuration” step (Figure 2.7).

Importantly, during the “action” step, we may need to take measurements repetitively. Therefore, it is better to use the standard commands for programmable instruments (SCIP) than the instrument drivers. During the measurements, Keithley 2182A measures the ΔV and stores it into its internal buffer. After that, Keithley 6221 fetches the buffer from 2182A and generates program-readable data. Therefore, we need two commands to program the measurements. They are “:TRACe:POINts:ACTual?” and “:TRACe:DATA:SElected? %s, %d”. The first one is used to check the current number of ΔV stored in the buffer. Because new measurements are stored after old measurements in the buffer. Therefore, we need the second command to fetch new measurements.

Figure 2.7. Block diagram of the LabVIEW program for R measurement with Keithley 6221 and 2182A. The yellow, green, blue and pink areas correspond to the instrument connection, instrument configuration, action, and connection close, respectively.

Chapter 3

Evolution of Spin Textures in Transition Metal Nanostructures

3.1 Background and motivation

Generally, the spin texture of a condensed matter system can be determined by its spin-related Hamiltonian (Equation 1.31). The most widely observed spin textures include ferromagnetic spin texture (FST, Figure 3.1a), canted ferromagnetic spin texture (CFST, Figure 3.1b), and antiferromagnetic spin texture (AST, Figure 3.1c). Besides, skyrmion, originally proposed by T. H. R. Skyrme [119], is a unique spin texture that exhibits a gradient transition of the spin direction (Figure 3.1d). Conventionally, the stabilization of skyrmion requires DM interaction [89,90]. The Hamiltonian of DM interaction is given by

$$H = \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j), \quad 3.1$$

where $\mathbf{D}_{ij}$ is the DM coefficient. $\mathbf{S}_i$ and $\mathbf{S}_j$ are adjacent local spins. Experimentally, skyrmions have been observed in noncentrosymmetric B20 compounds [120,121], such as FeGe [122] and MnSi [123], or ferromagnet/heavy metal heterostructure [124–128]. Skyrmions stabilized by DM interaction have attracted much attention in spintronic research [129–131]. A recent trend in the skyrmion-based study is incorporating skyrmion into advanced artificial structures for both fundamental studies and spintronic devices, such as racetrack memory [132–134], logic devices [135,136], and skyrmion magnonic crystals [137,138]. Since these skyrmions could be stabilized by interfacial interaction without DMI, these skyrmions are often referred to as artificial skyrmions [139–145]. Moreover, when skyrmions have an extended center region where all the spins are aligned (Figure 3.1e). These skyrmions are sometimes named skyrmion bubbles [124,146,147].

Figure 3.1. Commonly observed spin textures: (a) FST, (b) CFST, (c) AST, (d) skyrmion, and (e) skyrmion bubble.

One of the most widely used artificial structures is the nano-patterned heterostructure [139–142,148,149]. These nanostructures contain nanodisks with artificial sizes, thicknesses, and shapes. Skyrmion bubbles, nucleating right under these nanodisks, obtain additional tunability from the varied structure. Moreover, skyrmion bubbles in the nanostructure could interplay with each other and form a crystal-like group, referred to as artificial skyrmion lattice [139,141] or skyrmion crystal [142,144,148]. The idea of integrating artificial crystal order into skyrmions expands the spintronic research frontier [137,138]. Meanwhile, to further enhance and exploit the functionalities of the skyrmions, introducing new spin textures into the skyrmion is desirable but elusive. Embedding antiferromagnetic interlayer exchange coupling through a nonmagnetic spacer [150] is a promising solution. These artificial antiferromagnets, which are referred to as synthetic antiferromagnets (SAFs) [147,151–153], introduce special spin textures, such as antiferromagnetic skyrmion [154] and antiferromagnetically-coupled bilayer skyrmion [147,153]. Therefore, a combination of SAFs and skyrmion crystals might permit new functionalities and boost the control of skyrmions. More importantly, via the skyrmion crystals, we could generate multiple magnetization states, which potentially contribute to future neuromorphic spintronic devices.

In this chapter, we incorporate SAFs into nanostructures, which contain the top nanodisks and the continuous bottom layer with varied geometries. The existence of the skyrmion bubbles is proved by MFM images. MFM images and R_{xy} - B hysteresis loops further reveal the spin textures of nanostructure in different

magnetic fields. A magnetic phase diagram of the nanostructure is obtained by analyzing the R_{xy} - B hysteresis loop at different temperatures. Moreover, by increasing the thickness of the bottom layer, skyrmion bubbles can be switched from evolving into an FST to an AST. In the nanostructures with multiple nanodisks, the skyrmions bubbles below these nanodisks evolve into AST asynchronously. Based on this unique feature, we achieved a non-volatile multi-state, characterized by multiple levels of R_{xy} .

3.2 Sample preparation and magnetization imaging

3.2.1 Sample preparation

Figure 3.2a shows the schematic of the major layers of our nanostructures, which are $[\text{Pt/Co}]_N/\text{Ru}/[\text{Co/Pt}]_4$ from bottom to top. The nominal thicknesses of Pt, Co, and Ru layers are 0.6, 0.6, and 0.9 nm, respectively. The Ru interlayer serves as a spacer that generates the antiferromagnetic interlayer exchange coupling between the bottom $[\text{Pt/Co}]_N$ and top $[\text{Pt/Co}]_4$ layers adjacent to the Ru [150]. Additional layers of 4 nm Ta/4 nm Pt and 4 nm Ta were deposited on the bottom and top of the major layers to prevent oxidation and structure asymmetry. Because these additional layers do not contribute to the magnetization, we omit them in the schematics. All layers were deposited with DC magnetron sputtering at room temperature with Ar pressure $\approx 2.3 \times 10^{-3}$ Torr. The deposition rate of Pt, Co, and Ru were 0.14, 0.21, and 0.1 Å/s, respectively. The thicknesses of these layers are estimated with deposition rate and deposition time. The top $[\text{Pt/Co}]_4$ layers, layers above Ru, were etched into circular nanodisks. The etching stops when the Ru element was detected. The nanodisks have varied diameters (D , = 200, 300 and 400 nm) and quantities (Q , = 0, 1 and 9). The samples with no nanodisks serve as reference samples. R_{xy} measurements are conducted on all samples, while MFM measurements are conducted on the nanostructures with $Q = 1$. The bottom layer of samples used for R_{xy} measurement are etched into a Hall bar cross and contacted with gold electrodes (Figure 3.2b). In the nanostructure with $Q = 9$, the spacing between these nanodisks is equal to the D (Figure 3.2c).

Figure 3.2. Demonstration of the nanostructure. (a) Schematic of the nanostructure with the top multilayer patterned into nanodisks. Note that these multilayers demonstrates the order of deposition. They do not imply that the interfaces in the samples are atomically sharp. (b) Optical image of the Hall bar cross used for R_{xy} measurement. (c) SEM top view of nanostructure with $Q = 9$. Note that in the revised

3.2.2 Imaging magnetization via magnetic force microscopy

Figure 3.3a,b shows the 300 K R_{xy} - B hysteresis loop of $Q = 1$ and $S = 400$ nm nanostructures with $N = 2$ and 4, respectively. Both hysteresis loops exhibit multiple stages. The MFM images, which are taken from $+B_s$ to $-B_s$, and its measurement field are placed below the R_{xy} - B hysteresis loop (Figure 3.3c-l). The schematic spin textures corresponding to MFM images are placed below the MFM images. MFM is a technique for revealing the relative magnetization direction of the nanodisk and the bottom layer through the relative brightness/darkness. It uses a tiny ferromagnetic tip scanning above the film surface. When the film exhibits ferromagnetism, the tip will be attracted or repulsed due to the dipolar interaction. The attraction/repulsion as a function of the film surface area can be converted into an image that represents the magnetization distribution of the film. The MFM used in this study is a home-built MFM with a 20 T superconducting magnet. The H_c and H_s of the MFM tip

are 0.025 T and 0.2 T, respectively. The tip is lifted around 100 nm to the sample surface.

In the regions where $B > +B_s$ or $B < -B_s$, all the spins in the nanodisks and bottom layer are parallel. The spin textures of the nanostructures at these regions are FSTs. For the MFM images taken at the FST regions, the nanodisks area is darker than the surroundings (Figure 3.3c, g, h, l).

If we sweep the magnetic field from $+B_s$ to 0 T, all the spins in nanodisks are flipped and become antiparallel to the bottom spins. After the top spins are flipped, the spin textures of the nanostructures become ASTs. For the MFM images taken at the AST regions, the nanodisks area is brighter than the surroundings (Figure 3.3f, k). Since spins in the nanodisks are flipped from upward-pointing to downward-pointing, R_{xy} exhibits a small drop according to Equation 1.34. Moreover, the existence of AST proves the existence of antiferromagnetic interlayer exchange interaction. In the nanostructure with $N = 4$, the field region where AST exists is much larger than that in the nanostructure with $N = 2$. Therefore, we can conclude that the antiferromagnetic interlayer exchange interaction is much larger in the nanostructure with $N = 4$ compared with that in the nanostructure with $N = 2$.

If we further sweep the magnetic field crossing the $-B_c$, we observe a huge drop of R_{xy} from a positive to a negative value. This huge R_{xy} drop can only appear when the majority of the bottom spins are flipped. Moreover, both the MFM images that are taken after (Figure 3.3e, j) and before crossing B_c (Figure 3.3f, k) exhibit brighter nanodisk areas. This indicates that the spins in the nanodisk are still antiparallel to the bottom spins that are covered by the nanodisks. Because the spins below the nanodisks are not flipped, R_{xy} does not drop to the minimum near $-B_c$ (Figure 3.3e, j). The flipped spins that arouse the huge R_{xy} drops are in the area uncovered by the nanodisk. However, not all the uncovered bottom spins are fully flipped from upward-pointing to downward-pointing. The intralayer ferromagnetic exchange interaction tends to align the bottom spins in the same direction. Therefore, the direction of the bottom spins would exhibit a gradient transition from upward to downward (see the schematics in Figure 3.3e, j) at the boundary between the covered and uncovered areas. The bottom spins at the boundary and the spins under the nanodisk form a special spin texture, a skyrmion

bubble. The spins at the boundary form the domain wall of the skyrmion bubble. B_c is the field where the skyrmion bubble nucleates.

If we further sweep the magnetic field towards $-B_s$, the R_{xy} and MFM image of nanostructures with $N = 2$ and $N = 4$ behave differently. For the nanostructure with $N = 2$, a further increased negative field will shrink the skyrmion bubble. When the skyrmion bubble is smaller than the nanodisk, the spins in the nanodisk would be parallel to the covered bottom spins outside the skyrmion bubble but antiparallel to the spins at the centre of the skyrmion bubble. Therefore, the nanodisk area of the MFM image exhibits a brighter core encircled by a darker area (Figure 3.3d). When the field reaches $-B_s$, the skyrmion bubble is annihilated and all spins are parallel. For the nanostructure with $N = 4$, the interlayer antiferromagnetic exchange interaction is strong enough to align the spins in the nanodisk and covered bottom spins antiparallel. Therefore, the skyrmion bubble cannot be smaller than the nanodisk in the nanostructure with $N = 4$. A further increased negative field will annihilate the skyrmion bubble and arouse an AST (Figure 3.3i). When the field reaches the $-B_s$ of the nanostructure with $N = 4$, the AST will be switched to FST (Figure 3.3h). Because the nanostructure with $N = 4$ exhibits an additional AST region, its B_s is larger than that of the nanostructure with $N = 2$.

Figure 3.3. R_{xy} - B hysteresis loop and measured MFM images of nanostructures with $N = 2$ (a) and $N = 4$ (b). (c-l) MFM images of different stages of the nanostructure.

3.3 Structure-dependent magnetization

A major advantage of nanostructure compared with ordinary heterostructure is that we have more tuning knobs over the shape of the samples. Figure 3.4a shows the 300 K R_{xy} - B hysteresis loops of nanostructures with $N = 2$, $Q = 1$, D ranging from 200 to 400 nm, and a heterostructure with $N = 2$, $Q = 0$. All three nanostructures exhibit a B_c similar to the B_c of the $Q = 0$ heterostructure. This similarity further proves that the B_c of our nanostructure represents the flipping of the uncovered bottom spins. Moreover, all three nanostructures exhibit similar B_s . According to the MFM images (Figure 3.3c-f), the skyrmion bubble exists in the field range between B_c and B_s in the nanostructure with $N = 2$. Therefore, similar B_s and B_c in all nanostructures prove that the skyrmion bubble generated via the SAF and nanostructure is robust against the size of the nanodisks. This also implies that we can further decrease the size of the nanodisk to achieve a smaller skyrmion bubble with relatively unchanged robustness.

The flipped bottom spins near B_c include the spins that are fully flipped by the magnetic field and the partially flipped spins due to the intralayer ferromagnetic exchange interaction. When the proportion of the partially flipped bottom spins increases, the intralayer ferromagnetic exchange interaction becomes significant, leading to the increase of B_c . Figure 3.4b compares 300 K R_{xy} - B hysteresis loops of nanostructures with $D = 400$ nm, varied N , and Q . When we increase Q from 1 to 9, the proportion of the partially flipped bottom spins near B_c becomes 9 times larger. Therefore, we observe a B_c increase in the $Q = 9$ nanostructure compared with that in the $Q = 1$ nanostructure.

Figure 3.4. R_{xy} - B hysteresis loop of nanostructures with different geometries. (a) R_{xy} - B hysteresis loop of nanostructures with $N = 2$ and varied S . (b) R_{xy} - B hysteresis loop of nanostructures with $D = 400$ nm, varied N and Q . R_{xy} - B hysteresis loop of two reference samples with no nanodisks, $N = 2$ and $N = 4$ are plotted in (a) and (b) respectively for comparison.

3.4 Magnetic phase diagram based on anomalous Hall effect

To fulfill the applications of spintronic devices in different situations, we investigate the temperature-dependent robustness of the skyrmion bubbles. Figure 3.5a,b shows the R_{xy} - B hysteresis loop at different temperatures of the nanostructures with $N = 2$, and $N = 4$, respectively. Because the R_{xy} - B hysteresis loop is symmetric, we only need to analyze the part where the field is swept from $+B_s$ to $-B_s$.

Figure 3.5. R_{xy} - B hysteresis loop of nanostructures with $D = 400$ nm, $N = 2$ (a) and $N = 4$ (b) at different temperatures.

Figure 3.6 shows the magnetic phase diagrams extracted from the descending branch of the R_{xy} - B hysteresis loops. When we cool the nanostructures from 300 to 2 K, the field range of the skyrmion bubble slightly increases. This indicates the skyrmions bubble is even more robust at low temperatures.

Figure 3.6. T - B phase diagram of SAF nanostructures with $D = 400$ nm, $N = 2$ (a) and $N = 4$ (b).

3.5 Irreversible magnetization and multi-state

Before we discuss the potential application of the skyrmion bubbles in the nanostructure, I would like to classify two concepts. The part of the R_{xy} - B hysteresis loop with the magnetic field swept from $+B_s$ to $-B_s$ is called the major curve. At any unsaturated field (B_R) on the major curve, we can reverse the sweeping from B_R back to $+B_s$. The curve between B_R and B_s is called the reversal curve. Figure 3.7 shows the major and reversal curves of nanostructures with different N and Q . In the nanostructure with $N = 2$, independent from Q , the reversal curves coincide with the major curve (Figure 3.7a,b). When $B_R \approx -B_s$, we would observe a small hysteresis near $-B_s$ (Figure 3.7a). However, this hysteresis disappears when the magnetic field is swept away from $-B_s$. In the nanostructure with $N = 4$, independent from Q , reversal and major curves never coincide (Figure 3.7c,d).

Figure 3.7. Fraction of R_{xy} - B hysteresis loop and reversible curve of nanostructures at 300 K with (a) $N = 2$, $D = 400$ nm and 1 nanodisk (b) $N = 2$, $D = 400$ nm and 9 nanodisks (c) $N = 4$, $D = 400$ nm and 1 nanodisk (d) $N = 4$, $D = 400$ nm and 9 nanodisks.

The unique irreversibility of the reversal curves in the nanostructure with $N = 4$ becomes more significant when the nanostructure is cooled to 2 K (Figure 3.8). This irreversibility allows us to design a multistate device at working at zero magnetic fields.

Figure 3.8. Part of R_{xy} - B hysteresis loop and reversible curve of SAF nanostructures with $N = 4$, $D = 400$ nm, and 9 nanodisks at different temperatures.

Figure 3.9a compares the descending branch of R_{xy} - B of the nanostructure with fixed $N = 4$, $D = 400$ nm, but a varied $Q = 1$ and 9. Both curves have three distinct regions. They are denoted as (i) stabilization, (ii) annihilation, and (iii) AST region in the following discussion. When the magnetic field goes more negative, R_{xy} exhibits a slow decrease (stabilization) and a steep drop (annihilation), which correspond to the shrink of the skyrmion bubble and the transition into AST, respectively. Figure 3.9a shows that the range of annihilation field of the $Q = 9$ nanostructure is much larger than that of the $Q = 1$ nanostructure, indicating that the nine skyrmions in the $Q = 9$ nanostructure annihilate asynchronously. Based on this phenomenon, we could purposely nucleate or annihilate a different number of skyrmions to achieve different electrical states in the device with multiple skyrmions. Figure 3.9b demonstrates the multi-state of R_{xy} achieved based on the asynchronous annihilation of skyrmions. We define the base state as the state where the bottom layer is antiferromagnetically coupled to the nanodisks and skyrmion is absent. The base state can be achieved by sweeping the magnetic field along the major curve and return to 0 kOe from the antiferromagnet region. States with a finite number of skyrmions can be achieved through returning from different fields in the annihilation region. Figure 3.9b demonstrates three representative states, with magnetic fields returning from -2.4, -2, and -1.6 kOe. More importantly, all three states exhibit non-volatile and robustness, concluded from their repeatable R_{xy} at zero fields.

Figure 3.9. Skyrmion-based non-volatile multi-states. (a) R_{xy} - H of nanostructure with a different number of nanodisks. With more nanodisks capped, the skyrmion annihilation region becomes larger. (b) Three different zero-field states were achieved by reversing from different fields. Each state is non-volatile and repeatable.

3.6 Chapter summary

In this chapter, we achieve a tunable evolution of skyrmion bubbles in SAFs via engineering the geometries of nanodisk and bottom layer in the nanostructure. The annihilation of the skyrmion bubble shifts from evolving into the FST to evolving into AST when the bottom layer thickness increases. Most significantly, by controlling the proportion of the annihilated skyrmion bubble in the skyrmion bubble array, we achieve non-volatile multiple resistance states. Our results demonstrate the feasibility of controlling the skyrmion bubble's evolution via nanopatterning, which is useful to spintronic applications, such as magnonic crystals and spin-based neuromorphic devices.

Chapter 4

Bipolar Conduction and Giant Positive Magnetoresistance in Doped Transition Metal Oxide Heterostructures

4.1 Background and motivation

Induced by coupling the charge with other degrees of freedom, *e.g.* spin, orbit and lattice, emerging electronic phenomena spur new research interests and contribute to the future application [155–157]. For example, the unexpected two-dimensional electron gas [105] and gate-tunable superconductivity [158] arouse a mania in discovering the novel electrical properties, which seems unachievable in the original system. Among various fertile grounds for appealing electromagnetic behaviours, one tantalizing platform is the metallic system, where the charge may induce phenomena, such as charge-spin conversion [159] and charge density wave [160]. However, even though charge could be coupled, tailoring the charge carrier in a metallic system, including its type, density, and mobility, is still challenging. The major obstacle is the high carrier density and mobility dominate the electrical transport and overwhelm any modulated electrical signals. Therefore, a successful modulation of carriers in a metallic system could reveal both the hidden physics and their implication in further electronics.

To achieve the tunability of carrier behaviour in a metallic system, both the dopant and material systems are crucial. Doping with ultra-small dopants, such as deuterium (D), is a possible solution based on its successful modulations in semiconductors [161,162]. Significantly, the applications of ultra-small dopants in the creation of metal-insulator transition [163,164] and high-temperature superconductivity [165,166] demonstrate the huge potential for future electronic applications by introducing strongly correlated functionalities. Apart from a proper dopant, a metallic system with a tunable electronic structure is preferred. One promising material is titanium oxide (TiO_x). Figure 4.1 shows the schematic crystal structure and resistivity of TiO_x and Ti at room temperature. TiO_x has tunable oxidation states with the titanium ion charged from 2+ to 4+ and wide resistivity from $10^{-5} \Omega\cdot\text{cm}$ to $10^{17} \Omega\cdot\text{cm}$ [167]. Moreover, TiO_x is earth-abundant [168] and has been widely used in important applications, such as

photovoltaics [169], photocatalysis [170], and ultraviolet absorption [171]. Particularly, in the rocksalt titanium monoxide (Ti^{2+}O), the d_{xy} , d_{xz} , and d_{yz} orbitals overlap and form the partially filled t_{2g} orbit, leading to a unique metallic behavior [110,172], different from the other semiconducting TiO_x families. Interestingly, TiO_x has a varied stoichiometric O/Ti ratio [173], which could induce hole conduction [174]. More importantly, if electron and hole conduction coexist in a TiO_x -based samples and are tunable, we can obtain an unsaturated GMR [175], which could be used as the magnetization states read head [7].

Figure 4.1 Schematic crystal structure and resistivity of TiO_x and Ti.

In this chapter, we took advantage of both TiO_x and ultra-small dopants to generate holes in the metallic TiO_xD_y . Structural, electronic, and stoichiometric analyses reveal that majority of the film maintains a rocksalt structure of Ti monoxide even after D doping. The electron mobility after a proper D doping significantly increases by an order of magnitude compared with the undoped one. Holes also exhibit comparably high mobility proved by fitting the nonlinear Hall effect with a two-channel model. Moreover, a giant positive magnetoresistance (MR) is observed and explained by the electron-hole compensation. Further investigation shows that the generation of holes and high carrier mobility can be tailored through controlling the D concentration.

4.2 Sample preparation and basic characterization

4.2.1 Sample preparation

Figure 4.2a shows the schematic of the sample fabrication and resulting sample structure. Incorporating ultra-small dopants into oxides is challenging [162,176], but into metals is relatively well-investigated [177–179]. For the sake of accurate quantification, D is better than H, because distinguishing between H dopant and H contaminants from the environment is challenging while distinguishing D from H is relatively easy [180]. Therefore, we fabricate the TiO_xD_y film with a two-step process, *i.e.* deuteration and oxidation. Experimentally, we sputtered 11 nm Ti films on SrTiO_3 (STO) substrates at 500 °C. The sputtering gas was a mixture of D_2 and Ar with four different D_2 :Ar ratios, namely 0:1, 0.1:1, 0.3:1, and 0.5:1. Then, in the oxidation process, we utilized both the lattice oxygen from the STO [181,182] activated during the 500 °C growth and environmental oxygen from the atmosphere after the growth.

4.2.2 Atomic structure and stoichiometry characterization

To evaluate the atomic structures of the TiO_xD_y film, we conducted scanning transmission electron microscopy (STEM). Figure 4.2b shows a cross-section STEM image of the TiO_xD_y /STO with a sputtering gas ratio of D_2 :Ar = 0.1:1. The total thickness of the film is ≈ 11 nm. The surface region (≈ 2 nm) and the interface region (≈ 1 nm) are amorphous. The majority of the film (≈ 8 nm) shows a rocksalt structure whose lattice parameter ≈ 4.18 Å, very close to that of the rocksalt TiO lattice parameter, 4.185 Å. Usually, TiO film could be epitaxially deposited on the substrate if their lattice mismatch is small, such as the $\text{TiO}[100](001)$ on $\text{MgO}[100](001)$ substrate [183] and $\text{TiO}[\bar{1}12](111)$ on $\text{Al}_2\text{O}_3[\bar{1}1\bar{2}0](0001)$ substrate [184]. However, in our work, TiO and STO have a $\approx 7\%$ lattice mismatch. This single crystallinity of TiO is unexpected. We hypothesize that the interfacial layer (≈ 1 nm), the transition layer between the film and substrate, releases the lattice mismatch, and stabilizes the rocksalt structure of TiO_xD_y . The rocksalt structure is also confirmed by X-ray diffraction (XRD) (see Figure 4.2c). The XRD peak of the film appears at 43° , which is very close to the theoretically predicted 43.5° of TiO [185]. The other peak at 46.5° is generated from the STO substrate.

We then estimate the D/Ti ratio in the film by Rutherford backscattering (RBS) and elastic recoil detection analysis (ERDA). The result is shown in Figure 4.2d. D is well introduced into the film even with the smallest D₂:Ar ratio = 0.1:1, and more D is doped when the D₂:Ar increases from 0.1:1 to as high as 10:1. The D concentration was measured with ERDA using the NEC 9SDH-2X3 MV pelletron tandem accelerator at Fudan University. The background pressure of the chamber during ion beam analysis was less than 1×10^{-4} Pa. A 19 μm mylar foil in front of the detector was used to absorb scattered helium. The ERDA spectra were converted into D concentration profiles by Alegria 1.0 code [186] with a systemic error below 7%. Additionally, the composition of the film was determined by RBS with a 3.5 MeV helium ions beam perpendicular to the surface of the sample. Ti concentration of the sample was given by RBS spectrum unfolding results analyzed with SIMNRA 6.03 code. Based on the RBS and ERDA measurements, we could obtain the atomic ratio of D/Ti distribution as a function of film depth. Each data point in Figure 4.2d is the average of the maximum and minimum of the D/Ti distribution. The error bar represents the maximum and minimum D/Ti values.

Figure 4.2. Atomic structure and chemical component characterization of $\text{TiO}_x\text{D}_y/\text{STO}$. (a) Schematic of $\text{TiO}_x\text{D}_y/\text{STO}$ fabrication process. (b) Atomic structure of $\text{TiO}_x\text{D}_y/\text{STO}$ ($\text{D}_2:\text{Ar} = 0.1:1$) cross-section characterized by STEM. The thickness of the film is around 11 nm, with an around 2 nm surface region and an around 1 nm interface region. (c) XRD of $\text{TiO}_x\text{D}_y/\text{STO}$ ($\text{D}_2:\text{Ar} = 0.1:1$), confirming the rocksalt structure of the film. (d) D/Ti ratio in TiO_xD_y film as a function of the sputtering gas ratio characterized by RBS and ERDA.

4.2.3 Oxidation state analysis

To investigate the Ti valence across the film, we carried out the electron energy loss spectroscopy (EELS) spectrum mapping at 17 different positions (marked in Figure. 4.3a) with an interval of 0.9 nm from the interface to the surface. Figure 4.3b,c shows the EELS results of Ti $L_{2,3}$ and O K edge, respectively. In the STO (spectra #1-4), Ti $L_{2,3}$ edges spectra show four peaks, a signature of Ti^{4+} . Correspondingly, a large O K edge pre-peak at 528 eV is also indicative of oxygen bonded to Ti^{4+} states. Moving into TiO_xD_y film (from #5 onwards), Ti $L_{2,3}$ edge spectra has only two peaks and a significant redshift, implying that Ti is in a much lower valence state ($\text{Ti}^{2+}/\text{Ti}^{3+}$) [187–189]. The

surface 2 nm region (#13-17) shows a slight blueshift, indicating a higher oxidation state. In comparison with the previous study [187] which offered a comprehensive list of Ti_xO_y compounds, we confirm that the mid-region of the film is predominantly Ti^{2+} , while the interfacial region and surface region are dominated by Ti^{3+} .

Based on the structure and component characterization, we can conclude that the D doped TiO, with some deviation in the O/Ti atomic ratio, constitutes the majority of the film.

Figure 4.3. Ti oxidation state characterization. (a) The high-angle annular dark-field (HAADF) image acquired simultaneously while taking the EELS spectrum imaging. The schematic image next to the HAADF image denotes the relative positions of 17 EELS spectra of (b) Ti $L_{2,3}$ and (c) O K edge.

4.3 Nonlinear Hall effect and bipolar conduction

Both the ρ_{xy} and ρ_{xx} in this work were measured via Keithley 6221 and Keithley 2182A with a VDP geometry. We first measured the temperature-dependent resistivity ($\rho_{xx}-T$) of the doped TiO_x . The result is shown in Figure 4.4. The negative correlation between ρ_{xx} and T from 2 to 300 K shows that this film is metallic. This metallic state, from the electrical point of view, proves that our film is dominated by TiO because previous works reported that only TiO is metallic while Ti_2O_3 and TiO_2 are semiconductors [110].

Figure 4.4. ρ_{xx} - T of the undoped TiO_x film.

To analysis on whether the D doping can tailor the carrier of the conductor, we measured the ρ_{xy} . Figure 4.5a-c shows the field-dependent ρ_{xy} (ρ_{xy} - B) of TiO_xD_y with $\text{D}_2:\text{Ar} = 0.1:1$ at different temperatures. ρ_{xy} is linear field-dependent from 300 to 30 K, which corresponds to pure electron conduction. Strikingly, below 30 K, the ρ_{xy} - B starts to exhibit a nonlinearity and reaches the maximum ρ_{xy} at 10 K. From 10 to 0.3 K, the nonlinearity gradually diminishes. To analyze the nonlinearity, we fit the ρ_{xy} - B with the two-band correlated by the Drude model [190–192] (Equation 1.22). In addition, during the fitting, n_1 , n_2 , μ_1 , and μ_2 are constrained by the zero-field ρ through the relation of $e(n_1\mu_1 + n_2\mu_2) = 1/\rho$. The fitting result is shown in Figure 4.5a-c as well for comparison. Figure 4.5d,e shows the fitted n and mobilities, respectively. Above 30 K, the TiO_xD_y film is pure electron-conducting. At 30 K, holes are generated which corresponds to the nonlinear Hall effect. Below 30 K, the hole density (n_h) dramatically increases and exceeds electron density (n_e) at 3 K. Explaining the nonlinear Hall effect as a coexistence of electron and hole is consistent with previous studies [193,194]. Another exciting result is the high mobility of both carriers. Figure 4.5e shows that the fitted mobilities are up to around $4 \times 10^3 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ for holes and $2 \times 10^4 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ for electrons at 10 K. Note that the amorphous surface and interface regions of $\text{TiO}_x\text{D}_y/\text{STO}$ are dominated by Ti^{3+} (Figure 4.3b), which are insulating at low temperature [110]. These high-mobility carriers must be from the mid-region. The mid-region rocksalt TiO_xD_y layer has an O/Ti ratio larger than 1 because its Ti valance state is slightly larger than 2+ (Figure 4.3b). This indicates that the rocksalt TiO_xD_y exhibits a cation vacancy. This vacancy arouses acceptor

defects and can potentially exhibit hole conduction [174]. Moreover, the n_e of the film at 300 K is close to the previous study on TiO [173]. Therefore, we conclude that the bipolar conduction in our film is the intrinsic properties of the rocksalt TiO_xD_y layer.

Figure 4.5. Nonlinear Hall effect of TiO_xD_y . ρ_{xy} of TiO_xD_y (D_2 : Ar = 0.1:1) from 300 to 30 K (a), 30 to 10 K (b), 10 to 0.3 K (c). Fitted n (d), and mobility (e) as a function of temperature. The coexistence of electron and hole below 30 K explains the nonlinearity of the ρ_{xy} below 30 K.

To date, there is no report of the bipolar conduction with high-mobility carriers in titanium oxide yet [110,182]. Therefore, we need to confirm the relation between the hole and D dopants. We first compare the ρ_{xx} - T of TiO_xD_y films with different D_2 :Ar sputtering gas ratios. Figure 4.6a shows that all TiO_xD_y films are metallic. This is consistent with the existing study on metallic TiO [110]. At any temperature below 300 K, the ρ_{xx} decreases when we increase the D_2 :Ar ratio. Quantitatively speaking, at both 300 and 1.5 K, the ρ drops around an order

of magnitude when the $D_2:Ar$ ratio increases from 0:1 to 0.5:1. This huge ρ_{xx} drop confirms that D is doped into the film and proves that D has a significant impact on the film conductivity. To map this D enhanced conductivity onto bipolar conduction. We then compare the $\rho_{xy}-B$ (Figure 4.6b) and fitted the n (Figure 4.6c, top) and μ (Figure 4.6c, bottom). We first observed a giant electron mobility increase. TiO_xD_y with $D_2:Ar = 0.1:1$ exhibits a high electron mobility, $2 \times 10^4 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ which is 10 times as large as the undoped one ($D_2:Ar = 0:1$). Moreover, TiO_xD_y with $D_2:Ar = 0.1:1$ has the greatest degree of nonlinearity and the largest n_h accordingly. Comparatively, the nonlinearity and the n_h of the TiO_xD_y with $D_2:Ar = 0.3:1$ is much smaller. The nonlinearity can be better demonstrated by comparing the dash lines (Figure 4.6b), the $\rho_{xy}-B$ low-field extrapolations, with the $\rho_{xy}-B$ curves. Despite the decreased n_h , hole mobility is relatively unchanged. When we further increase or decrease $D_2:Ar$ to 0.5:1 or 0:1, the $\rho_{xy}-B$ becomes completely linear. This indicates an optimal amount of D dopant is crucial for this nonlinearity and both excessive and insufficient D doping can inhibit the generation of holes.

Figure 4.6. Control experiment to investigate the requirement for high-mobility bipolar conduction. $\rho_{xx}-T$ (a) and $\rho_{xy}-B$ (b) of the TiO_xD_y with different sputtering gas ratios ($D_2:Ar$). The dash lines are the low-field extrapolations, which are plotted to demonstrate the nonlinearity. (c) Fitted n and mobility of TiO_xD_y at 10 K. The one with $D_2:Ar = 0.1:1$ yields the highest hole density.

4.4 Emerging giant magnetoresistance

Following the carrier investigation, we first measured the MR of TiO_xD_y with $D_2:Ar = 0.1:1$ at different temperatures (Figure 4.7). We found an unexpected giant MR, 915% at 6 T, and 10 K.

Figure 4.7. MR of TiO_xD_y ($\text{D}_2:\text{Ar} = 0.1:1$) at different temperatures.

Figure 4.8a compares the ρ - T at 0 and 6 T. Importantly, these two R_s - T curves coincide at the temperature region where the ρ_{xy} - B is linear ($T > 30$ K). The MR becomes significant only below 30 K, which is the exact transition temperature of ρ_{xy} - B from linear to nonlinear. Furthermore, neither doped STO [195] nor TiO_x [196] has been reported to exhibit such high MR. Therefore, the giant positive MR is an intrinsic behaviour of bipolar conduction. Giant unsaturated positive MR has been reported in some semimetal [175], which is naturally a bipolar conductor because its Fermi level crosses both the valance and conduction band. Differently, in our tailored TiO_xD_y , holes are manually generated and exhibit high mobility and n comparable to those of electrons. The electron and hole conduction channels can compensate each other and lead to an unsaturated large MR. This can be further proved by the comparison of the 10 K MR with different doping states (Figure 4.8b). When the $\text{D}_2:\text{Ar}$ ratio is changed to 0.3:1, MR at 6 T is accordingly reduced to around 250%. The pure electron conduction TiO_xD_y ($\text{D}_2:\text{Ar} = 0:1$ or $0.5:1$) has an MR less than 150% at 6 T, which is 6 times smaller than the one of TiO_xD_y with $\text{D}_2:\text{Ar} = 0.1:1$.

Figure 4.8. Giant positive MR of TiO_xD_y . (a) ρ - T with the out-of-plane field = 0 and 6 T. (b) ρ_{xy} - B of the TiO_xD_y with different sputtering gas ratios ($D_2:Ar$)

4.5 Chapter summary

In this chapter, we report an observation of bipolar conduction accompanied by a giant positive magnetoresistance in TiO_xD_y films. To overcome the challenges in intercalating the D into a crystalline oxide, a series of TiO_xD_y were formed by sequentially doping Ti with D and surface/interface oxidation. Intriguingly, while the electron mobility of the TiO_xD_y increases by an order of magnitude larger after doping, the emerging holes also exhibit high mobility. The high mobility is potentially related with the single crystal structure of cubic TiO_x . Moreover, the bipolar conduction induces a giant magnetoresistance up to 900% at 6 T, which is ~ 6 times higher than its conventional phase. Our study paves a way to empower conventional materials in existing electronics and induce novel electronic phases.

Chapter 5

Tunable Magnetism in Complex Transition Metal Oxide Superlattices with Spin-orbit Interaction

5.1 Background and motivation

In the pursuit of unexpected functionalities and high-performance spintronic devices, intensive efforts have been devoted to controlling the magnetism of magnetic materials. Among various possible controlling approaches, interfacial engineering promises accessibility to not only the magnetic modulation but also a strong interfacial coupling, arousing emerging phenomena and applications [77,104]. Recently, spurred by the unprecedented advances in complex oxide fabrication [197], the interface-based method now becomes feasible.

The perovskite is one of the complex oxides that could nurture strong interfacial effects. It has a cubic structure where cations (A) sit at the corners, oxygen atoms sit at the face centres, and transition metal atom (B) sits at the cubic centre (Figure 5.1a). The six oxygen atoms in a unit cell form an oxygen octahedron. Because the oxygen octahedron is not spherically symmetric, the five degenerated d orbits will be split into triply degenerated t_{2g} orbits and doubly degenerated e_g orbits (Figure 5.1b). In those perovskites whose oxygen octahedrons elongate or shorten, the degenerated e_g and t_{2g} are further split [198]. The rich electronic structure of transition atoms in perovskites leads to a large number of emerging phenomena. Here I introduce one of these phenomena, called the double exchange, that leads to ferromagnetism [199]. In a perovskite, if the electron configuration is reconstructed via phenomena, such as electron accumulation [200], one transition metal ion could obtain 1 more electron than its adjacent transition metal ion (Figure 5.1c). Because two adjacent transition metal ions are connected by O^{2-} , one electron in the 2p orbits of O^{2-} could hop to an empty e_g orbit while the electron in the e_g orbits could fill the 2p orbit [199] (Figure 5.1c). In the end, the local moments of the nearest transition metal ions always prefer the same directions, leading to ferromagnetism.

Figure 5.1. Schematic crystal and electronic structure of perovskite. (a) Face centre structure of perovskite. (b) d orbits split into t_{2g} and e_g orbits. (c) Schematic of double exchange mechanism.

The key to the design of a controllable magnetism through the interface is to introduce a spin-correlated interfacial coupling. One of the most tantalizing choices is the coupling between 3d and 5d electrons. The heterostructure of Mn (3d)/Ir (5d) complex oxides are particularly attractive because the entanglement of 3d and 5d electrons exhibit rich magnetic behaviours [108,200,201] and strong SOI [202–204]. For instance, emerging ferromagnetism and topological Hall effect have been reported in the $\text{SrMn}^{4+}\text{O}_3/\text{SrIr}^{4+}\text{O}_3$ (SMO/SIO) [205–207] and $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO)/SIO [208,209] heterostructure, respectively. Moreover, through atomic control of the coupling between the iridate and manganite, a tunable in-plane ferromagnetic anisotropy characterized by both magnetization and anisotropic magnetoresistance (AMR) was reported at the $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3/\text{SIO}$ [210,211] heterostructures. Up to now, while interesting results have been achieved at the $\text{Mn}^{4+}/\text{Ir}^{4+}$ or $\text{Mn}^{3+/4+}/\text{Ir}^{4+}$ interfaces, the little study explores the perovskite interfaces of pure Mn^{3+} and Ir^{4+} , which has a unique polar/nonpolar structure. With the $\text{Mn}^{3+}/\text{Ir}^{4+}$ interface providing an extra polar discontinuity at the interface [212], a stronger interfacial coupling is expected, possibly leading to a dramatic modulation of magnetism. This modulated magnetism could then be implemented in building magnetization-based devices.

In this chapter, through atomically changing the stacking of 3d/5d electrons, we achieved the magnetic-order modulation in $\text{LaMn}^{3+}\text{O}_3/\text{SIO}$ superlattices, which manifests as a change of magnetization, rotation of ferromagnetic easy axis, and transition of AMR symmetry. Both magnetization and magnetotransport measurements show that the magnetic order becomes more interface-dominated while we decrease the superlattice period. The comparison with the single-layer LMO and SIO and in-depth theoretical prediction demonstrate that the underlying mechanism for these behaviours is related to the Mn-Ir coupling and the interfacial Rashba effect.

5.2 Sample preparation and basic characterization

5.2.1 Sample preparation

Figure 5.2a shows the schematic structures of the superlattices $[(\text{SIO})_m-(\text{LMO})_m]_n$. Here m represents the number of the unit cell (uc) of SIO or LMO. n represents the repeats of $(\text{SIO})_m-(\text{LMO})_m$. We prepared four superlattices, namely $[(\text{SIO})_8-(\text{LMO})_8]_1$, $[(\text{SIO})_4-(\text{LMO})_4]_2$, $[(\text{SIO})_2-(\text{LMO})_2]_4$ and $[(\text{SIO})_1-(\text{LMO})_1]_8$ (denoted as SL881, SL442, SL224 and SL118 for short). The superlattices were epitaxially grown on (001)-orientated SrTiO_3 (STO) substrates by pulsed laser deposition at 700 °C and 100 mTorr oxygen pressure. The frequency of the pulsed laser was 2 Hz. The energy density of the laser was 1.8 J/cm^2 . During the cooling process of the samples, the oxygen pressure was increased to 200 mTorr. The 3d/5d interfacial effect is controlled via atomically changing the repeating period ($2m$). From SL881 to SL118, the period was shortened while the total thickness of each constituent was fixed. Thus, as the period is shortened, more interfaces are created, and a larger interfacial effect is expected. The high structural quality of the superlattice samples was confirmed by the *in-situ* reflection high-energy electron diffraction (RHEED) (Figure 5.2b). Every oscillation in Figure 5.2b represents the epitaxial growth of one unit cell (uc) LMO or SIO. The periodic oscillation of the four superlattice samples indicates a layer-by-layer growth mode.

Figure 5.2. In-situ sample deposition monitoring. (a) Schematic of the $[(\text{SIO})_m-(\text{LMO})_m]_n$ superlattices. (b) In-situ RHEED oscillations, confirming the structural quality of the superlattice samples

5.2.2 Crystal and electronic structure characterization

Figure 5.3a shows the XRD-diffraction of the four superlattices. Albeit our samples are very thin, the first-order superlattice satellite peaks can still be observed, demonstrating a good crystallinity. Since the electronic structure of Mn decides the underlying mechanism for the magnetism, we studied the electronic state of Mn through the Mn $L_{2,3}$ edges X-ray absorption spectroscopy (XAS) measured at 85 K (Figure 5.3b). XAS is a technique that measures the ratio of the absorbed X-ray intensity when the X-ray frequency is varied. Since the absorption depends on the electron energy level, XAS is widely used for detecting the electronic structures. The XAS of 2 and 8 uc LMO are also presented here for reference. Consistent with the previous report, the Mn^{2+} spectral peak is observed in our single-layer LMO due to the electron accumulation [200]. Same as the 8 uc LMO, the XAS of SL881 also shows an Mn^{3+} spectral peak together with a hump at the Mn^{2+} position. This agrees with our previous result that SL881 has relatively weak interfacial coupling and its electronic structure of Mn is mainly dominated by the LMO bulk part. However, with more Ir-Mn created, the Mn^{2+} peak disappears. A reasonable explanation for this would be the intrinsic electron accumulation in LMO is suppressed or counteracted. Because when the repeating period drops, the polar LMO layers will be continuously separated by nonpolar SIO and forming a nonpolar/polar/nonpolar structure. Therefore, there would be opposite polar discontinuities at the top and bottom of the LMO, and the electron accumulation

along the two directions will reduce each other. Since the electron accumulation is gradually modulated from SL881 to SL118, the underlying mechanism for the ferromagnetism also undergoes a transition from the intrinsic charge transfer inside LMO to the emerging electronic correlation at the 3d/5d interface.

Figure 5.3. Crystal and electronic structure characterization. (a) XRD of the $[(SIO)_m-(LMO)_m]_n$ superlattices. (b) XAS of the $[(SIO)_m-(LMO)_m]_n$ superlattices. XAS of 2 uc and 8 uc LMO are presented as references.

5.3 Magnetic characterization

In a magnetic condensed matter system, the magnetic order has a significant impact on the magnetotransport. Therefore, we studied the magnetism of the four superlattices in detail. Figure 5.4a shows the (100)-direction temperature-dependent magnetization measured with a superconducting quantum interference device (SQUID). SQUID is a widely used method for measuring the magnetism of a film. Different from the MFM technique, SQUID can only capture the neat magnetization of the whole film. However, SQUID is often integrated with a cooling system. Therefore, we could use SQUID to study temperature-dependent magnetization. The measurement is conducted during warming the samples with a 0.1 T magnetic field after cooling them in a 1 T magnetic field. The result shows a decreasing M_s of the superlattice when the repeating period is shorter. To be noted, when we replace the SIO with STO, the magnetization of $[(STO)_1-(LMO)_1]_8$ is becomes much smaller than the one of SL118 which indicates the interfacial effect is likely to be the interfacial coupling between Ir and Mn. Then, we investigate the magnetic order through the measurement of the in-plane field-dependent magnetization (M - B) at 10 K (Figure 5.4b). The hysteresis loop reveals the ferromagnetic nature of all four superlattices. More importantly, the disappearing hysteresis of the reference

sample $[(\text{STO})_1-(\text{LMO})_1]_8$ indicates the Ir plays a crucial role in the ferromagnetism. Quantitative comparison of saturation M_s and B_c , summarized in Figure 5.4c, demonstrates that both B_c and M_s can be tailored more than 100% from SL881 to SL118. This also shows the feasibility of tuning the magnetism through Ir-Mn interfacial engineering. To understand this behavior, we should first find out the source of the superlattice ferromagnetism. Previous work has reported Ir-moment is much weaker than Mn-moment [210]. Therefore, the magnetization measurement is dominated by Mn. LMO is an antiferromagnetic material [213] and will show ferromagnetism when it is in thin-film form through an intrinsic electron accumulation called “polar catastrophe” [108,200]. When the polar LMO epitaxial is grown on a nonpolar substrate like STO, there will be a potential build-up that can drive the electrons of LMO to accumulate at the bottom. When the thickness of the LMO becomes larger than 3 uc, this accumulation can initiate a phase transition of LMO from antiferromagnetism to ferromagnetism [200]. This theory well explains the absence of ferromagnetism in the reference sample $[(\text{STO})_1-(\text{LMO})_1]_8$ because the thickness of individual LMO is below 3 uc. However, with the same structure by simply replacing the Ti with Ir, SL118 shows ferromagnetism. Such a transition not only confirms the existence of the interfacial coupling between SIO and LMO but also proves the interfacial coupling is the source for the ferromagnetism in SL118.

Figure 5.4. Magnetization of the $[(\text{SIO})_m-(\text{LMO})_m]_n$ superlattices characterized by SQUID. (a) (100)-direction temperature-dependent magnetization measured 0.1 T magnetic field. (b) (100)-direction M - B measured at 10 K. A reference sample $[(\text{STO})_1-(\text{LMO})_1]_8$ is also presented to (c) The changing of B_c and M_s summarized from the M - B measurement.

Apart from the modulation of the magnetization, we also achieved the tilt of the ferromagnetic easy axis. Figure 5.5a, b shows the Mn L edge X-ray magnetic circular dichroism (XMCD) spectra [214]. XMCD signal is the difference between the left-circular polarized and right-circular polarized XAS signals. When the film is not ferromagnetic, these two XAS signals are identical and the XMCD signal is 0. Accordingly, when the XMCD signal is not 0, the film must be ferromagnetic. The XMCD in this study was measured at 85 K under 1 T magnetic field with 45° and out-of-plane (OOP) incident X-rays, respectively. The magnetic field applied was in the same direction as the X-ray. The out-of-plane signals are magnified 5 or 10 times (marked in Figure 5.5b) for comparison with the 45° -incident signals. Figure 5.5a shows that all four superlattices have almost the same moment along 45° direction. Since our SQUID result (Figure 5.4b) has shown that the in-plane magnetization is gradually decreasing when there are more interfaces. As a result, the spins of the four superlattices must be oriented differently, otherwise, it is impossible to have a similar net moment along the 45° direction. The OOP XMCD (Figure 5.5b) further validates this conclusion. There is no out-of-plane XMCD of SL881, indicating the spins are fully in the in-plane direction. When we shorten the superlattice period from SL881 to SL118, we observe an increasing out-of-plane XMCD signal indicating the ferromagnetic easy-axis tilts away from the in-plane.

Figure 5.5 Magnetization of $[(\text{SIO})_m-(\text{LMO})_m]_n$ superlattices characterized by XMCD at 85 K. The X-ray incident direction is (a) 45° and (b) 90° to the film plane. The 90° XMCD of SL118 is magnified 5 times while the 90° XMCD of SL224, SL442, and SL881 is magnified 10 times.

5.4 Characterizing interfacial transport by magnetoresistance

Figure 5.6 shows the temperature-dependent sheet resistance (R_s - T) of the four superlattices, single layer SIO and 8uc LMO. All superlattices exhibit a semiconducting behaviour with their R_s falling between the conducting 8 uc SIO and insulating LMO. Compared with the extremely insulating LMO and 2 uc SIO, the measurable R_s of the low-repeating-period superlattice indicates that there is an interfacial effect that could greatly modulate the transport. Moreover, SL881 has only one interface, therefore, the relatively weak interfacial effect does not impose a significant impact on its transport compared with the other superlattices.

Figure 5.6 R_s - T of the $[(\text{SIO})_m-(\text{LMO})_m]_n$ superlattices, single layer SIO, and 8 uc LMO.

We also investigate the interfacial coupling through magnetoresistance measurement. Figure 5.7a shows the (100)-direction in-plane (IP) MR measured at 30 K. SL881 and 8 uc SIO share a similar MR. This is expected because the interfacial effect of SL881 is relatively weak, and the conducting is dominated by the 8uc SIO. As the superlattice period is shortened, there is a marked increase of MR magnitude. Since the 2uc SIO or LMO are expected to be completely insulating at 30 K (Fig. 1d), their resistance will be independent of the magnetic field. Consequently, this marked MR increase is not a pure behaviour of SIO. Therefore, there should be an interfacial coupling between the SIO and LMO,

which influences the magnetotransport behaviour of the superlattice. The inset in Figure 5.7a shows the MR hysteresis loop, which is caused by ferromagnetism. Previous works have reported that Mn^{3+} is the dominating element in LMO/SIO superlattice that arouses ferromagnetism and single-layer LMO is insulating at 30 K. Therefore, the MR hysteresis in LMO/SIO superlattice shows there is charge transport in the interface region between SIO and LMO. We further confirm this conclusion by comparing the MR- B and M - B hysteresis loop of SL224 measured at 10 K with the magnetic field along (100)-direction (Figure 5.7b). Both loops show the same B_c . Based on the abrupt giant MR and MR hysteresis, we can conclude that when the superlattice period is shortened, the transport property of the superlattice shifts from bulk SIO dominating to the LMO/SIO interface dominating, where both LMO and SIO contribute.

Figure 5.7. IP MR characterization. (a) IP MR of $[(\text{SIO})_m-(\text{LMO})_m]_n$ superlattices and 8 uc SIO. (b) Comparison between MR- B and M - B hysteresis loop of SL224 at 10 K. The same B_c position indicates that LMO contributes to the conducting

5.5 Rashba spin-orbit interaction and weak antilocalization

We attribute the interfacial coupling to the Rashba SOI. Experimentally characterizing Rashba SOI is usually challenging. The most widely used method is to measure the OOP MR and fit the B_{so} via Equation 1.31. However, the result of the fitting is accurate only when the feature of WAL, a positive MR at low field, is significant. When a condensed matter system exhibits a strong WL, WAL may not be observed via MR measurement. Therefore, despite the potential strong SOI, obtaining the B_{so} via fitting the MR is impossible.

Figure 5.8a shows the OOP MR of $[(\text{SIO})_m-(\text{LMO})_m]_n$ superlattices. The MR of SL118 exhibits no WAL signal but a strong WL signal. Nevertheless, It

does not justify that its WAL is weaker than the WAL of other superlattices. The absence of positive MR at the low-field region only proves that the WL is much larger than its own WAL. A similar strong negative MR is also observed in SL442. Therefore, investigating the Rashba SOI via fitting the MR of $[(\text{SIO})_m-(\text{LMO})_m]_n$ superlattices is impossible. However, to determine whether the SIO/LMO interface yields a large Rashba SOI, we prepare another heterostructure, $(\text{SIO})_2-(\text{LMO})_2-(\text{SIO})_6$. It possesses more interfaces than 8 uc SIO and fewer interfaces than SL224. From 8 uc SIO to $(\text{SIO})_2-(\text{LMO})_2-(\text{SIO})_6$ to SL224, the thickness of SIO is fixed while more interface regions are created. The MRs of the three samples are shown in Figure 5.8b. 8 uc SIO exhibits neither strong WL nor WAL. Compared with 8 uc SIO, both $(\text{SIO})_2-(\text{LMO})_2-(\text{SIO})_6$ and SL224 exhibit much stronger WAL, which could only arise from the Rashba SOI at the SIO/LMO interface. Figure 5.8c,d shows the fitting of the ΔG of $(\text{SIO})_2-(\text{LMO})_2-(\text{SIO})_6$ and SL224 via Equation 1.31. While the WL is relatively unchanged in SL224 and $(\text{SIO})_2-(\text{LMO})_2-(\text{SIO})_6$, the Rashba SOI is stronger in SL224.

Figure 5.8. OOP MR characterization of SIO/LMO heterostructures. (a) OOP MR of $[(\text{SIO})_m-(\text{LMO})_m]_n$ superlattices (b) OOP MR of SL224, 8uc SIO and $(\text{SIO})_2-(\text{LMO})_2-(\text{SIO})_6$. (c, d) Fitting of the ΔG of SL224 (c) and $(\text{SIO})_2-(\text{LMO})_2-(\text{SIO})_6$ (d).

5.6 Anisotropic magnetoresistance

5.6.1 Phenomenological analysis

More information on the interfacial transport is revealed through investigating the anisotropy of the magnetotransport, we found the IP anisotropic magnetoresistance (AMR) symmetry could be tailored by changing the repeating period. The AMR was measured by rotating the in-plane 9 T magnetic field and calculated through

$$AMR(\theta) = \frac{\rho[B(\theta)] - \rho[B(\theta=0)]}{\rho[B(\theta=0)]}, \quad 5.1$$

where θ represents the angle between the magnetic field and the fixed current direction. Figure 5.9a shows the experimental IP AMR and its fitting by the phenomenological equation [215,216]

$$AMR(\theta) = A_0 + A_2 \sin[2(\theta - \theta_1)] + A_4 \sin[4(\theta - \theta_2)], \quad 5.2$$

where A_2 and A_4 denote the coefficient of the two-fold and four-fold symmetric terms, respectively. Note that the AMR of SL881 is magnified 10 times and still too noisy to be fitted. Figure 5.9b shows the fitting results, which are given in the form of A_4/A_2 . When the repeating period is shortened, the AMR gradually shifts from four-fold symmetry dominating ($A_4/A_2 > 1$) to two-fold symmetry dominating ($A_4/A_2 < 1$). The SL224, which is the intermediate state, has an AMR that lies in between these two limits.

Figure 5.9. IP AMR of the $[(\text{SIO})_m-(\text{LMO})_m]_n$ superlattices (b) In-plane anisotropic magnetoresistance (AMR) of four superlattices with the in-plane magnetic field rotating from 0° to 360° . The empty circles are the experimental results, and the solid lines represent the fitting curves. (c) The fitting results of AMR are presented in an A_4/A_2 form.

5.6.2 Mechanism exploration

We used the first-principles calculation to model the LMO/SIO superlattices with a periodic boundary condition. Figure 5.10a shows a representative schematic of an SL22. We obtain the Rashba SOI energy ($E_{\text{SOI_Rashba}}$) by excluding the intrinsic SOI energy ($E_{\text{SOI_intrinsic}}$) from the total SOI energy (E_{SOI}), *i.e.*, $E_{\text{SOI_Rashba}} = E_{\text{SOI}} - E_{\text{SOI_intrinsic}}$. By using the density functional theory (DFT) calculation embedded in the Vienna *ab initio* Simulation Package (VASP), we calculated the E_{SOI} of the superlattices, bulk LMO, and SIO. The intrinsic SOI originates from the atomic orbit and is independent of symmetry. Then, using the isolated LMO and SIO as the references, we obtain the $E_{\text{SOI_intrinsic}}$ of the superlattices as the average of the E_{SOI} of the bulk LMO and SIO, given in the form of $E_{\text{SOI_intrinsic}} = 0.5(E_{\text{SOI_LMO}} + E_{\text{SOI_SIO}})$. Figure 5.10b shows that the Rashba SOI is enlarged 5 times when the repeating period is shortened from SL44 to SL11. The enhancement of the Rashba effect is interesting and may come from various reasons, such as the asymmetric interfacial structure [83,210,217] and built-in electric field [86]. Based on our first-principles calculation results, we hypothesis the enhancement of the Rashba effect comes from the asymmetric interfacial structure. Our result shows that the bond length of interfacial O-Ir-O (Figure 5.10a) is monotonically decreasing from SL44 to SL11 (Figure 5.10b). This decreased bond length enlarges the overlaps between different orbitals and can result in a larger gradient of internal electric potential (ΔV), and therefore a larger Rashba SOI energy.

To investigate how the Rashba SOI affects the magnetotransport properties, we numerically simulate the AMR behaviour based on the first-principles results. We modelled the magnetotransport properties of the electrons at the LMO/SIO interface with a generalized Kondo lattice described by the Hamiltonian [218,219]

$$H = -t \sum_{\langle i,j \rangle, \sigma} c_{i\sigma}^\dagger c_{j\sigma} + i\lambda_R \sum_{\langle i,j \rangle, \sigma\sigma'} c_{i\sigma}^\dagger (\vec{\sigma} \times \hat{r}_{ij})_{z, \sigma\sigma'} c_{j\sigma'} - \mu \sum_i n_i - J_K \sum_i \mathbf{S}_i \cdot \mathbf{s}_i - B \sum_i \hat{\mathbf{n}} \cdot \mathbf{S}_i, \quad 5.3$$

where $c_{i\sigma}^\dagger$ ($c_{i\sigma}$) denotes the creation (annihilation) operator of an itinerant electron at site $\mathbf{r}_i$ with spin σ . The first term is the kinetic energy, involving nearest neighbour hopping t . The second term represents the Rashba spin-orbit coupling term arising from the heavy metal element Ir. μ is the chemical potential controlling the density of electrons. The next term is the Kondo interaction, which describes the coupling of the electron spin $\mathbf{s}_i = \sum_{\alpha\beta} c_{i\alpha}^\dagger \vec{\sigma}_{\alpha\beta} c_{i\beta}$ at each site $\mathbf{r}_i$ to

the localized moment $\mathbf{S}_i$ of the magnetic Mn ions. In the present model, the localized spins are treated as classical vectors of unit length ($|\mathbf{S}_i| = 1$). The final term is the magnetic field, applied in-plane, such that $\mathbf{B} = B\hat{n} = (B\cos\theta, B\sin\theta, 0)$.

When the repeating period is shortened, the J_K increases accordingly. The longitudinal conductivity, σ_{xx} , is calculated with the Kubo formula [220],

$$\sigma_{xx} = \frac{2\pi e^2}{Nh} \sum_{m,n \neq m} \left(\frac{f_m - f_n}{\varepsilon_m - \varepsilon_n} \right) \frac{\eta |\langle m | J_x | n \rangle|^2}{(\varepsilon_m - \varepsilon_n)^2 + \eta^2}, \quad 5.4$$

where the sum runs over all energy levels, N is the total number of sites, $f_{m(n)}$ is the Fermi-Dirac distribution function for single-particle eigenstates $|m\rangle$ and $|n\rangle$ with energy $\varepsilon_{m(n)}$, and η is the scattering rate of conduction electrons from the local moments. J_x is the current operator along $\hat{x}$ direction.

$$J_x = -i \sum_{j\sigma} [t(c_{j\sigma}^\dagger c_{j+\hat{x}\sigma} - H.c.) - i\lambda_R (c_{j\sigma}^\dagger (\vec{\sigma} \times \hat{r}_{j,j+\hat{x}})_{z,\sigma\sigma'} c_{j+\hat{x}\sigma'} - H.c.)] \quad 5.5$$

Since our magnetic field is IP, both ordinary Hall effect or AHE are negligible ($\sigma_{xy} \approx 0$). Therefore the longitudinal resistivity is computed as $\rho_{xx} = \frac{\sigma_{xx}}{\sigma_{xx}^2 + \sigma_{xy}^2} \approx 1/\sigma_{xx}$. Figure 5.10c shows the theoretically-predicted AMR behaviour.

When we enhance the SOI term λ , the dominating symmetry of the AMR changes from four-fold to two-fold, which is qualitatively in agreement with the experimental observation. This simulation result proves that the coupling between the spin-orbit coupled state of Ir^{4+} could impact the symmetry of the ferromagnetic order of Mn^{3+} when their interfacial coupling is prominent.

Figure 5.10. Theoretical investigation of LMO/SIO superlattices. (a) Schematic of the LMO/SIO superlattice structure used in the first-principle calculation. (b) Bond lengths and Rashba SOI energies were obtained by first-principles calculation. (c) Numerical simulation of anisotropic magnetoresistance (AMR) with different Rashba SOI strengths.

5.7 Chapter summary

In this chapter, through atomically stacking the 3d/5d electrons, we achieved a systematical modulation of magnetic order, including modification of magnetization, the tilt of the ferromagnetic easy axis, and symmetry transition of the anisotropic magnetoresistance. An interfacial coupling emerges and is enhanced by introducing more Mn (3d)/Ir (5d) interfaces into a unique polar-nonpolar LMO/SIO superlattice. In-depth first-principles calculation and numerical simulation show that these magnetic behaviours could be understood by the 3d/5d electron correlation and Rashba spin-orbit coupling. This atomic stacking of different electrons provides a new route to engineer magnetic properties synchronously and could contribute to future spintronic applications.

Chapter 6

Conclusion and Outlook

6.1 Conclusion

To contribute to the next-generation electronic and spintronic devices, systematic investigations should be devoted to magnetotransport studies.

Chapter 3 studies the evolution of spin textures in nanostructured Pt/Co multilayers. The nanostructure exhibits multiple spin textures, *i.e.*, skyrmion bubble, AST and FST. Moreover, we found that the skyrmion bubbles are annihilated into an AST when the bottom layer thickness increases. Significantly, by manually control the number of skyrmion bubbles that are annihilated, we achieved a robust multi-state.

Chapter 4 reveals emerging bipolar conduction in a D-doped TiO_x . The dopant, D, plays an important role in arousing this bipolar conduction. Both electrons and holes exhibit $\mu > 10^3 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$. Moreover, the electron mobility of the D-doped TiO_x is around 10 times as large as that of the undoped ones. Significantly, we observe a GMR, up to 900% at 6 T. This GMR intrinsically is induced by this high-mobility bipolar conduction.

Chapter 5 reveals emerging interfacial conduction in the SIO/LMO superlattice. The Rashab SOI increases when the period of the superlattice decreases. A Rashab SOI-induced WAL can be observed in certain samples. Moreover, we observe an emerging AMR symmetry transition between 2-fold and 4-fold when the superlattice changes. The underlying mechanism is also attributed to the increased Rashab SOI.

Chapters 3, 4 and 5 contain three studies that focus on the different aspects of magnetization-based multistate. The study in chapter 3 explores how to build multistate with different magnetizations. The study in chapter 4 explores how to build giant magnetoresistance which is used to read the multiple magnetization states. The study in chapter 5 explores how to control the magnetization via interfacial effect.

6.2 Outlook

6.2.1 Evolution of spin textures in transition metal nanostructures

Our work successfully demonstrates the robustness of our skyrmion bubbles and the feasibility of multistate based on the skyrmion bubbles. Based on the current results, there are two research directions that we can follow to push this research further.

To improve the functionality of the skyrmion-based multistate, we could investigate how compact the skyrmion bubbles could be while retaining the robust multistate. More compact skyrmion bubbles allow us to build smaller multistate devices without damaging their functionality. Besides, currently, each nanodisk could generate four bits, which is determined by the relative M_z orientations of the nanodisk and its bottom layers. If more Ru layers are inserted in the nanodisk, we could potentially obtain more states.

Another good route is to fabricate a nanostructure with periodically ordered nanodisks. Under these nanodisks, we could obtain periodically ordered skyrmion bubbles. These periodically ordered skyrmion bubbles and their capping nanodisks form a skyrmion-based magnonic crystal [137]. Magnonic crystal possesses a spin wave, which is analogous to the electron wave in a real crystal. When the skyrmion bubbles become the building blocks of magnonic crystals, we may observe emerging magnetic phenomena and discover new spintronic applications.

6.2.2 Bipolar conduction and giant positive magnetoresistance in doped transition metal oxide heterostructures

Our work reports the emerging bipolar conduction and GMR in D-doped TiO_x . In the future, we could investigate whether similar phenomena could be discovered in other transition metal oxides after D. Moreover, we find that the $\text{D}_2:\text{Ar} = 0.1:1$ gives birth to the largest hole concentration and MR. We could investigate the underlying mechanism that leads to the hole conduction from a theoretical point of view. We could design additional control experiments to demonstrate whether the $\text{D}_2:\text{Ar} = 0.1:1$ is the best ratio with the theoretical results.

6.2.3 Tunable magnetism in complex transition metal oxide superlattice with

spin-orbit interaction

Our work demonstrates the tilts of the ferromagnetic easy axis induced by the SOI of 5d transition metal oxide. We could investigate whether we could use this tilted easy axis to achieve field-free magnetic switching. Moreover, our 3d/5d superlattices exhibit a significant interfacial transport. We could use other methods shown in Figure 1.17, such as gating, to tune the properties of the carriers at the interface.

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Appendix. List of publications

First and co-first author publications

1. **K. Huang#**, L. Wu#, M. Wang, N. Swain, M. Motapothula, Y. Luo, K. Han, M. Chen, C. Ye, A. J. Yang, H. Xu, D. Qi, A. T. N'Diaye, C. Panagopoulos, D. Primetzhofer, L. Shen, P. Sengupta, J. Ma, Z. Feng, C.-W. Nan, and X. Renshaw Wang. Tailoring Magnetic Order via Atomically Stacking 3d/5d Electrons to Achieve High-Performance Spintronic Devices, **Applied Physics Reviews** **7**, 011401 (2020).

- Selected as a featured article by the editor

- # contribute equally

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