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Spin-Orbit Torque Generation and Manipulation in Epitaxial Complex Oxides

By

Xiaoxi Huang

A dissertation submitted in partial satisfaction of the
requirements for the degree of
Doctor of Philosophy
in
Materials Science and Engineering
in the
Graduate Division
of the
University of California, Berkeley

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Professor: Ramamoorthy Ramesh, Chair

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by

Xiaoxi Huang

Abstract

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Xiaoxi Huang

Doctor of Philosophy – Materials Science and Engineering

University of California, Berkeley

Professor Ramamoorthy Ramesh, Chair

This dissertation presents a systematic study on spin-orbit torque generation and manipulation in all oxide model systems consisting of ferromagnet $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$, SrRuO_3 , multiferroic BiFeO_3 and spin-orbit coupled SrIrO_3 . Efficient spin-orbit torque generation is crucial in the field of spintronics for the applications of spintronics-based memory and logic devices that requires the writing of magnetic states (i.e., switching the magnetization orientations). The development in efficient spin-orbit torque generation is staggered by the scarce of material candidates and the lack of extra degrees of freedom for spin current manipulation. The primary aim of this dissertation is to demonstrate how complex oxides can be utilized to generate spin currents efficiently and provide a viable pathway to control spin currents. In this dissertation, I will identify a complex oxide model system for spin current generation, with specific interest to investigate and understand the influence of electronic interaction enabled by the epitaxial interface on the spin-orbit torque generation. I will demonstrate that an epitaxial interface is able to facilitate spin current transmission and enable electronic interaction in the heterostructure; and the heterostructure interface, which is overlooked in the field of spintronics, is of vital importance for enhancing spin-orbit torque efficiency. Furthermore, the capability of BiFeO_3 of transmitting spin current via antiferromagnet magnons is demonstrated with various techniques. Two pathways-modifying interface termination and using electric fields to control the ferroelectric polarization in BiFeO_3 , which in turn changes the spin current transmission, are demonstrated. Strikingly, bistable high/low spin transmission states are observed when reversing the ferroelectric polarization of BiFeO_3 , suggesting a coupling between spin propagation and ferroelectric polarization. In summary, the work presented in this dissertation reveals the important role that epitaxial interface plays in spin-orbit torque generation and demonstrates that multiferroic BiFeO_3 is a good candidate for spin current transmission and manipulation. Additionally, the ferroelectric polarization controllable spin propagation in multiferroics offers an alternative to realize spintronics-based memory devices and the introduction of multiferroic materials to the field of spintronics opens up a new avenue for electrical control of spin currents.

To my parents for your unconditional love

To my grandparents for your sheltering

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Chapter 1

Introduction

This chapter provides a brief introduction to the technological background as well as the key concepts and definitions that are frequently referred to in this dissertation. To start with, the real-world problem of realizing next generation energy efficient memory and logic devices is presented at the very beginning to serve as a motivation for the work presented in this dissertation. Then a brief introduction to the physical mechanisms that are most commonly used for spin current generation is followed, providing the physical basis for resolving the aforementioned issues. Next a survey on the prevalent material systems that are studied for spin current generation in this field is provided, motivating materials innovation for spin current generation. What follows is an introduction to epitaxial complex oxides for spin current generation, detection and manipulation, which I propose could be the material candidates for next generation memory and logic devices. Finally, an outline that summarizes the organization of this thesis is given at the end of this chapter.

1.1 Towards next generation energy efficient memory and logic devices

Since the development of Complementary Metal-oxide-semiconductor (CMOS) technology in the 1980s, the evolution of human civilization has been deeply intertwined with the advances in semiconductor industry. The technological revolution in semiconductor industry that brought about advanced memory and logic devices such as microprocessor, random-access-memory (RAM) and et al has made devices with powerful computational capabilities accessible to people. Ever since, the demand for computational devices with fast processor, larger memory and smaller size has been ever-lasting, which serves as a driving force to integrate more and more transistors on a single chip. Empirically, according to Gordon Moore, the number of transistors in a dense integrated circuit doubles about every two years. This insatiable appetite for denser integrated circuit can only be met by shrinking the dimension of the transistors, which has miniaturized transistors to the sizes of 10 nanometers. The energy consumption and heat generation as a consequence of humongous transistor integration on chips are tremendous, and the electricity consumption associated with information and communication sector is estimated to account for 11% of the global electricity consumption in 2020 (Fig. 1.1a). This alarming fact triggers research on scalable spintronic memory and logic devices beyond CMOS that allow for progressive miniaturization and improve efficiency (reduce switching energy). Spintronics is a field that utilizes the spin and charge degrees of freedom to fulfill the requirements of controlling the properties of materials and devices for the next generation memory and logic applications such as spin transfer torque-magnetic random-access memory. Fig. 1.1b shows the performance (write energy vs time) for various spintronic memory cells. By amping up the efficiency for spin current generation, the write energy for spin torque-based memory cells can be optimized to be comparable to static random-access memory (SRAM) (Fig. 1.1b). Note that only spin transfer torque (STT) memories have been commercialized, while the rest are still at various stages of research and development. The performance of various types of logic circuits are summarized in Fig. 1.1c. Field-effect transistor (FET) is in general faster than spintronic logic, because charging the capacitors in the gate is faster than the precession of magnetization. Nevertheless, spintronic logic is non-volatile, while most of the FETs are volatile, and the recently proposed magneto-electric spin orbit (MESO) device[1] is predicted to reach extremely low operation energies (Fig. 1.1c). Given the potential of spintronics for achieving next-generation energy efficient memory and logic devices, intense studies have been devoted to this burgeoning field.

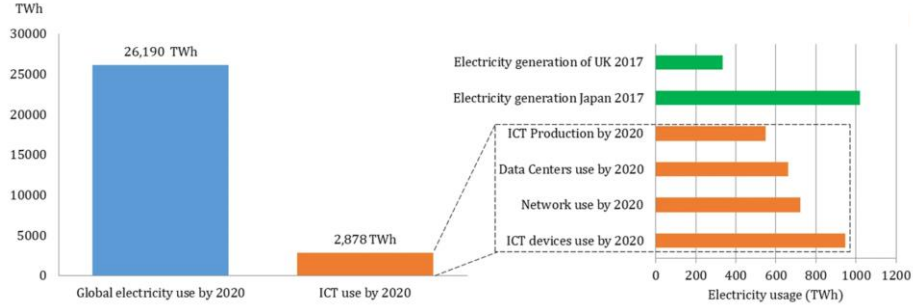
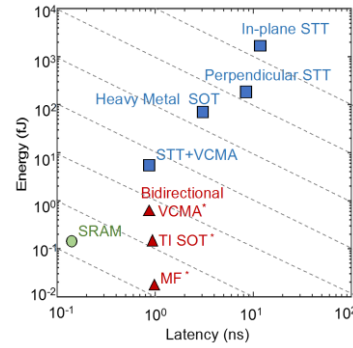
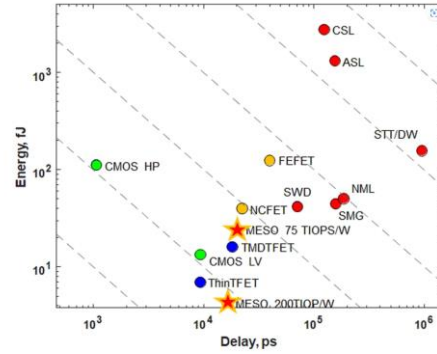
a**b****c**

Figure 1.1: Energy consumption for information and communication technology (ICT) and performance of next generation spintronic devices. **a**, Expected electricity consumption of the ICT sector by 2020. The electricity generation of the United Kingdom and Japan is listed as a comparison to the electricity consumed by ICT. **b**, STT: spin transfer torque, SOT: spin orbit torque, TI: topological insulator, VCMA: voltage controlled magnetic anisotropy. **c**, CMOS HP: high-performance transistors from the 2018 process generation (0.73 V supply), CMOS LV: low voltage (0.3 V) transistors, FEFET: ferroelectric field-effect transistors, NCFET: negative capacitance FET, STT/DW: spin-transfer-torque domain-wall device, ASL: all-spin logic, CSL: charge-spin logic, NML: nanomagnetic logic, SMG: spin majority gate, magnetoelectric, SWD: spin wave device, with magnetoelectric transduction, ThinTFET: 2D-material vertical tunnel FET, TMDTFET: transition-metal dichalcogenide tunnel FET, MESO: magneto-electric spin-orbit logic, two versions with different material parameters resulting in different computing throughput per dissipated power. Data from Ref. [1]. Figure 1 is adapted from Ref. [2].

The generation, detection and manipulation of spin currents have been the most important topics in the field of spintronics, and Rashba-Edelstein effect as well as spin Hall effect are the two best studied mechanisms for spin current generation.

1.2 Underlying mechanisms for spin current generation

1.2.1 Spin momentum locking and Rashba-Edelstein effect

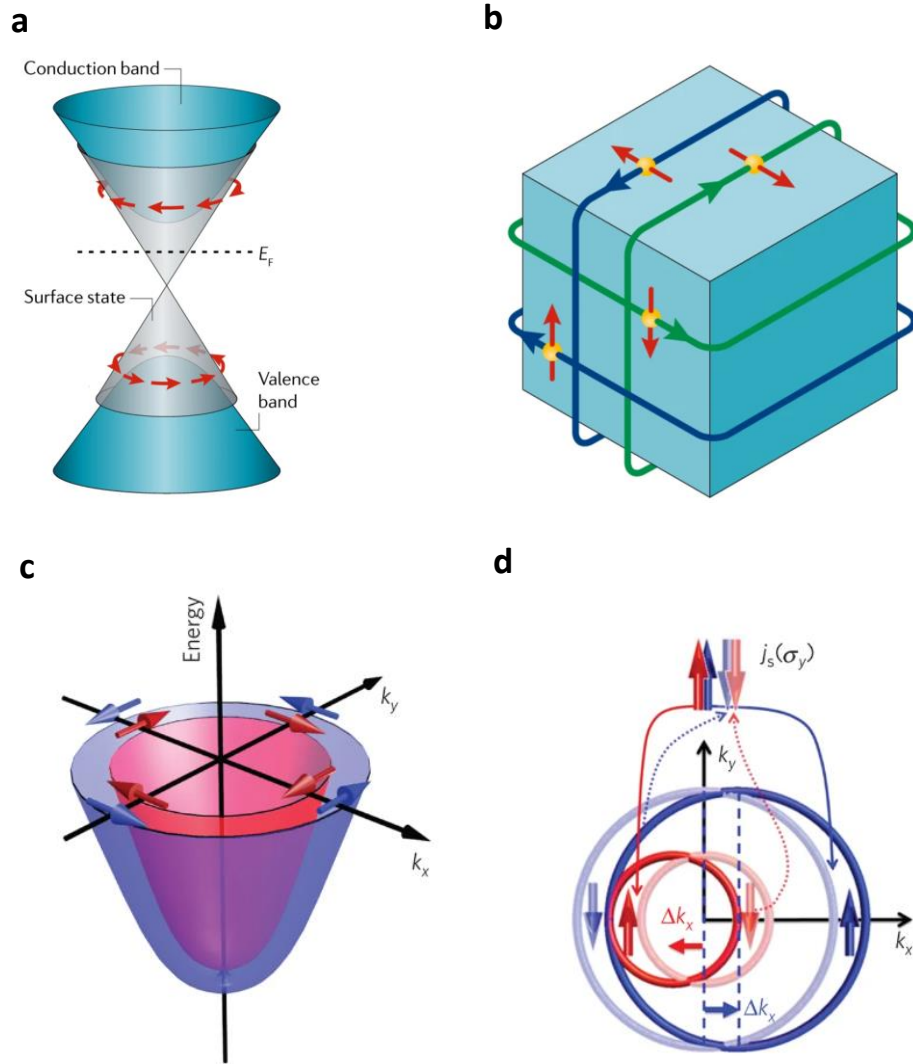


Figure 1.2: Spin current generation by spin momentum locking and Rashba effect. **a**, The massless Dirac-like dispersion of the surface state with spin momentum locking in a topological insulator. The surface state band connects the bulk valence and bulk conduction band. **b**, Real-space picture of the surface state in a topological insulator. Electrons with spins pointing up and down move in opposite directions. **c**, Sketch of a simplified Rashba type spin texture at equilibrium. At the Fermi level, there are two Fermi contours (inner contour is red and outer contour is blue) showing opposite spin

textures (red arrows and blue arrows for spin texture in the inner and outer Fermi contours respectively). **d**, The principle of inverse Edelstein effect, the injection of a spin current j_s with spins oriented along y direction (σ_y) generates a charge current along the x direction. **a** and **b** are adapted from Ref. [3], and **c** and **d** are adapted from Ref. [4].

Spin momentum locking is the constrain of spin orientation perpendicular to the electron momentum in topological insulators that exhibits insulating bulk state but metallic surface states populated by massless Dirac fermions[5-7]. If the Fermi level (E_F) lies within the bulk bandgap, the topological insulator shows metallic conduction at the surface but is insulating in the bulk (Fig. 1.2a). And the 2D surface state is described by the Hamiltonian

$$H = v_F(-\kappa_y\sigma_x + \kappa_x\sigma_y), (1.1)$$

where v_F is the Fermi velocity of the linear dispersion and σ_x and σ_y are the Pauli matrices for spin. This Hamiltonian implied spin momentum locking of the massless Dirac electrons, which means electrons with spin pointing up and down move in different directions (Fig. 1.2b). When a current flows in the surface state, more forward going electron states are occupied than backward going electron states, which necessarily means that the flow of charge is accompanied by a non-equilibrium surface spin accumulation at a right angle to the electron momentum because of spin momentum locking in the surface states.

Rashba effect is a manifestation of spin orbit coupling (SOC) in crystals, where the spin degeneracy associated with spatial inversion symmetry is lifted by inversion symmetry breaking induced electric field at the heterostructure interface[8, 9] or in the bulk of a crystal[10, 11]. The Rashba SOC (H_R) can be modeled by the interaction between spin Pauli matrices vector σ and momentum vector k , and can be expressed as the following Hamiltonian[8]:

$$H_R = \alpha_R(k \times \hat{z}) \cdot \sigma, (1.2)$$

where α_R is the Rashba coefficient and $\hat{z}$ is the unit vector along the direction where the inversion symmetry is broken. Owing to this interaction, the Rashba SOC generates a chiral spin angular momentum texture in the momentum space – spin momentum locking (SML) (Fig. 1.2c). To unravel the Rashba splitting and spin texture, a 2D free electron Hamiltonian with a Rashba SOC is introduced:

$$H = \frac{\kappa^2}{2m} + \alpha_R(k \times \hat{z}) \cdot \sigma, (1.3)$$

where $\kappa = \sqrt{\kappa_x^2 + \kappa_y^2}$. By solving the Schödinger equation $H|\psi\rangle = E|\psi\rangle$, one can get Dirac cone like energy splitting with original parabolic energy dispersion, $E_{\pm}(\kappa) = \frac{\kappa^2}{2m} \pm \alpha_R\kappa$ (Fig. 1.2c). The flow of a charge current results in the non-zero spin accumulation[12] coming from the uncompensated spin-textured Fermi surface (Fig. 1.2c). The converse effect, namely spin to charge conversion, is termed as inverse Edelstein effect (IRE) (Fig. 1.2d), which has been discovered at Ag/Bi interface[13].

1.2.2 Spin Hall effect

Spin Hall effect outstands several other possible mechanisms to generate spin currents as the most widely studied mechanism for spin current generation and manipulation. Spin Hall effect is – when a charge current passes through a non-magnetic material, a transverse spin imbalance will be generated with spin orientation perpendicular to the plane of the two currents (Fig. 1.3a). And the inverse process is the inverse spin Hall effect (ISHE) (Fig. 1.3b). This effect was first theoretically predicted in 1971 by Dyakonov and Perel[14] and later brought to the attention of the spintronics community by Hirsch[15] and Zhang[16]. Since the first experimental observation of spin Hall effect [17, 18] made two decades ago, everlasting efforts have been devoted to the search of materials for spin charge interconversion as well as current induced spin-orbit torques (SOT).

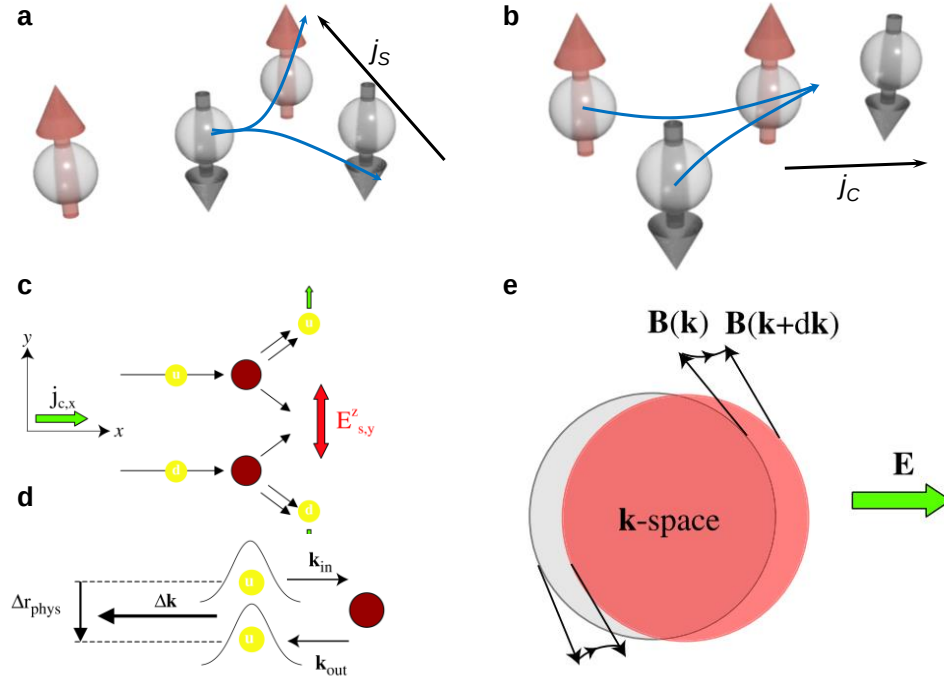


Figure 1.3: Spin Hall effect and the mechanisms. **a**, Spin Hall effect. **b**, Inverse spin Hall effect. **c**, Semiclassical picture for skew scattering. **d**, Semiclassical picture for side-jump. Δr_{phys} is the trajectory shift for the back scattered wave packet. **e**, Intrinsic spin Hall effect. **c**, **d**, **e** are adapted from Ref. [19].

There are two distinct extrinsic physical mechanisms for spin Hall effect, differing in the roles that external impurities play. The extrinsic mechanism is controlled by the spin orbit interaction with impurities, and two types of mechanisms have been identified: i, skew scattering and ii side-jump.

Skew scattering – It has been known that spin carrying particles are scattered asymmetrically by a central potential in the presence of spin orbit interaction. The orbital and spin angular momentum L_z , S_z are conserved separately in the case of spin orbit interaction with spherically symmetric impurities. The scattering cross section depends on

not only the scattering angle but also the relative signs of L_z and S_z . Therefore, a spin-up electron will be more strongly scattered, let's say, in the states of a positive L_z than in the states of a negative L_z , resulting in more electrons scattered to the left than to the right (Fig. 1.3c). And the opposite is true for electrons with down spins (Fig. 1.3c).

Side-jump – The side-jump originates from the anomalous form of the velocity operator in a system with spin orbit coupling[20-22]. Let us take the physical position operator for the conduction band of zincblende semiconductor for example[19].

$$r_{\text{phys}} = r + \frac{\lambda^2}{4\hbar} p \times \sigma, \quad (1.4)$$

where r_{phys} is the physical position operator in the positive energy space, r is the canonical position operator, p is momentum and σ is the vector for Pauli matrices. Taking the time derivative of r_{phys} , we get:

$$v = \frac{\hbar k}{m^*} + \frac{2\lambda_c^2}{\hbar} \nabla V_{\text{imp}}(r) \times \sigma, \quad (1.5)$$

Considering a semiclassical picture, an electron is described not by a plane wave but by a wave packet. Let $\hbar\Delta k$ be the average change of momentum of an electron in collision with an impurity. Obviously, this change takes place in an extremely short interval of time during which the electron collides with the impurity. Therefore, $\nabla V(r)$ dominates over the first term in the velocity equation and the first term can be disregarded. By integrating v over time, we get the displacement of the wave packet:

$$\Delta r_{\text{phys}} = \int v(t) dt = -2\lambda_c^2 \Delta k \times \sigma, \quad (1.6)$$

This is the side-jump effect (Fig. 1.3d).

Intrinsic mechanism – Besides the extrinsic (impurity related) mechanisms, the intrinsic mechanism, which is associated with the spin dependent band structure of the materials, can also contribute to the spin Hall effect. The essence of intrinsic spin Hall effect lies in the precession of spin about the k dependent magnetic field $B(k)$ that characterizes the band structure of the material. As qualitatively depicted in Fig. 1.3e, the Fermi distribution is displaced along the direction of the electric current (charge current). By changing the value of k , which in turn changes the value of $B(k)$, the electric field forces the spins out of alignment with the effective magnetic field $B(k)$. In order to realign with the effective magnetic field, the spins tilt away from its original position, and this tilting goes in opposite direction on the opposite side of the Fermi distribution. Thus, a transverse spin current is generated.

1.3 Material candidates for spin current generation

The theoretical work of Hirsch[15] and Zhang[16] on spin Hall effect triggered unprecedented interest in experimental discovery of spin Hall effect in the spintronics community[23]. The first experimental observations of spin Hall effect[17, 18, 24] were inspired by the intrinsic spin Hall effect proposal that focused on semiconductors and suggested that the optical activity of these materials be utilized for detecting spin Hall effect. Specifically, circularly polarized electroluminescence (suggested by Ref. [25]) and magneto optical Kerr effect (Ref. [25, 26]) were the two techniques that were commonly used in observing spin accumulation due to spin Hall effect. The spin distribution across

the semiconductor channel was imaged by Kato et al.[17] using magneto optical Kerr effect (Fig. 1.4a), showing that the spin polarization is out of the plane and has opposite signs on the two edges of the channel, consistent with the theoretical prediction of spin Hall effect. Wunderlich et al.[18], whereas, utilized coplanar p-n diodes to detect circularly polarized electroluminescence at the opposite edges of the 2D hole system(Fig. 1.4b, Ref. [24]). Their experiments showed that when an electric field is applied across the hole channel, a spin polarization with out of plane orientation was detected and has opposite signs for the two edges. Kato et al ascribed the observed signal to the extrinsic mechanism, based on a weak dependence on crystal orientation for the strained samples, while Wunderlich attributed their observation to the intrinsic mechanism. Remarkably, these two optical imaging of spin distribution evidenced the successful detection of spin Hall effect and inspired vigorous research on detecting and utilizing spin Hall effect electronically.

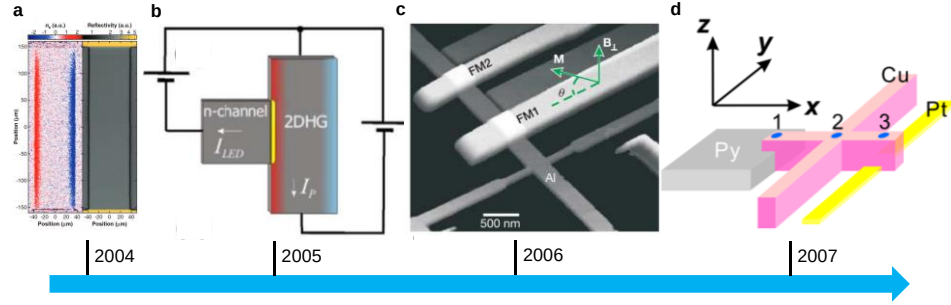


Figure 1.4: Milestones for the experimental observation of spin Hall effect. **a**, Two-dimensional image for spin density and reflectivity, respectively, for the unstrained GaAs sample at $T = 30$ K and $E = 30$ mV μm^{-1} . **b**, A 2DHG channel bordered by an n-type region that forms light-emitting-diode (LED). The application of a charge current I_P induces spin accumulation on the two edges. **c**, Atomic force microscope image of a device. A thin Al Hall cross is oxidized and contacted with two ferromagnetic electrodes with different widths (FM1 and FM2). A magnetic field perpendicular to the substrate, $B_{\perp}$, sets the orientation of the magnetization of FM1 (FM2), which is characterized by an angle, θ . **d**, A schematic illustration of the fabricated device for spin current detection. The figures are adapted from Ref. [17, 24, 27, 28].

After the realization of optical detection of spin accumulation generated by spin Hall effect, intense study was triggered, targeting at the electrical generation, manipulation, and detection of spin current. In 2006, Valenzuela et al.[27] successfully observed a charge imbalance converted from the injected spin current as a result of the spin Hall effect, opening a new arena for spin charge interconversion in the field of spintronics. In their experiments, the ferromagnetic electrode FM1 is used to inject spin-polarized current through the Al_2O_3 tunnel barrier into the Al channel (Fig. 1.4c). The injected current is driven away from the Hall cross, where only a pure spin current flows as a result of spin injection. The spin current induces a transverse charge imbalance, in the presence of spin Hall effect, and gets detected as a measurable voltage signal at the transverse probes. In parallel, Kimura et al. studied spin charge interconversion in a Py/Cu/Pt structure (Fig. 1.4d), in which Py and Pt serve as spin generator and detector interchangeably. Py was

first used as the spin current injector and the inverse spin Hall effect was measured in the Pt strip. Then the measurement geometry was reversed, where Pt with spin Hall effect served as the spin injector and the spin current was detected by measuring the non-local spin valve signal on the Py electrode. Spin Hall conductivities for both the direct and inverse spin Hall effect were determined to be the same, suggesting that spin Hall effect be a practical mechanism for spin current generation and detection. The achievement of electrical generation and detection of spin current was immediately followed by explosive search of material candidates for spin charge interconversion.

1.3.1 Heavy metals

Heavy metals – Despite the great potential that spin current has for encoding information, the study of spin Hall effect has only been considered as a scientific exploration out of curiosity since the highest efficiency for charge spin conversion was just a few percent[29, 30] at the early stages. However, the search for metals with large charge spin conversion efficiency (defined as spin Hall angle $\theta_{SH} = \frac{\hbar}{2e} \frac{J_S}{J_C}$) has since been a keen pursuit of researchers in the spintronics community. The best studied material in the following years, without any doubt, was Pt, where spin current sufficiently large to excite magnetization dynamics was achieved[31]. In their experiments, a pure spin current was injected into a Pt thin film by spin pumping, and it was observed to generate a transverse electromotive force that critically depends on the spin polarization direction (Fig. 1.5a). Systematic research was carried out later on to measure the intrinsic spin Hall angle of Pt accurately. M. Morota et al.[30] used a non-local spin valve structure to determine the intrinsic spin Hall effect of Pt to be around 0.021. Y. Wang et al.[32] took into account the spin diffusion in NiFe and measured the intrinsic spin Hall angle of Pt by systematically varying the thickness of NiFe. The intrinsic spin Hall angle was determined to be 0.068 at room temperature and no temperature dependence was found in their study. W. Zhang et al.[33], on the other hand, studied the role of Pt-Ferromagnet interface transparency in determining the intrinsic magnitude of spin Hall effect in Pt (Fig. 1.5b). By performing a comprehensive study on the dependence of spin Hall angle on different interfaces and the thickness of different ferromagnet, they found that the intrinsic spin Hall angle of Pt was as high as 0.19 ± 0.04 , after taking into account the interface transparency of these interfaces. These studies on determining the intrinsic spin Hall angle of Pt not only unraveled the spin Hall effect in Pt but also set the criteria that one should apply when interpreting the spin Hall angle of any materials measured by various techniques.

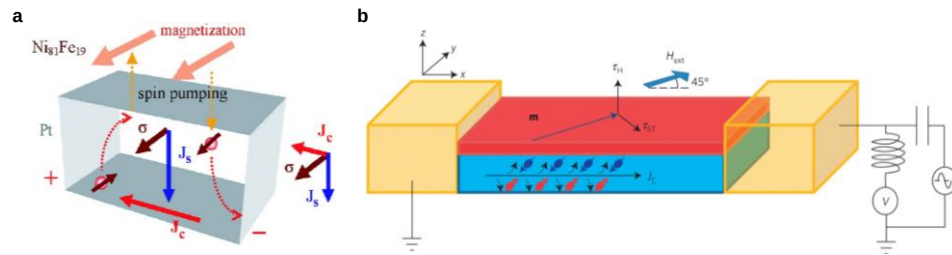


Figure 1.5: Spin current induced magnetization dynamics. a, Schematic illustration of spin pumping and inverse spin Hall effect. J_s and J_c denote the spin pumping generated pure spin current and the charge current generated by the inverse spin Hall effect, respectively. σ is the spin polarization direction of the spin current. The dotted arrows in Pt layer are the representation of the electron motion bent by the spin orbit interaction, responsible for the inverse spin Hall effect. Adapted from Ref. [31]. b, Illustration of spin torque ferromagnetic resonance (ST-FMR) measurement geometry. H_{ext} is the external magnetic field, m is the magnetic moment of the ferromagnet, τ_H is the field like torque generated by Oersted field due to the charge current in the Pt layer, and τ_{ST} is the spin torque due to the spin current generated by Pt. Adapted from Ref.[33].

An important advantage associated with the large spin Hall angle in metals is that the spin orbit torque in these metals such as Pt and Ta are sufficiently large to switch the magnetization of the adjacent ferromagnet. Liu et al.[34] demonstrated a prototypical device (Fig. 1.6a), where a magnetic tunnel junction (MTJ) consisting of CoFeB (3.8 nm)/MgO (1.6 nm)/CoFeB (1.6 nm) is integrated on top of the Ta channel. The spin current generated in the Ta channel transfers its spin angular momentum to the adjacent ferromagnet (free layer in the MTJ structure) and reorients the magnetization of the free layer with large enough spin currents. The parallel and anti-parallel configuration of the magnetization of the two magnetic layers can be consistently achieved (low and high MTJ resistance, Fig. 1.6b), indicating that heavy metals can be potentially used as the spin current source for MTJ based memory and logic devices. Compared to spin transfer torque induced magnetization switching, where spin angular momentum is carried by an electric current that is spin-polarized by the fixed layer of the MTJ, there are several advantages of the devices that utilize spin Hall effect to generate spin currents. First of all, the separation of the low impedance writing channel (Ta) from the high impedance reading channel (MTJ cell), as demonstrated by the three-terminal device in Fig. 1.6a, mitigates the deterioration of the insulating layer in the tunnel junction by large writing currents with repeated cycles, and allows for optimization of the tunnel junction for large read-out signal. In addition, identical currents required for switching magnetization back and forth (from parallel to anti-parallel and the reversing process) is another advantage associated with the three-terminal device, due to the inherent symmetry of spin Hall effect. Last but not the least, the proposed three-terminal device based on spin Hall effect is relatively simple and easy to fabricate, compared to other proposed three-terminal devices based on spin transfer torque.

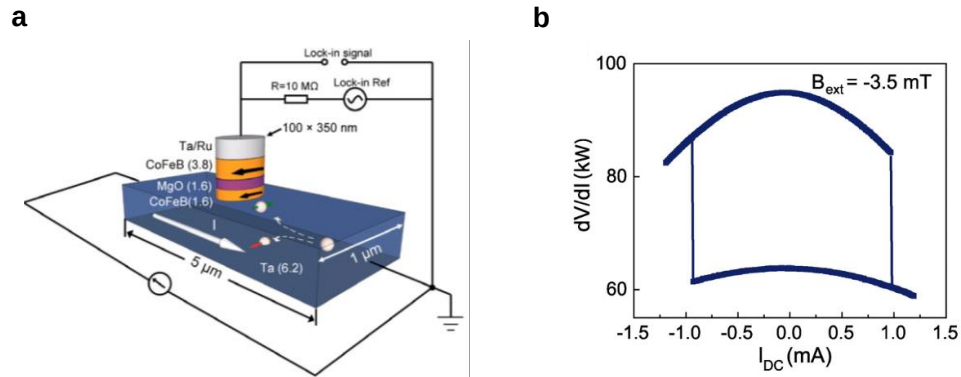


Figure 1.6: Spin Hall effect induced switching for in-plane magnetized nanomagnet. **a**, Schematic for a three-terminal spin Hall effect device and the circuit for measurements. **b**, Tunnel magnetoresistance as a function of applied dc current. An in-plane external magnetic field of -3.5mT is applied to compensate the dipolar coupling of the free layer and fixed layer. Adapted from Ref.[34].

1.3.2 Two-dimensional electron gases

Two-dimensional electron gases (2DEGs) – The study of spin charge interconversion in 2DEGs can be dated back to as early as the experimental observation of spin Hall effect[18], where a separation of spin polarization is observed on the two edges of the two dimensional hole gas channel. The low efficiency for spin current generation in these semiconductor-based two-dimensional systems, however, hinders the application of spin charge conversion to practical devices. The emergence of 2DEGs at oxide interface[35], which exhibit abundant properties such as interface conductivity with high mobility[35], superconductivity[36], and traces of magnetism[37-39], has attracted ever-growing research enthusiasm in the condensed matter physics. In addition, Caviglia et al.[40] showed that the quasi 2DEGs formed at the interface, where inversion symmetry breaking is present, shows strong gate-tunable Rashba spin orbit coupling, thus making the oxide interface a promising candidate for efficient spin-charge interconversion. The spin-charge interconversion in 2DEGs at these interfaces can be attributed to the so-called Rashba effect[8], as a result of the combination of the spatial inversion symmetry breaking that creates a built-in electric field along the direction normal to the interface and the spin orbit coupling in these oxides that together form the interface. In particular, with low temperature mobility comparable or even larger than that of Silicon, and the gate-tunable transport properties, STO-based 2DEGs are promising candidates for spintronics applications, which have already shown unparalleled high spin-charge interconversion efficiencies[4]. The spin to charge conversion experiments were demonstrated by Lesne et al.[4] in STO/LAO/NiFe system using a spin pumping set up shown in Fig. 1.7a. NiFe is used to pump a pure spin current into the 2DEGs at the STO/LAO interface, where it is converted into a 2D charge current. The effective spin to charge conversion efficiency, λ_{MEE} is determined to be 6.7 nm at 10 K. Strikingly, the conversion efficiency can be tuned

by the back-gate voltage as shown in Fig. 1.7b, where a sign flipping of the spin to charge conversion was observed across zero volt.

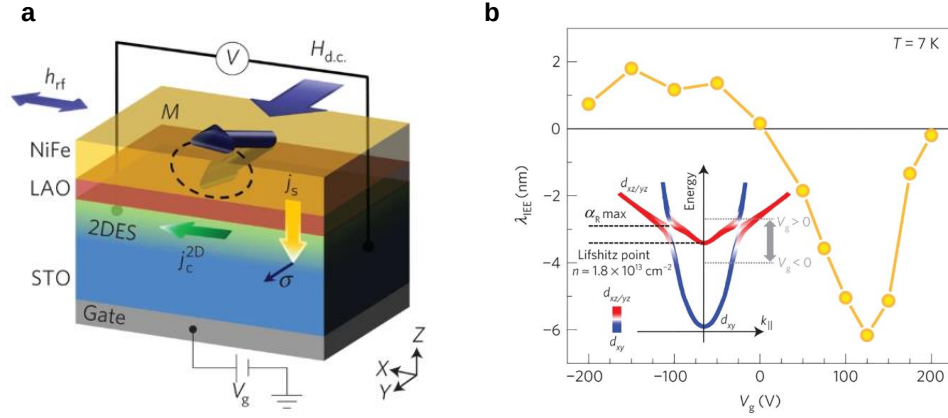


Figure 1.7: Gate-tunable spin-charge interconversion in 2DEGs at LAO/STO interface. a, Illustration of the spin pumping experiments performed in this study. b, Gate-dependent inverse Edelstein effect length, λ_{IEE} . Inset: Sketch of the band structure of the 2DEGs. Adapted from Refs. [4, 41-43].

The LAO/STO 2DEGs system studied at 10K by Lesne et al. was further investigated by several other groups up to room temperature. At 300K, the dielectric constant of STO decreases to 300, as compared to the dielectric constant of 20000 below 4K, inhibiting the generation of a large electric field. To make matters worse, the mobility of charge carriers in STO/LAO 2DEGs is known to be two orders of magnitude lower at high temperature than at low temperature. The spin to charge (λ_{IEE}) as well as charge to spin (q_{DEE}) efficiencies for STO/LAO 2DEGs at various temperatures are summarized in Table 1.1 (The spin-charge interconversion efficiencies for various interfaces are summarized in Table 1.1).

Oxide interface	Spin-charge interconversion efficiency	Refs.
LaAlO ₃ /SrTiO ₃	$\lambda_{IEE} = 6.4$ nm at T = 10K	[4]
	$\lambda_{IEE} = 6.7$ nm at T = 20K	[44]
	$\lambda_{IEE} = 1$ nm at T = 75K	[45]
	$q_{DEE} = 0.63$ nm ⁻¹ at T = 300K	[46]
Al/SrTiO ₃	$\lambda_{IEE} = 28$ nm at T = 15K	[47]
Ar-irradiated SrTiO ₃	$\lambda_{IEE} = 0.23$ nm at T = 300K	[48]
Cu/Bi ₂ O ₃	$\lambda_{IEE} = -0.17$ nm at T = 300K	[49]
Ag/Bi ₂ O ₃	$\lambda_{IEE} = 0.15$ nm at T = 300K	[49]

Table 1.1: Spin-charge interconversion efficiency for 2DEGs at oxide interfaces. λ_{IEE} : Inverse Edelstein length; q_{DEE} : charge to spin conversion efficiency by direct Edelstein effect. Adapted from Ref. [50].

With the realization of gate-tunable spin to charge conversion in 2DEGs[4], electric field control of Rashba SOC has been studied extensively, to achieve next generation energy efficient logic devices[1]. For example, semiconductor GeTe has been predicted to

exhibit a bulk Rashba band structure, in which the switching of polarization reverses the spin texture of the Rashba split Fermi contour[11]. Because of the high-leakage nature of GeTe, the experimental evidence of ferroelectricity was only obtained in thin film GeTe by piezoresponse force microscopy[51]. A successful demonstration of ferroelectrically controllable spin to charge conversion was reported by Noël et al.[52]. In their study, the ferroelectric like behavior under high electric field in SrTiO₃[53] was utilized to control the inverse Edelstein effect in the 2DEGs at the SrTiO₃/Al interface. It was shown that both the electrical conductivity and spin to charge conversion efficiency display hysteretic dependence on the applied gate voltages. The output voltage was fully reversed upon the change of polarity of the applied electric field pulses in the spin pumping experiments and remains stable. This hysteretic behavior of the spin to charge conversion efficiency reveals another important advantage associated with spintronics-based memory and logic devices – bipolarity. The bipolar dependence of spin to charge conversion efficiency on ferroelectricity is crucial to implement a ferroelectrically controlled memory and logic device (Fig. 1.8) akin to the magnetoelectric spin orbit (MESO) devices[1]. The concept of a ferroelectrically controlled memory and logic device is displayed in Fig. 1.8a and Fig. 1.8b. At the interface between the SOC material and the ferroelectric material, electrons are accumulated or depleted depending on the orientation of the ferroelectric polarization. The reversal of local electric field by ferroelectric polarization at the interface with Rashba SOC could in principle reverse the chirality of spin texture of the Fermi contours. Therefore, the charge currents converted from the spin current generated by the ferromagnet will undergo a reversal of the direction upon the switching of ferroelectric polarization, which lays the basis for the design of devices such as bipolar memory.

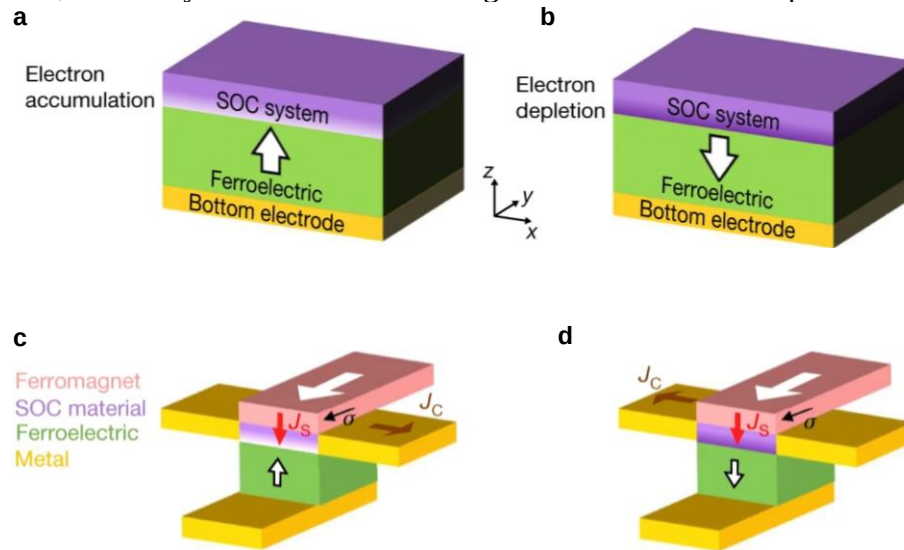


Figure 1.8: Ferroelectrically controlled spin to charge conversion. **a, b**, Schematic illustration of ferroelectric Rashba device. The ferroelectric material is shaded in green and the spin orbit coupling (SOC) layer is shaded in purple. Upon switching, electrons are accumulated in the SOC material or depleted, generating an electric field whose sign depending on the orientation of the ferroelectric polarization. **c, d**, Non-volatile device operated by ferroelectrically controlled Rashba SOC. A spin

current is generated by the top ferromagnet and gets converted by the 2DEGs at the SOC material and Ferroelectric interface. The direction of charge current reverses upon the switching of the polarization of the ferroelectric. Adapted from Ref. [52].

1.3.3 Topological insulators

Topological insulators – Another important class of materials for spin-charge interconversion is topological insulators, in which the spin of the charge carriers is locked at a right angle to the momentum. Due to the presence of spin momentum locking, topological insulators are considered as promising candidates for efficient spin current as well as spin orbit torque generation. As shown in Fig. 1.9a and Fig. 1.9b, when an in-plane charge current is applied to the surface states of a topological insulator, more forward going electron states are occupied than the backward going electron states, which, due to the spin momentum locking of the orientation of the spin and the momentum in the surface states, necessarily generates a non-equilibrium surface spin accumulation. When coupled to an adjacent ferromagnet, this non-equilibrium spin accumulation can transfer spin angular momentum to the magnet by exerting a spin transfer torque, which can induce magnetization dynamics[54] or switching of the adjacent magnet[55]. The two vector components of the current induced torque, in the $\hat{m} \times (\hat{m} \times \sigma)$ (in-plane) and $(\hat{m} \times \sigma)$ (out-of-plane) directions, are shown in Fig. 1.9a. The figure of merit of primary interest for topological insulators is spin-torque ratio (spin orbit torque efficiency), θ_{SOT} , that is the strength of the in-plane torque (damping like torque) per unit charge current density in the spin current source material. And θ_{SOT} for Bi_2Se_3 is determined by Mellnik et al.[54] to be 2.0 – 3.5 at room temperature, which outperforms heavy metals and 2DEGs. The efficient spin current and spin-torque generation in topological insulators makes topological insulators a promising candidate for spintronic devices, because θ_{SOT} fundamentally determines the current needed for efficient magnetic manipulation[34, 56, 57]. The current-induced efficient switching of magnetization by topological insulators is revealed by several topological insulator/magnet heterostructures[55, 58]. A heterostructure consisting of a topological insulator and a magnet is shown in Fig. 1.9c, in which a dc current conducts on the y direction. With dc currents and assisting magnetic fields along $\pm y$ directions, four stable states are possible (Fig. 1.9d). It was found by Fan et al.[55] that with an assisting magnetic field on the y direction, the z component of the magnetization can be switched deterministically by sweeping a dc current. The critical current density required for switching is determined to be below $8.9 \times 10^4 \text{ A cm}^{-2}$ at 1.9K, showing great potential for low-power magnetization switching.

The large spin-charge interconversion efficiency suggested by spin momentum locking in topological insulators have attracted intense research interest, and Table 1.2 gives a summary of the spin-charge interconversion efficiencies for some of the most studied topological insulators.

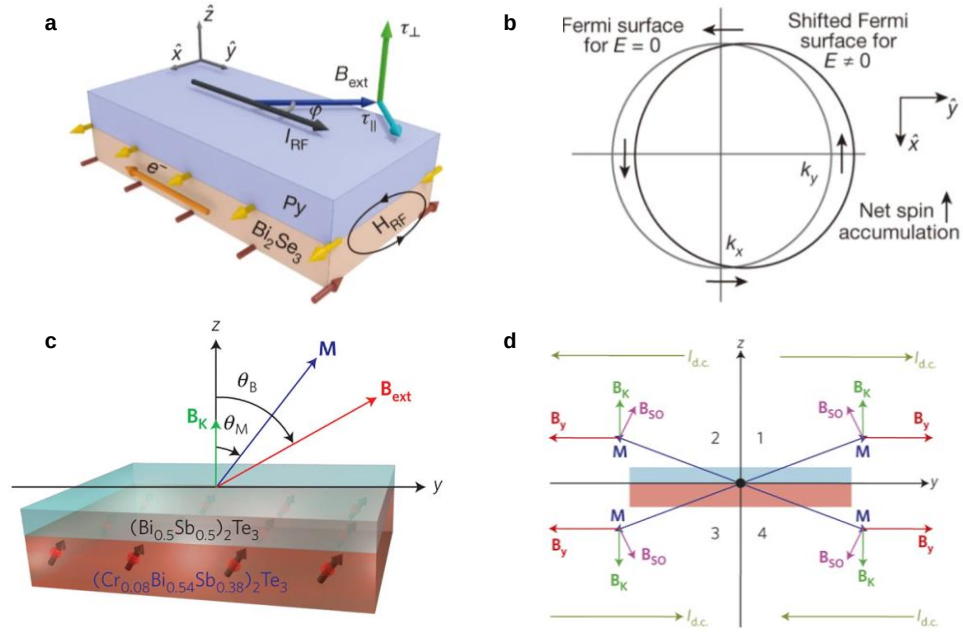


Figure 1.9: The mechanisms of current induced spin accumulation and magnetization switching in topological insulators. **a**, Illustration of the heterostructure and the coordinate system. Yellow and red arrows denote spin moment directions. Py, permalloy. **b**, Schematic illustration of the mechanism by which an in-plane current in the topological insulator surface states generates a non-equilibrium spin accumulation, on account of locking between spin directions and wavevector for electrons in the surface states. The arrows denote the direction of spin magnetic moments, which are opposite to the spin angular momenta. Adapted from Ref. [54]. **c**, Schematic of the heterostructure for magnetization switching. B_{ext} is the external magnetic field and M is the magnetization of the bottom magnetic topological insulator layer. B_K is the out of plane anisotropy field. **d**, Schematic of the four stable magnetization states (panels 1-4) when passing a large dc current, and applying an assisting external magnetic field B_y in the $\pm y$ directions. B_{SO} and B_K are the effective spin orbit field induced by dc current and the anisotropy field respectively. Adapted from Ref. [55].

Materials	Spin orbit torque efficiency, θ_{SOT}	Refs.
Bi_2Se_3	2.0 – 3.5 at $T = 293\text{K}$	[54]
	0.42 at $T < 50\text{K}$	[59]
	0.16 at $T = 293\text{K}$	[60]
	18.6 at $T = 293\text{K}$	[61]
	1 – 1.75 at $T = 293\text{K}$	[62]
$(\text{BiSb})_2\text{Te}_3$	0.4 at $T = 293\text{K}$	[60]
	2.5 at $T = 293\text{K}$	[63]
Bi_2Te_3	0.08 at $T = 293\text{K}$	[63]
SnTe	1.4 at $T = 293\text{K}$	[63]

Table 1.2: Spin charge interconversion efficiency for various topological insulators.

1.4 Complex oxides

This section introduces the broad range of functional properties that complex oxides host and an elaboration on their relevance as well as potential application to the field of spintronics.

The fundamental building blocks ABO_3 of perovskite complex oxide is shown in the top left of Fig. 1.10 with the eight A site cations on the eight corners of the cubic unit cell and the six B site cations located on the six face centers of the cubic unit cell. Nearly 75% of the elements in the periodic table can be incorporated either on the A site or on the B site, offering a great abundance of materials with the same structure. Furthermore, the abundance of materials with the same structural motif provides us great flexibility in heterostructure fabrication (Top right in Fig. 1.10), giving rise to endless interfaces and heterostructures for emergent phenomena. This large variety of materials and interfaces are the main contributors to the extensive diversity of physical properties we observe in complex oxides. Electronically, a wide range of electronic transport properties from insulators ($LaAlO_3$, $NbTiO_3$ and $LaVO_3$) [65], semiconductors ($SrTiO_3$ and $BaSnO_3$), metals ($SrVO_3$ and $SrRuO_3$) and superconductors (YBCO) are observed. Magnetically, common magnetic ground states of para- and diamagnetism ($SrVO_3$ and $SrTiO_3$), ferromagnetism ($SrRuO_3$ and $GdTiO_3$) and anti-ferromagnetism ($LaVO_3$ and $SmTiO_3$) are reported. Moreover, structural polarization related states such as pyroelectricity, piezoelectricity and ferroelectricity are also seen in complex oxides. Intriguingly, some of these properties emerge in the same material, enabling an intrinsic coupling of multiple ordering parameters that were initially thought to be mutually exclusive. An excellent example of the coexistence of multiple ordering parameters is multiferroic materials such as $BiFeO_3$ [66] and $TbMnO_3$ [67], in which magnetization and ferroelectric polarization coexists. Colossal magnetoresistance materials [68, 69] such as $La_{0.7}Sr_{0.3}MnO_3$ [70], on the other hand, have magnetism and orbital ordering that strongly affect the electronic transport properties. Last but not the least, the coexistence of metal-like free carrier concentration and electrical polarization has been observed in polar metals such as $LiOsO_3$ [71]. The wide range of properties that complex oxides host fulfill the spintronics requirements from magnetism, strong spin orbit coupling to multiferroicity.

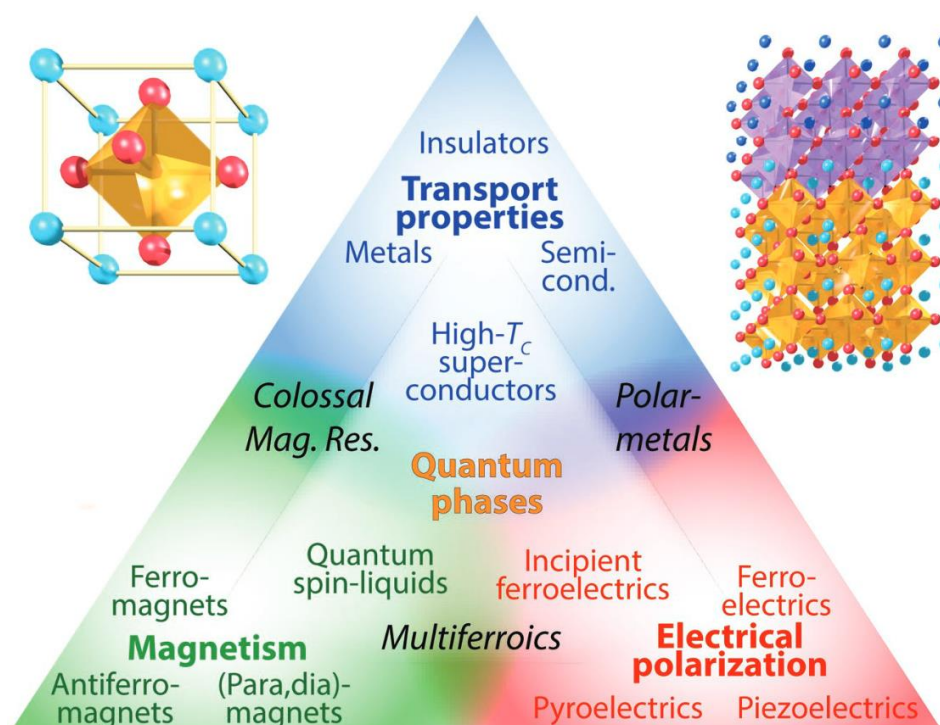


Figure 1.10: The wide range of functional properties and electronic phases hosted by ABO₃ perovskite family of complex oxides. The BO₆ octahedral unit is located at the center of the unit cell, with A site atoms on the eight corners (Top left). The simple ABO₃ structures allows for a large variety of heterostructures with diverse properties (Top right). Colossal Mag. Res.: Colossal magnetoresistance; Semi-cond.: Semiconductor. Adapted from Ref. [64].

Besides the abundant properties that complex oxides harbor, the capability of modifying the complex interaction between the charge, spin, orbital and lattice degrees of freedom can give rise to new behaviors at the oxide heterostructure interfaces as illustrated in Fig. 1.11. The most obvious effect at an interface is the breaking of symmetry caused by the chemical discontinuity that leads to a modification of electronic and structural properties. A broken symmetry by bringing two complex oxide materials with different chemical potentials together can lead a charge transfer, which in turn alters the doping level of the layers. This could in principle induce new electronic phases at the interface, as exemplified by the metallic and magnetic orders at the manganite interface and superconducting state at the LaAlO₃/SrTiO₃ interface. Furthermore, the close proximity in energy of different structural, electronic and magnetic phases makes complex oxides highly sensitive to epitaxial strain that is generated at the interface between two materials with different lattice parameters but grown epitaxially. The application of biaxial strain through epitaxy is powerful method in controlling the lattice distortions, which in turn influence the electronic properties. When two polar materials come into contact, the electrostatic interactions also come into play, and the boundary conditions at the interface lead to an electrostatic coupling between the two layers. Furthermore, competition

between different interactions at the interface and frustration can also give rise to new structural and electronic phases. The tunability on the interaction between charge, spin, orbital and lattice degrees of freedom can essentially provide us extra knobs when studying complex oxide spintronics.

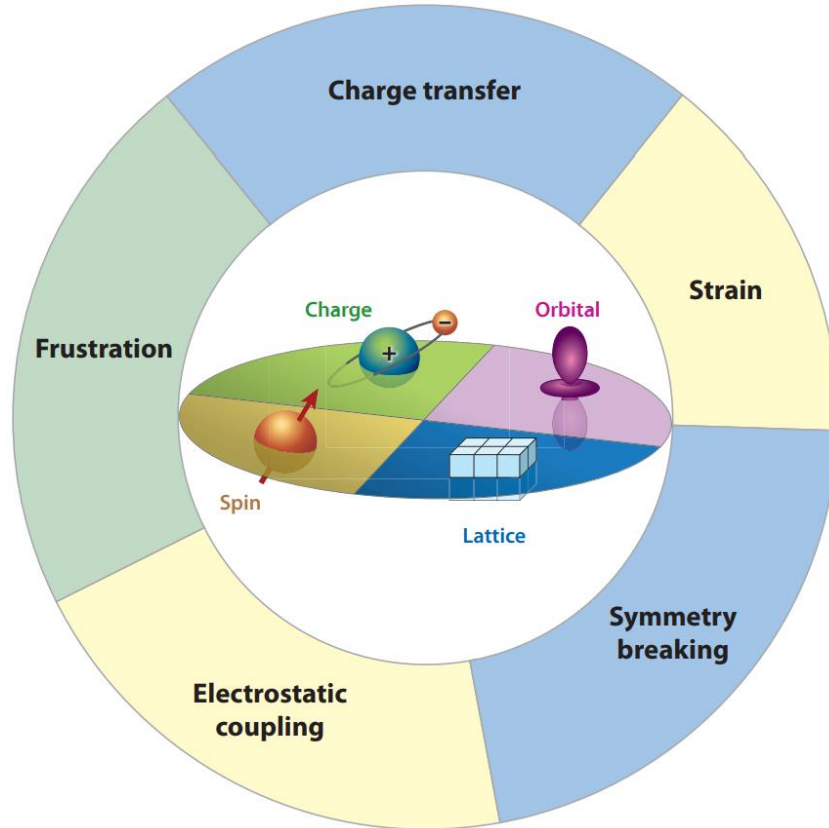


Figure 1.11: Interaction between different degrees of freedom and tunability of properties in complex oxides. Rich properties of complex oxides arise from the complex interaction between the charge, orbital, spin and lattice degrees of freedom. These interactions can be modified at the interface between different oxides through the effects of local symmetry breaking, charge transfer, electrostatic coupling, strain and frustration. Adapted from Ref. [72].

Given the large variety of properties and complex interactions that complex oxides host, a great number of complex oxides can be identified as potential candidates for spin current generation, manipulation and detection in spintronics. One of the focuses in the field spintronics is efficient spin current generation, and conventionally heavy metals with large spin orbit coupling such as Pt, W, Ta stand out as materials with large spin Hall angles. Strong spin orbit coupling is one of the many intriguing properties that complex oxides harbor, and large spin orbit coupling in iridates family (e.g., SrIrO_3) and PtCoO_2 has been theoretically and experimentally reported. The material candidates with large spin orbit coupling and even topological behavior are summarized in Table 1.3. Another class of material in demand for spintronics study is ferromagnetic materials, which usually serve

as detectors for magnetization perturbation induced by spin-orbit torques. CoFe, CoFeB with high Curie temperature and large saturation magnetization are the most common conventional ferromagnetic metals used for spintronics applications. Their oxide counterparts such as $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$, $\text{Y}_3\text{Fe}_5\text{O}_{12}$, which exhibit high Curie temperature and decent saturation magnetization at room temperature are good ferromagnetic candidates for all oxide heterostructures.

Property	Material	Structure
Ferromagnet	$\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ [70]	Perovskite, ABO_3
	$\text{Sr}_2\text{FeMoO}_6$ [73]	Double perovskite
	$\text{Y}_3\text{Fe}_5\text{O}_{12}$ [74, 75]	Garnet
Spin-orbit coupling	SrIrO_3 [76] Sr_2IrO_4 [77]	Perovskite, ABO_3
	$\text{Pb}_2\text{Ir}_2\text{O}_7$ [78] $\text{Bi}_2\text{Ir}_2\text{O}_7$ [79]	Pyrochlore
	PtCoO_2 [80]	Delafossite
	BiFeO_3 [66] YMnO_3 [81]	Perovskite

Table 1.3: Material candidates in the family of complex oxides for spintronics application.

Recently, significant research efforts in the field of spintronics have been attracted to antiferromagnets, among which Néel vector switching by spin-orbit torque[82] and spin transport with the assistance of antiferromagnet magnons[83] show great potential for low energy, high density memory and logic devices. Specifically, insulating antiferromagnets have shown to have the capability of carry spin angular momentum by the propagation of spin waves[84-86], by which magnetization switching[87, 88] has been made possible, with the flowing of electrons in the antiferromagnets. Moreover, spin transmission in some antiferromagnets with easy-plane[89] or easy-axis[83, 90] anisotropy can occur for the distance up to several micrometers, greatly preserving the spin coherence over long distances. However, despite the advantages magnon current has over spin current, the most effective way employed so far to modulate the spin transport in antiferromagnets is to use a constant magnetic field or magnetic field pulses, of which up to several tesla might be required depending on the strength of anisotropy of the antiferromagnet[83, 89, 90]. The application of utilizing the aforementioned advantages exhibited by conventional antiferromagnets to practical spintronic devices is hindered by the requirement of energetically prohibitive magnetic fields to control the spin dynamics. Multiferroic materials such as BiFeO_3 [66], in which antiferromagnetism and ferroelectricity coexist, on the other hand provide us with the feasibility to control spin transport with electric fields rather than magnetic fields, due to the intrinsic coupling between ferroelectric and antiferromagnetic orders[91-93]. The controlling of ferroelectric order by an electric field, however, costs energy typically thousands of times smaller than that of magnetic order, indicating that switching the ferroelectric order that in turn change the antiferromagnetic order could be a viable path to energy efficient spintronic devices.

1.5 Organization of dissertation

This dissertation is organized as follows:

First, chapter 1 provides the motivation and an introduction to the scientific background pertinent to this study. Following is a review on the prominent physical mechanisms for spin current generation as well as the classes of materials that have been most well studied in the field of spintronics. This chapter is finished with a brief introduction to a new class of material-complex oxides that are proposed as a promising candidate for spin current generation, manipulation and detection.

Chapter 2 will start with a literature review on the materials that I am going to study as well as an introduction to the synthesis technique that is employed for the growths of the materials. Lastly, the characterizations on physical properties from structure, magnetism to spin transport are thoroughly discussed.

Chapter 3 will investigate the significance of epitaxy in realizing efficient spin current generation in all oxide system. Presented in this chapter is a comprehensive study on the structure, electronic interaction and magnetism that are unique to the epitaxial oxide heterostructure utilizing various techniques; and the efficient spin current generation as a manifest of the epitaxial interface is highlighted. In the end, I come to a conclusion that epitaxy is a powerful tool we can make use of for achieving efficient spin current generation. This also motivates me to explore the abundant physical properties that complex oxides host to manipulate the spin current that can be efficiently generated in epitaxial all oxide systems.

In chapter 4, I will introduce another degree of freedom to control spin current in multiferroic BiFeO₃. This chapter starts with the investigation on the capability of BiFeO₃ of transmitting spin current with resorting to various techniques such as ST-FMR and direct magnetization switching. Then the tunability of spin propagation in BiFeO₃ by ferroelectric polarization is demonstrated; and bistable spin transmission states with high/low ratio around 40% is achieved. In chapter 4, I utilize RHEED assisted PLD with atomic scale precision to control the interface termination, which determines the bulk polarization of BiFeO₃. This motivates the manipulation of ferroelectric polarization by electric fields highlighted in chapter 5.

Chapter 5 discusses the control of spin transmission in BiFeO₃ via electric fields besides interface termination. I will show that the spin transmission in BiFeO₃ can be tuned by the polarity of the applied external fields, which essentially manipulate the ferroelectric polarization as the interface termination does. The ST-FMR measurements on the spin transmission in BiFeO₃ with opposite polarizations agrees with the results in Chapter 4. Lastly, I will demonstrate that the spin torque that arises from BiFeO₃ is sufficiently efficient to switch the magnetization of a ferromagnet with perpendicular magnetic anisotropy.

Chapter 6 serves as a brief summary of the findings presented in this dissertation and leaves the readers with suggested future directions to investigate in this field.

Chapter 2

Growth and characterization of all oxide heterostructures

This chapter provides an introduction to the synthesis as well as characterization techniques I employed in this study. It starts with some quick surveys on the properties of the materials explored in this study; and they are SrIrO_3 with large spin orbit coupling, BiFeO_3 with multiferroicity and $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ with room temperature ferromagnetism. Following is an introduction to the synthesis technique – Pulsed Laser Deposition that is employed for the growths of the thin films. Then the characterization of the physical properties of the heterostructures from structure, magnetism, to spin transport is discussed thoroughly.

2.1 Model system

2.1.1 SrIrO_3 – Iridate with large spin orbit coupling

Spin orbit coupling (SOC), which locks the momentum of an electron to its spin, has been shown essential for giving rise to many novel physical phenomena such as spin-charge interconversion. SrIrO_3 , a typical metallic member of the strong spin orbit coupling iridate family, therefore is a promising candidate for efficient spin current generation (spin orbit torque generation). Besides strong SOC, physical properties such as metallicity and magnetism are crucial for efficient spin current generation as well. These properties in principle originate from the competition between strong SOC (ζ_{SO}) and other interactions including crystal field splitting (Δ) and electron correlation (U), due to their comparable strength in these material systems. As a prototypical SOC system, iridates have a SOC $\zeta_{\text{SO}} \sim 0.4$ eV, comparable to their oxide octahedra crystal field ($\Delta \sim 2$ eV) and electron correlation ($U \sim 0.5$ eV). The strong SOC of SrIrO_3 has been experimentally confirmed by the weak anti-localization effect measurements[76]. This strong SOC has been shown to be essential for superconductivity[98], Weyl semimetal[99], and quantum Hall effect[100, 101] in various iridates. In particular, in the case of Ruddelsden-Popper (RP) structure type $\text{Sr}_{n+1}\text{Ir}_n\text{O}_{3n+1}$ iridates, a strong SOC splits the t_{2g} band that forms as a result of crystal field splitting into $J_{\text{eff}} = 1/2$ and $J_{\text{eff}} = 3/2$ effective bands. Then electron correlation U can further lift the $J_{\text{eff}} = 1/2$ band into upper Hubbard band (UHB) and lower Hubbard band (LHB), forming a half-filled $J_{\text{eff}} = 1/2$ band[95] (Fig. 2.1b). Owing to the enhanced interlayer hopping induced by three-dimensionality in SrIrO_3 , the $J_{\text{eff}} = 1/2$ band mixes with the $J_{\text{eff}} = 3/2$ band at a certain level (Fig. 2.1c), thus making it a paramagnetic semimetal[77, 102]. Noteworthily, the semi-metallicity of SrIrO_3 fulfills the carrier conduction requirement for efficient spin-charge conversion. In addition, SrIrO_3 was predicted to host Dirac cone in the electronic band structure and can be modified into various topological phases[96, 103] depending on the broken inversion symmetry (Fig. 2.1d, e), offering possibilities to achieve energy efficient spintronic devices. Based on semi-metallicity, strong SOC and possible topological phases, SrIrO_3 was chosen as the spin current source in the study for this dissertation.

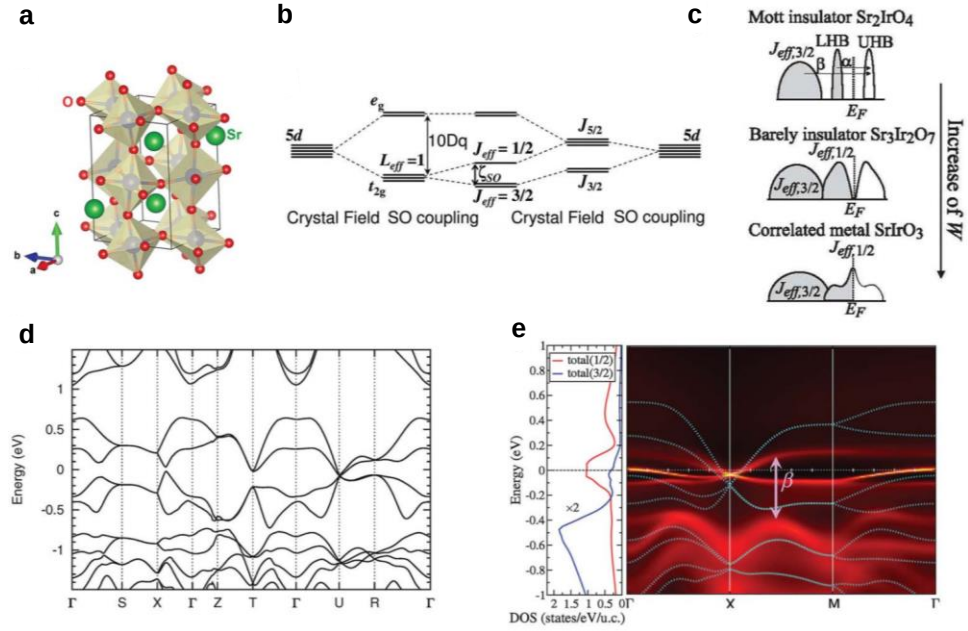


Figure 2.1: Crystal and electronic structure of orthorhombic SrIrO_3 . Adapted from Ref.[94] **a**, Crystal structure of orthorhombic SrIrO_3 . **b**, $5d$ level splitting by crystal field and spin orbit coupling. Adapted from Ref. [95]. **c**, Schematic diagram of $5d$ $\text{Sr}_{n+1}\text{Ir}_n\text{O}_{3n+1}$ compounds, which are well described by the effective total angular momentum J_{eff} states due to strong spin orbit coupling. E_F represents Fermi level and the arrow indicated the direction for the bandwidth W increase. Adapted from Ref. [77]. **d**, LDA calculation with Hubbard $U = 2$ eV and spin orbit coupling tuning parameter $\alpha = 2.0$. Adapted from Ref. [96]. **e**, Calculation with LDA + DMFT method. The dotted lines are obtained by the generalized gradient approximation plus U (GGA + U) method, the background bright curves correspond to the calculated results through LDA + DMFT. Adapted from Ref. [97].

2.1.2 BiFeO₃ – Multiferroic material

BiFeO_3 investigated since the late 1950's is the only room temperature multiferroic material, in which magnetic order and ferroelectric order coexist. Much of the early research interest was focused on the magnetoelectric coupling of BiFeO_3 ; however, these studies investigated the bulk crystal rather than the thin film, which was first stabilized in 2003[66] and dominates the research on BiFeO_3 since then. In the bulk, BiFeO_3 possesses a rhombohedrally distorted perovskite structure in which a spin cycloid is formed with period length of ~ 64 nm[104] and a Néel temperature of ~ 643 K[105]. Strain in BiFeO_3 thin films can break the spin cycloid and favors a G-type antiferromagnetic order wherein canting of the moments can produce a weak, long-range net magnetization ($\sim 0.1\mu_B$ per Fe)[66, 106], arising from the Dzyaloshinskii Moriya Interaction (DMI). The G-type antiferromagnetism is driven by the super exchange interaction between neighboring d^5 -

Fe^{3+} atoms and mediated by the O^{2-} between them. All the nearest spins point antiparallel to one another along the $[111]$ orientation (Fig. 2.2c) and in the (111) planes magnetic spins are ferromagnetically aligned (Fig. 2.2b). At the same time, BiFeO_3 thin films possess robust ferroelectricity, with a Curie temperature ~ 1100 K and large polarization $> 90 \mu\text{C}/\text{cm}^2$ [107]. Note that the origin of the ferroelectric nature in BiFeO_3 , different from traditional ferroelectrics, is that the two 6s electrons in Bi^{3+} hybridize with the surrounding oxygen ion, leading to a large displacement of Bi cation relative to the oxygen octahedra along the $[111]$ directions (Fig. 2.2a).

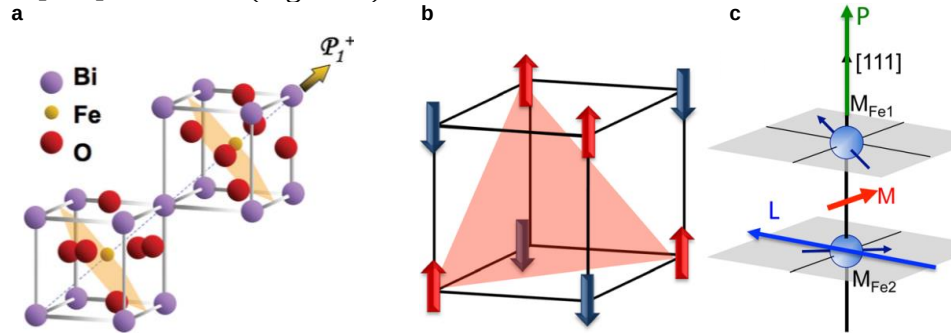


Figure 2.2: Ferroelectricity and antiferromagnetism of BiFeO_3 thin film. **a**, Schematic of the crystal structure of BiFeO_3 and the ferroelectric polarization on the $[111]$ direction. Adapted from Ref.[108]. **b**, Schematic of a G – type antiferromagnetism, in which the antiferromagnetic spins lie in the (111) plane and the canting of these spins forms a small magnetization that lies in the plane shown in **c**. Adapted from Ref.[109].

2.1.3 $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ – ferromagnetic material

In 1950's, ferromagnetism and magnetism were found in doped manganites, whose parent manganites were antiferromagnetic and insulating, drawing extensive research interest in the field of condensed matter physics. The intense study on the magnetic and electric properties of these manganites has given birth to important physical discoveries such as the colossal magnetoresistance (CMR) effect in $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_3$ (LCMO) thin film[68, 69]. The finding of CMR by doping LaMnO_3 inspired an everlasting research enthusiasm on manganites with large variety of A site cations, dopants and doping levels. Of all the possible compounds, $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ (LSMO) has attracted enormous research interest and has been the best studied manganites, due to the high Curie temperature that LSMO displays at a certain doping level.

A phase diagram for $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ is shown (Fig. 2.3), where the parent compound LaMnO_3 is on the left side. In LaMnO_3 , in which the Mn cation has a d^4 orbital configuration, its ground state is characterized with an orbital ordering pattern, thus making it antiferromagnetic (A-type) and Mott insulating. On the right side is the SrMnO_3 , in which Mn cation has a d^3 orbital configuration. Compared to LaMnO_3 , SrMnO_3 has G-type antiferromagnetic ordering and is band insulating. With the progressive doping of Sr into LaMnO_3 , various magnetically and charge ordered phase emerge. Among all the possible phases, one that is of our interest for this dissertation is the ferromagnetic metallic

phase that forms at 1/3 Sr doping level, due to the outstanding ferromagnetic properties – Curie temperature 380 K and almost 100 percent spin polarization.

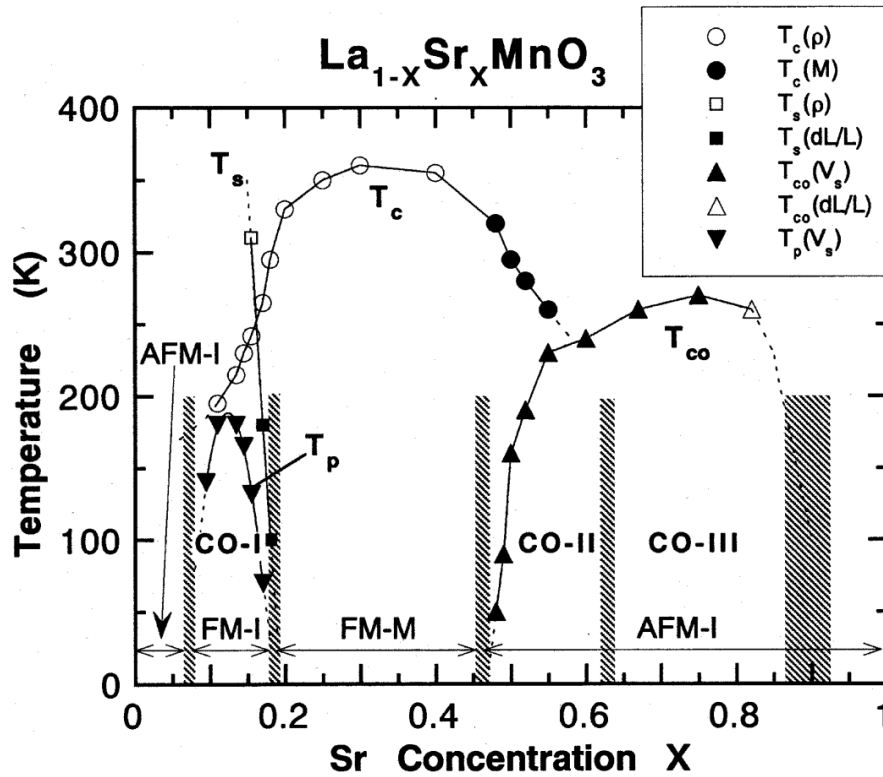


Figure 2.3: Phase diagram of $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ as a function of temperature and the concentration of dopant Sr. T_c : Curie temperature; T_s : structural transition temperature; T_{co} : charge ordering temperature; T_p : Polaron ordering temperature; The shaded regions correspond to the borders of three types of charge orderings (CO-I, CO-II, CO-III). AFM-I: antiferromagnetic insulator; FM-I: ferromagnetic insulator; FM-M: ferromagnetic metal. Adapted from Ref. [70].

2.2 Fabrication of the heterostructures

The section features the growth of high-quality single crystal thin films that were used to accomplish the scientific exploration of this dissertation. The technique I employ to synthesize the heterostructures in this study is the high-pressure reflection high-energy electron diffraction (RHEED) assisted pulsed laser deposition (PLD). During a growth, the target with the same composition as the thin films is ablated by a laser beam with high energy density. The material on the target surface is locally vaporized and forms an ablation plume that deposits on a substrate at high temperature (Fig. 2.4a). Since RHEED only gathers information from the surface layer of the sample, RHEED is employed to monitor the growth process. For a layer-by-layer growth, the atoms first spread two-dimensionally on the sample surface to complete a full layer and then grow the subsequent

layer. Such a periodic surface roughness change can be monitor by RHEED intensity oscillations. Fig. 2.4b shows a typical layer-by-layer growth, which results in an atomically smooth and fully covered atomic layers. At initial stage, RHEED intensity decreases as more and more atoms are deposited, which results in a deterioration of the surface roughness. The intensity reaches the minimum when the surface is half covered. After that, the intensity starts to recover and reaches the maximum again, indicating that the surface is fully covered and becomes atomically smooth again. Thus, by resorting to RHEED monitoring, the growth mode can be identified and the thickness can be determined by counting the RHEED oscillation periods.

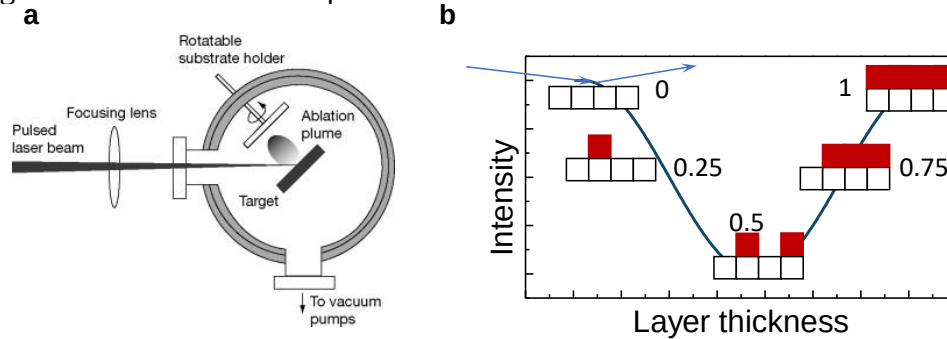


Figure 2.4: RHEED assisted PLD. **a**, Schematic diagram of a PLD process. Adapted from Ref. [110]. **b**, RHEED intensity oscillation during a layer-by-layer growth.

For all the heterostructures studied in this dissertation, (001)-oriented SrTiO_3 (STO) single crystal was used as the substrates for growth due to its lattice match to the thin films investigated in this study. The commercial STO substrates are first etched by buffered HF acid and then thermally annealed at 1000°C for 3 hours in oxygen environment, to obtain an atomically smooth and TiO_2 terminated surface. A successful treatment for the STO substrate is confirmed by atomic force microscopy (AFM), showing perfect atomic height terraces (inset in Fig. 2.5c). The sharp RHEED diffraction pattern (top left in Fig. 2.5a) also supports an atomically flat surface. Usually, LSMO is the first layer to grow on the STO substrate at 700°C , an oxygen partial pressure of 150 mTorr, a laser energy density of 1.5 J/cm^2 and a frequency of 2Hz. The RHEED oscillation (Fig. 2.5a) is recorded during the growth, showing a layer-by-layer growth mode. The atomically flat surface resulting from the layer-by-layer growth is confirmed by the AFM scans. Layer-by-layer growths for various materials (SRO, SIO, BFO) were also achieved with optimized growth condition, as displayed by the RHEED patterns as well as RHEED intensity oscillations in Fig. 2.5. The optimized growth conditions for the materials used in the study for this dissertation are summarized in Table 2.1.

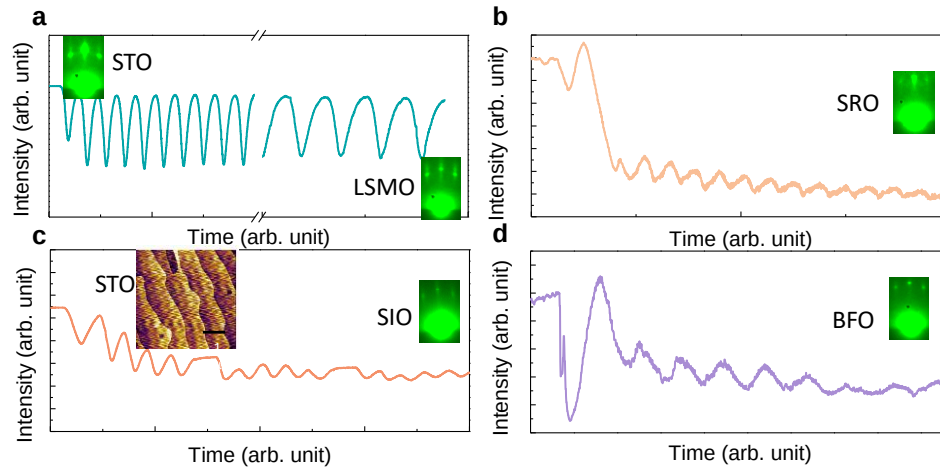


Figure 2.5: RHEED patterns and RHEED intensity oscillations monitored during the growth of **a**, LSMO; **b**, SRO; **c**, SIO; and **d**, BFO respectively.

Materials	Temperature (°C)	Pressure (mTorr)	Laser energy (J/cm ²)
La _{0.7} Sr _{0.3} MnO ₃	700	150	1.5
SrRuO ₃	700	100	1.5
SrIrO ₃	700	100	1.3
BiFeO ₃	700	100	1.5

Table 2.1: Recipe of layer-by-layer growth for various materials.

2.3 Characterization of the heterostructures

2.3.1 Structural characterization of the heterostructures

Atomic force microscopy (AFM) imaging of the (001)-oriented STO substrate is shown in Fig. 2.6a. Large atomic terraces are seen, indicating a successful treatment of the substrates. LSMO, BFO, SIO thin films grown with the optimized growth conditions in Table 2.1 were characterized with AFM (Fig. 2.6b, c, d), showing atomic terraces and confirming the layer-layer-layer growth mode observed with RHEED.

The structure of the heterostructures were characterized with both X-ray diffraction (XRD) and reciprocal space mapping (RSM). The 2θ - ω scan for a bilayer sample LSMO/SIO on (001)-oriented STO substrate is shown in Fig. 2.7a, which reveals that both LSMO and SIO are pure phase and in-plane strained to the STO substrate. The thickness fringes (Kiessig fringes) along the SIO (002) and LSMO (002) diffraction peaks indicate high crystalline quality and sharp interface structure. The same features are observed in the XRD scan for a trilayer sample LSMO/BFO/SIO on (001)-oriented STO substrate. The RSM scan for the bilayer sample further confirms that both LSMO and SIO are coherently strained to the STO substrate, with in-plane lattice constant equal to that of the substrate.

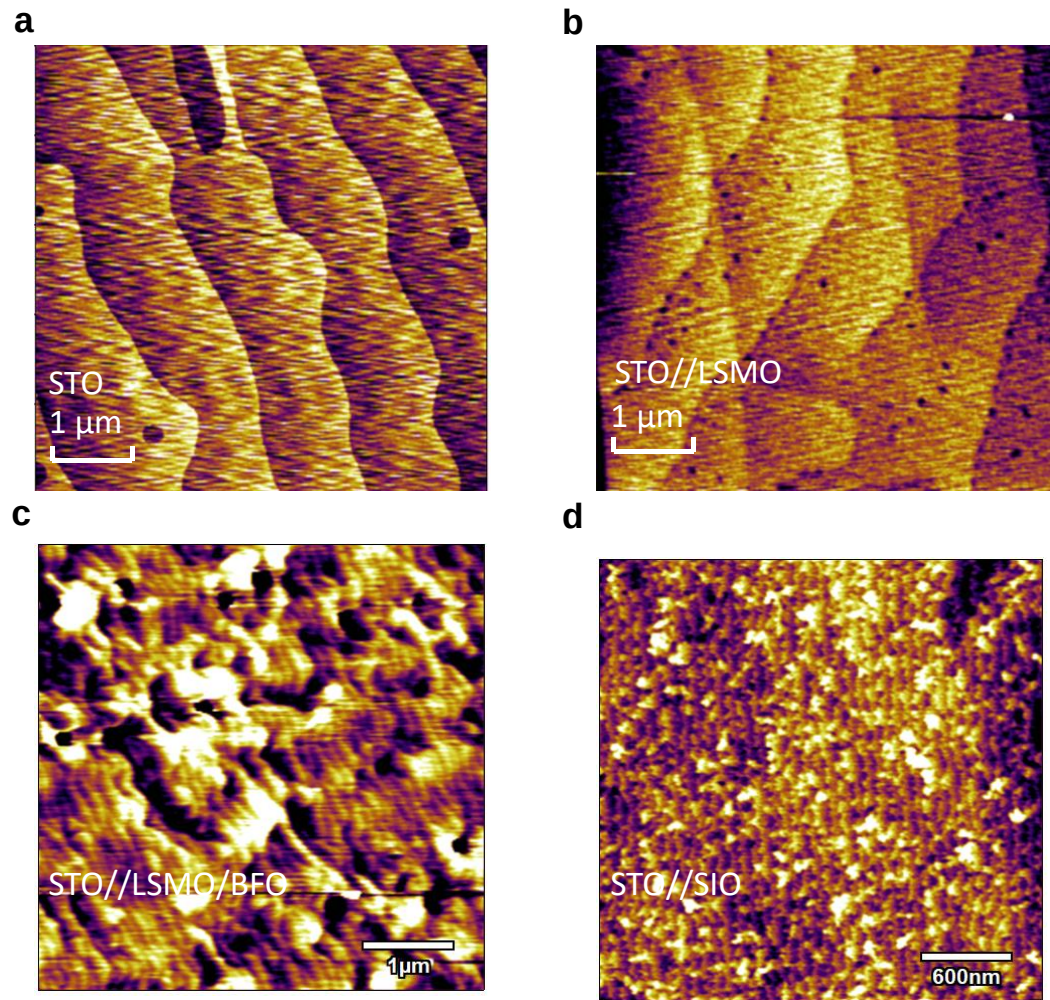


Figure 2.6: Atomic force microscopy imaging of STO, LSMO, BFO, and SIO respectively.

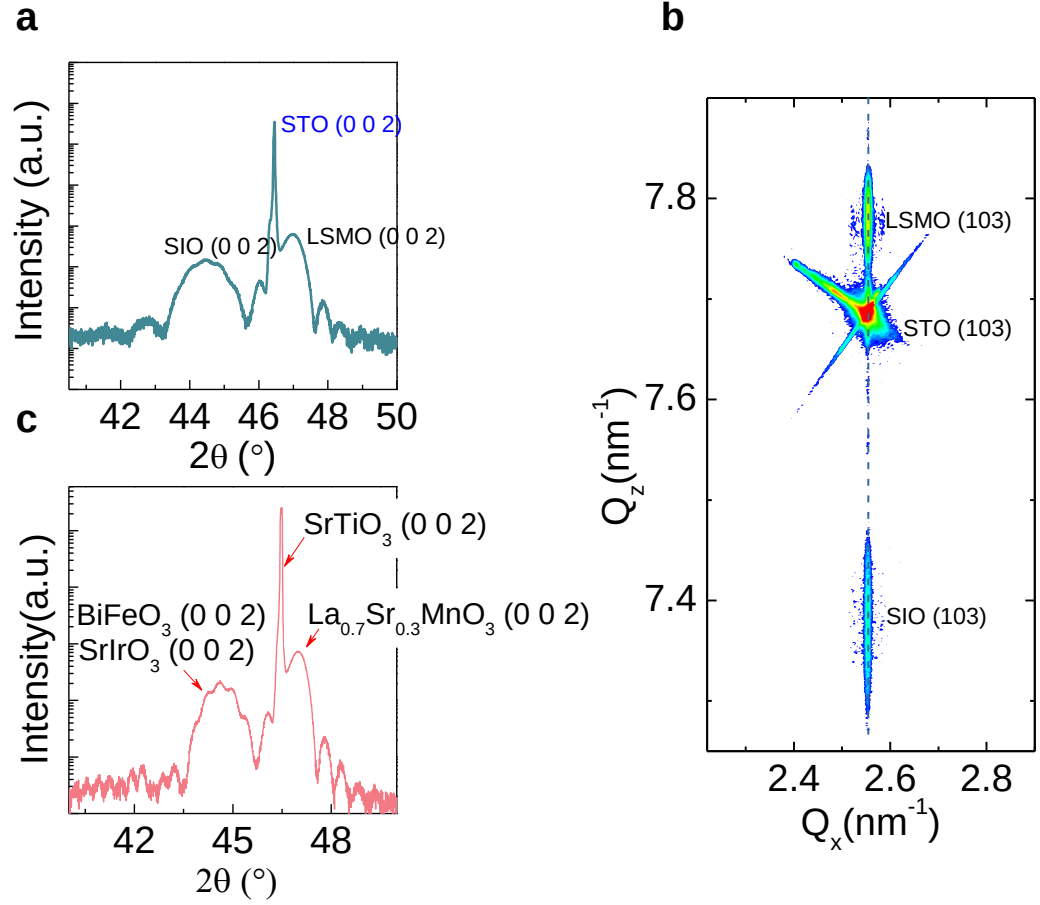


Figure 2.7: Structural characterization for the heterostructures by X-ray diffractometry. a, X-ray diffraction (XRD) 2θ-ω scan for a bilayer sample STO (001)//LSMO (20 nm)/SIO (10 nm). b, Reciprocal space mapping (RSM) for the same bilayer sample along the (103) zone axis of the STO substrate. c, XRD 2θ-ω scan for a trilayer sample STO (001)//LSMO (20 nm)/BFO (5 nm)/SIO (10 nm).

2.3.2 Magnetic properties of La_{0.7}Sr_{0.3}MnO₃ thin films

The magnetization of a 20 nm LSMO thin film grown on (001)-oriented STO was recorded with magnetic field of 100 Oe applied in the plane when cooling down the sample from 400 K to 10 K, showing a Curie temperature approximately 345 K (Fig. 2.8a). The magnetization of the same LSMO film measured as a function of the strength of the in-plane magnetic field is displayed in Fig. 2.8b. A coercive field of ~ 25 Oe and a saturation magnetization of ~ 300 emu/cc were determined from the scan, showing good agreement with that of high-quality LSMO thin films in literature[111].

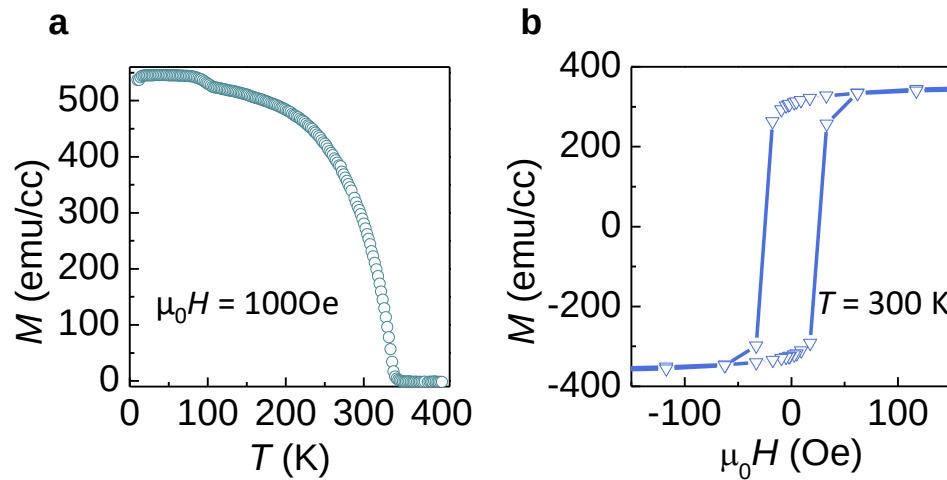


Figure 2.8: Magnetic properties of STO/LSMO (20 nm) by magnetometry. **a**, Saturation magnetization as a function of temperature. An in-plane magnetic field of 100 Oe is applied along the sample edge. **b**, Magnetization as a function of in-plane magnetic fields at room temperature.

2.3.3 Ferroelectric properties of BiFeO₃ thin films

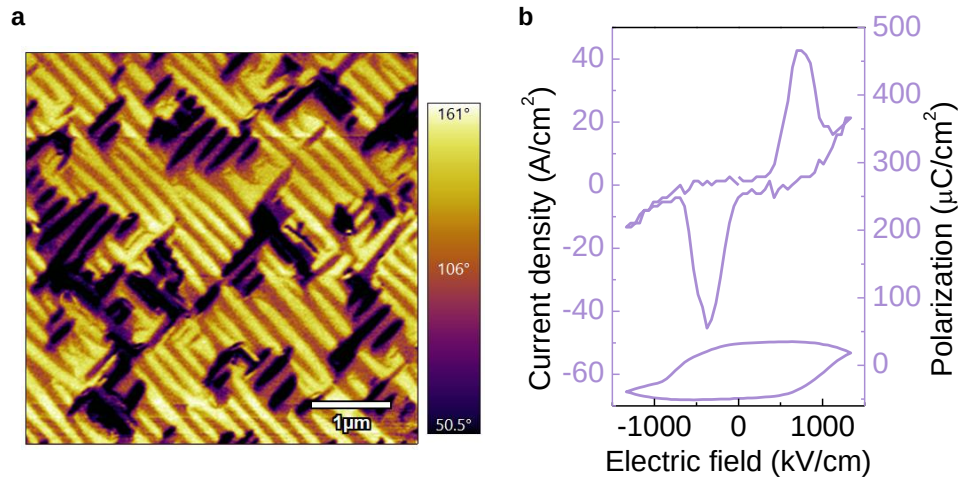


Figure 2.9: Ferroelectric characterization of BFO films. **a**, In-plane ferroelectric domain structures of LSMO/BFO. **b**, Ferroelectric polarization hysteresis loop of a BFO thin film (left axis); Ferroelectric switching current density as a function of applied electric field (right axis).

The ferroelectric polarizations of BFO lie on the eight body diagonal directions of the cubic lattice[112], due to the rhombohedral perovskite structure of BFO. These ferroelectric polarizations can be mapped out with piezoresponse microscopy (PFM) technique, which can simultaneously record the out-of-plane and in-plane piezoresponse signals. An in-plane ferroelectric domain structure of a LSMO (20 nm)/BFO (20 nm) thin film is shown as an example (Fig. 2.9a). The in-plane PFM image suggests the presence

of stripe-like ferroelectric domains separated by 71° domain walls. The ferroelectric polarization hysteresis loop of a BFO thin film was measured with Radiant Technologies Inc. RT 6000S ferroelectric tester probe station. An example scan for a sample LSMO (20 nm)/BFO (20 nm)/SIO (10 nm), in which where a hysteresis develops with an electric field exceeding 500 kV/cm is displayed in Fig. 2.9b.

2.3.4 Electrical transport

Epitaxial SrRuO_3 (SRO) thin film grown on (001)-oriented STO substrate with the optimized conditions in Table 2.1 displays layer-by-layer growth mode (Fig. 2.5b). The high-quality SrRuO_3 thin films undergo a paramagnetic to ferromagnetic phase transition at 150 K[113]. Below the Curie temperature, epitaxial SrRuO_3 shows good ferromagnetism with perpendicular magnetic anisotropy, which makes it a good ferromagnetic material for studying magnetization switching by current induced spin-torques[114]. The magnetic properties of SrRuO_3 in a bilayer sample ($\text{SrRuO}_3/\text{SrIrO}_3$) was carried out with anomalous Hall effect measurements. A dc current of 100 μA was applied in the channel along y direction and the transverse voltage was measured when sweeping the magnetic field along the out-of-plane direction (Fig. 2.10a). Example AHE scans at 50 K, 70 K and 90 K are displayed in Fig. 2.10b, c, d, showing good ferromagnetic properties with perpendicular magnetic anisotropy at these temperatures. The magnetic coercive fields are seen to progressively decrease when the sample temperature goes up, consistent with previous reports[115].

LSMO, a room temperature ferromagnet metal, serves as the current induced spin torque detector besides SrRuO_3 . The resistance of a 20 nm LSMO film on (001)-oriented STO substrate as a function a temperature is shown in Fig. 2.11a and the resistance decreases as temperature decreases, showing good metallic behavior. Moreover, the planar Hall resistance is recorded when varying the angle between the external magnetic field (50 Oe) and current direction. A sinusoidal $\sin 2\theta$ angular dependence is observed, indicating that no magnetic anisotropy is present in the film plane (Fig. 2.11d). This observation is consistent with the cubic structure of the STO (001) substrates. On the contrary, strong in plane anisotropy is observed in the planar Hall resistance measurements (magnetic field 50 Oe) for NdGaO_3 (NGO) (001)/LSMO (20 nm) (Fig. 2.11c), due to the biaxial strain exerted by the substrate[116]. NdGaO_3 has an orthorhombic crystal structure with the lattice parameters of $a = 5.43\text{\AA}$, $b = 5.50\text{\AA}$, and $c = 7.71\text{\AA}$. When grown on NdGaO_3 (001) substrate, a compressive strain (-1.04%) and a tensile strain (0.23%) are applied on the x and y axes of LSMO film, giving rise to a magnetic easy axis on the $[010]_o$ direction. Moreover, the resistance of a 10 nm SIO film on STO (001) substrate decreases as the sample cools down (Fig. 2.11b), suggesting a metallicity of SIO film at this thickness. Importantly, the resistance match between LSMO and SIO can mitigate the current shunting issue in the LSMO/SIO heterostructures, which is a notoriously prominent problem in heterostructures consisting of a metallic ferromagnet (e.g., CoFe, CoFeB) and a spin-orbit coupling layer (e.g., heavy metals, topological insulators).

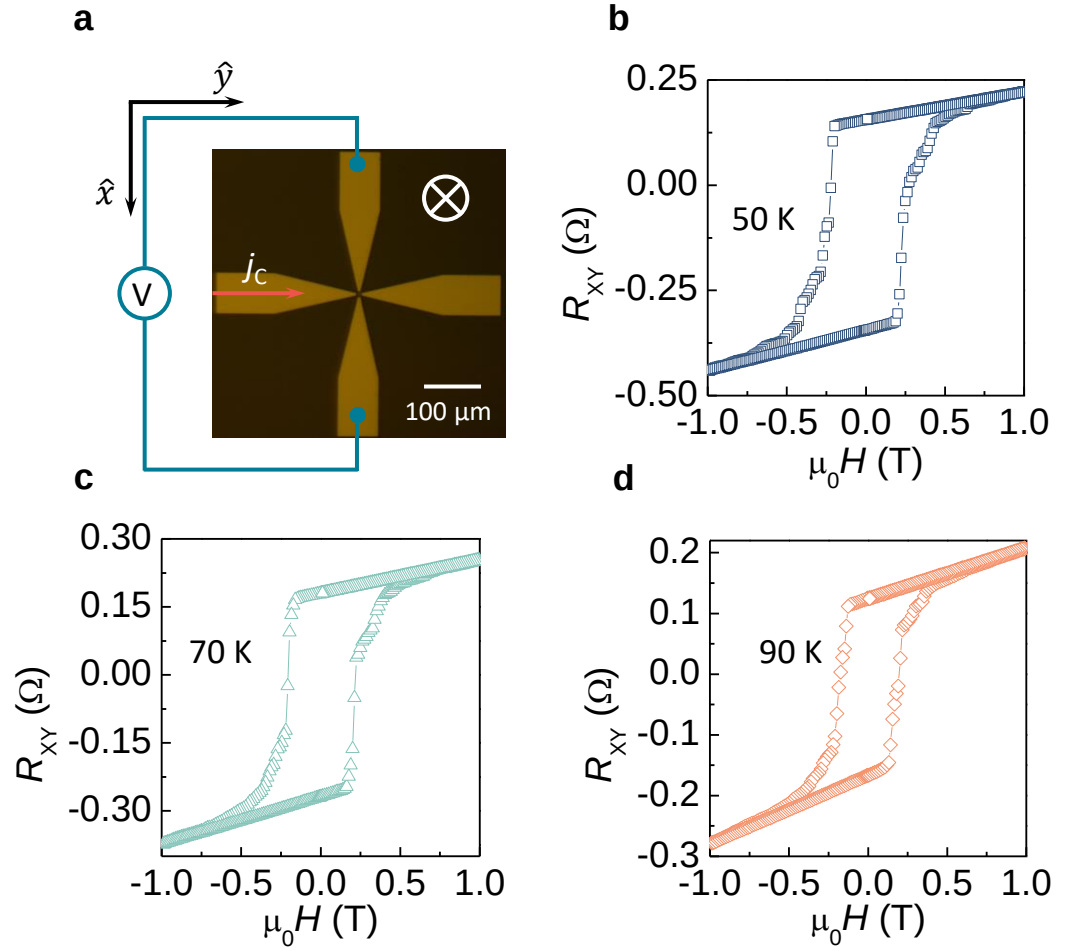


Figure 2.10: Temperature dependent anomalous Hall effect (AHE) of STO (001)//SRO (7 nm)/SIO (13 nm). **a**, An optical image of the Hall device for AHE measurement. The magnetic field sweeps along the out-of-plane direction. **b**, **c**, **d**, The anomalous Hall resistance as a function of magnetic field at 50 K, 70 K, and 90 K respectively.

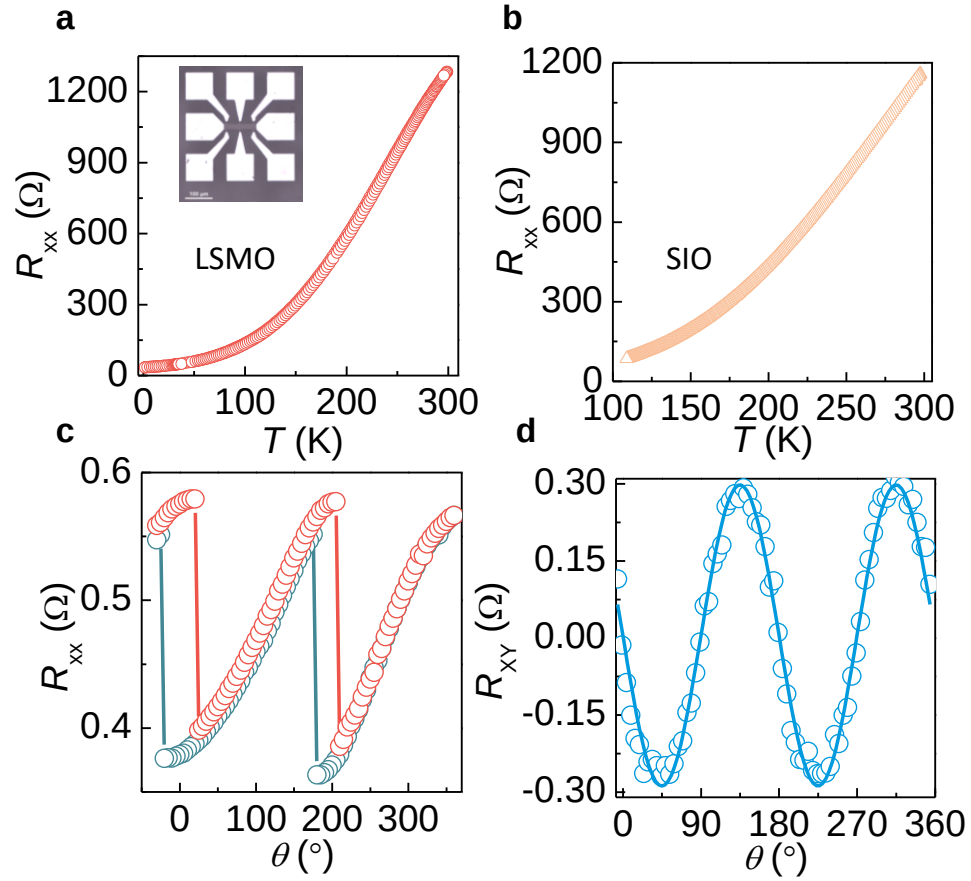


Figure 2.11: Transport properties of LSMO and SIO. **a**, Longitudinal resistance (R_{xx}) as a function of temperature for STO/LSMO (20 nm). **b**, Longitudinal resistance (R_{xx}) as a function of temperature for STO/SIO (10 nm). **c**, Transverse resistance as a function of in-plane field angle (θ) for NGO (001)/LSMO (20 nm). The strength of the in-plane magnetic field is fixed at 50 Oe. **d**, Transverse resistance as a function of in-plane magnetic field angle (θ) for STO (001)/LSMO (20 nm). The strength of the in-plane field is fixed at 50 Oe.

2.4 Spin transport

2.4.1 Spin-torque ferromagnetic resonance measurement

The strength of spin orbit torque generated in the all oxide heterostructures is characterized with a well-established technique-Spin torque ferromagnetic resonance (ST-FMR) developed previously to measure spin orbit torque efficiency from materials with spin orbit coupling. As shown in Fig. 2.12a, I applied an oscillating radio frequency current J_C to the strip in the in-plane configuration, with an in-plane external magnetic field H_{ext} at a 45° angle with respect to the current direction. Due to the spin Hall effect in SrIrO_3 , a

spin current J_S with spin polarization on x direction (denoted as blue and red arrows) accumulates at the LSMO/SIO interface. The spin angular momentum carried by the spin current exerts a damping-like torque ($\tau_{DL} \propto m \times (m \times \sigma)$) and a field-like torque ($\tau_{FL} \propto m \times \sigma$) on the adjacent ferromagnet LSMO. When the frequency of the applied current and the strength of the applied external magnetic field satisfy the ferromagnetic resonance condition, the magnetization of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ starts oscillating, yielding a resistance oscillation due to the anisotropic magnetoresistance of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$. Mixing the applied current and the anisotropic magnetoresistance of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ results in a measurable dc voltage, V_{mix} shown in Figure 2.12b, which is fitted by the sum of a symmetric Lorentzian and an anti-symmetric Lorentzian. The ST-FMR mixing dc voltage can be written as[117]:

$$V_{\text{mix}} = -\frac{1}{4} \frac{dR}{d\Phi} \frac{\gamma I_{\text{rf}} \cos\Phi}{\Delta 2\pi \left(\frac{df}{dH}\right)|_{H_{\text{ext}}=H_0}} [S F_S(H_{\text{ext}}) + A F_A(H_{\text{ext}})], \quad (2.1)$$

where $R(\Phi)$ is the device resistance as a function of the applied in-plane magnetic field angle Φ , I_{rf} is the microwave current flowing into the device, $\left(\frac{df}{dH}\right)|_{H_{\text{ext}}=H_0}$ is the field gradient of resonance frequency, $F_S(H_{\text{ext}}) = \frac{\Delta^2}{\Delta^2 + (H_{\text{ext}} - H_0)^2}$ is the symmetric Lorentzian function with half-width-half-maximum linewidth Δ , $F_A(H_{\text{ext}}) = \frac{(H_{\text{ext}} - H_0)\Delta}{\Delta^2 + (H_{\text{ext}} - H_0)^2}$ is the anti-symmetric Lorentzian function. S and A are the amplitudes of the symmetric and anti-symmetric Lorentzian functions respectively. Since $\tau_{DL}(\propto m \times (m \times \sigma))$ and $\tau_{FL}(\propto m \times \sigma)$ have a $\cos\theta$ and $\frac{dR}{d\Phi}$ has $\sin 2\theta$ angular dependence, both V_S and V_A have a $\sin 2\theta \cos\theta$ angular dependence on the external magnetic field[118], confirmed by the angular dependent measurements in Fig. 2.12c, d. The SOT efficiency can be expressed in terms of the extracted ST-FMR parameters[117]:

$$\xi_{||} = \frac{S}{A} \left(\frac{e}{\hbar}\right) \mu_0 M_{\text{S}} t_{\text{FM}} d_{\text{NM}} \sqrt{1 + \mu_0 M_{\text{eff}}/H_0}, \quad (2.2)$$

where M_S is the saturation magnetization of the ferromagnet, M_{eff} is the effective magnetization of the ferromagnet, and H_0 is the resonance field.

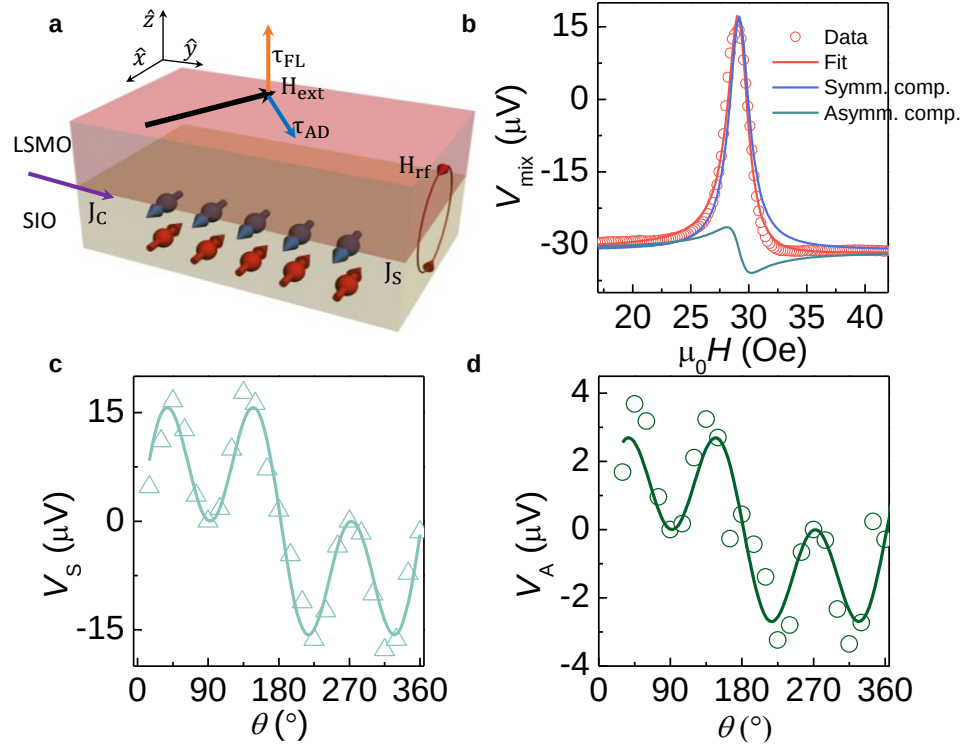


Figure 2.12: ST-FMR measurements. **a**, A schematic illustration of ST-FMR setup. The external magnetic field is applied at a 45° angle with respect to the microwave current direction. τ_{FL} : field-like torque; τ_{DL} : damping-like torque; J_C : charge current density; J_S : spin current density; H_{rf} : Oersted field induced by the microwave current. **b**, An example scan for ST-FMR measurements. The teal solid line represents the asymmetric component, while the blue solid line is a representation of the symmetric component. **c**, **d**, Angular dependence for the symmetric component and asymmetric component of the mixing voltage.

2.4.2 Second harmonic Hall measurements

ST-FMR is an effective technique that harnesses the spin orbit torque induced magnetization dynamics to identify the nature of the current induced torques and quantify their magnitudes. Besides ST-FMR (GHz frequency), an adiabatic (low frequency) harmonic Hall voltage measurement, which detects the change of the magnetization angle from its equilibrium value caused by spin orbit torque, can be employed to study the current induced effective fields[119]. For in-plane magnetized system (LSMO/SIO), a quasi-dc current is applied in the Hall device and the transverse second harmonic voltage is measured as a function of the angle of the applied in-plane magnetic field (Fig. 2.13a). The strength of the SOT from the SIO is characterized by measuring the second harmonic Hall voltage[119-121], which can be expressed by:

$$V_{xy}^{2\omega} = I_0 R_{PHE} \cos(2\Phi_M) \frac{[\tau_{FL} \cos(\Phi_M) + \tau_B]/\gamma}{H_{ext}}$$

$$-\frac{1}{2}I_0R_{\text{AHE}}\frac{\tau_{\text{DL}}\cos(\Phi_{\text{M}})/\gamma}{H_{\text{ext}} + \mu_0 M_{\text{eff}}} + V_{\text{ANE}}\cos(\Phi_{\text{M}}) + C, \quad (2.3)$$

where Φ_{M} is the angle between $\mathbf{M}$ and current flow direction, R_{PHE} and R_{AHE} represent the planar Hall and anomalous Hall resistance that appears as coefficients in the Hall resistance model ($R_{\text{H}} = R_{\text{PHE}}\sin(2\phi_{\text{M}})\sin^2(\theta_{\text{M}}) + R_{\text{AHE}}\cos(\theta_{\text{M}})$, where θ_{M} is the angle of $\mathbf{M}$ from z axis.), V_{ANE} is the anomalous Nernst voltage, M_{eff} is the effective magnetization, τ_{FL} is the out-of-plane field like (FL) torque, τ_{B} is the out-of-plane damping-like (DL) torque generated by spin polarization along the z -direction, τ_{DL} is the in-plane DL torque, and C is an angle-independent voltage offset. Fig. 2.13b shows the magnetic field angle-dependence of the second harmonic Hall signal fit to Eq. (2.3), which allows us to separate out the FL and DL contributions to the Hall signal. Separate curves depict the FL torque, combined in-plane DL torque and Nernst effect, and out-of-plane DL-torque in Fig. 2.13b. The $1/H_{\text{ext}}$ -dependence of the FL-Hall voltage is shown in Fig. 2.13d, with an expected linear trend given by Eq. (2.3). To separate the DL torque contribution to the $\cos(\Phi_{\text{M}})$ term from ANE contribution, $V_{\text{DL+ANE}}^{2\omega}$ is plotted as a function of $1/(H_{\text{ext}} + \mu_0 M_{\text{eff}})$ (Fig. 2.13c). Since the ANE contribution depends only on the magnetization and is independent of the magnitude of the external magnetic field as long as the magnet is saturated, the ANE signal is given by the y-axis intercept of the linear fit, while the contribution from DL torque τ_{DL} is extracted from the slope of the linear fit. Using the extracted τ_{DL} , the SOT efficiency is calculated by $\xi_{\parallel} = \frac{\tau_{\text{DL}}}{\gamma J c} \frac{M_{\text{stFM}}}{\hbar/2e}$ [122].

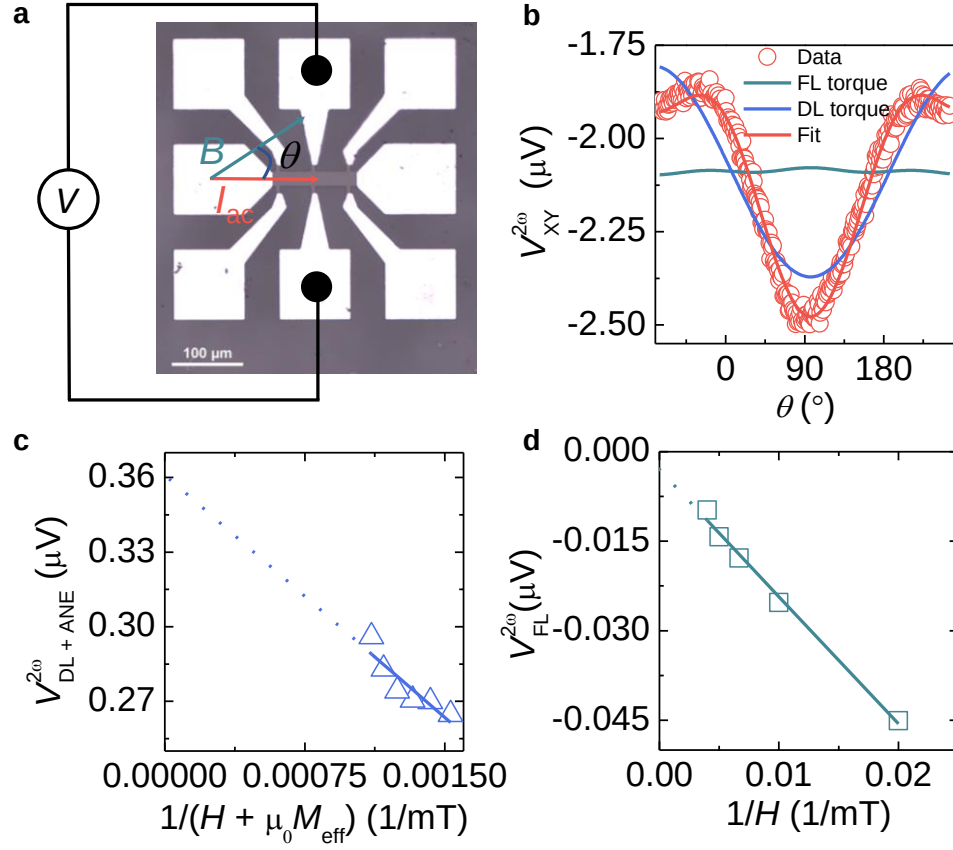


Figure 2.13: Second harmonic Hall measurements. **a**, A schematic illustration of the second harmonic Hall measurement geometry. The second harmonic Hall voltage is recorded while rotating the magnetic field in the plane. **b**, An example second harmonic Hall voltage as a function of magnetic field angle, with a magnetic field of 2500 Oe and a sample voltage of 4.8 V applied. The solid red curve is a fit to Eq. (2.3). The teal curve is the $\cos(2\Phi)\cos(\Phi)$ term (the field-like component). The blue curve is the $\cos(\Phi)$ term (the damping-like component + thermoelectric component). **c**, Dependence of the extracted damping-like and thermal electric voltage component. The blue curve is a fit to the data using Eq. (2.3). **d**, Dependence of the extracted field-like voltage component. The solid curve is a fit to the data using Eq. (2.3).

Chapter 3

Novel spin-orbit torque generation at room temperature in all oxide epitaxial $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{SrIrO}_3$ system

The efficient spin orbit torque generation lies at the center of spintronics research for next generation spintronics-based memory and logic devices. SOTs in conventional heavy metals and topological insulators have been explored extensively, while 5d transition metal oxides, which also host ions with strong spin-orbit coupling, are a relatively new territory in the field of spintronics. An all-oxide, SrTiO_3 (STO)// $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO)/ SrIrO_3 (SIO) heterostructure with lattice-matched crystal structure has been synthesized, exhibiting an epitaxial and atomically-sharp interface between the ferromagnetic LSMO and the high spin-orbit-coupled metal SIO. I use spin-torque ferromagnetic resonance (ST-FMR) to probe the effective magnetization and the SOT efficiency in LSMO/SIO heterostructures grown on STO substrates. Remarkably, epitaxial LSMO/SIO exhibits a large SOT efficiency, $\xi_{||} = 1$, while retaining a reasonably low shunting factor and increasing the effective magnetization of LSMO by $\sim 50\%$. Our findings highlight the significance of epitaxy as a powerful tool to achieve a high SOT efficiency, explore the rich physics at the epitaxial interface, and open up a new pathway for designing next-generation energy-efficient spintronic devices.

3.1 Background and introduction

Efficient electrical manipulation of magnetism is an ongoing pursuit for applications in next-generation information storage and logic functions. One promising pathway is to utilize current-induced spin torques originating from spin-orbit coupling (SOC)[123, 124], such as the bulk spin Hall effect (SHE)[34, 56, 125] or the interfacial Rashba-Edelstein effect (REE)[9, 126], where a charge current passed through a SOC metal generates a spin torque on an adjacent ferromagnet (FM). The key to realizing energy-efficient spin-orbit torque (SOT)-based switching of FM's is to maximize the anti-damping SOT efficiency (per unit current density) $\xi_{||} = \theta_{||} T_{\text{int}}$, where $\theta_{||}$ is the spin Hall ratio and T_{int} is an interfacial spin transparency factor. Consequently, there is tremendous on-going research focused on achieving such large SOT efficiencies by using materials with large SOC, such as elemental heavy metals (HM)[34, 56, 127-129], topological insulators (TI)[54, 55, 59, 130], with spin momentum locked interface states, and two-dimensional electron gases (2DEG) at oxide interfaces[4, 131, 132]. However, in HM's SOT efficiencies are typically below 0.5[133, 134], and in TI/FM and 2DEG systems the high resistivity of the spin Hall material causes current shunting through the magnet that compromises the overall efficiency per unit applied current[60, 62]. Here, I show that model epitaxial all-oxide heterostructures can achieve SOT efficiencies on par with some TI materials at room temperature[63], while allowing lower-resistivity SOC layers and high-resistivity oxide FMs that together decrease unwanted current shunting compared to TI or 2DEG/metallic FM structures. Moreover, I find an unusual dependence of the oxide FM magnetic anisotropy on the thickness of the oxide SOC material, highlighting the rich physics in correlated oxide spintronics systems.

Complex oxides, displaying a strong interplay between charge, spin, orbital, and lattice degrees of freedom, offer a scientifically-rich platform for both fundamental and application-oriented research. Their properties range from high-temperature superconductivity in cuprates, colossal magnetoresistance in doped manganites, and more recently, exotic band structure effects in perovskite iridates. Moreover, the sensitivity of complex oxides to epitaxial strain[135], interface chemistry, and crystal orientation provides additional degrees of freedom in tuning the electronic and magnetic structure and SOC effects. In particular, 5d transition metal oxides (TMO), with strong SOC and electron-electron correlation, give rise to intriguing fundamental physics, ranging from nontrivial quantum phases[136-138] to magnetic anisotropy manipulation[139] and intrinsic charge-spin interconversion[140-142]. However, despite this rich physics, in the few room temperature studies on charge-spin interconversion in 5d TMOs, the adjacent FM metal has typically been a non-epitaxial metal, such as SrIrO₃ (SIO)/Permalloy (Py)[140, 141], SIO/Co_{1-x}Tb_x [142] and SrRuO₃/Co[143]. The resulting SOT efficiencies, $\xi_{||} \leq 1$ [140-143], are comparable to their HM/FM counterparts. With this as the background, our hypothesis was that creating atomically perfect ferromagnet and SOC metal layers, coupled through chemically abrupt epitaxial interfaces, can lead to emergent electronic phenomena at the interfaces[139], as well as the opportunity to obtain more efficient charge-to-spin conversion. For our studies, we chose the correlated perovskite

manganite, $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) and the SOC metal perovskite iridate, SrIrO_3 (SIO), as model systems.

3.2 Structural and electronic study on $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{SrIrO}_3$ heterostructure

3.2.1 Structural characterization

$2\theta - \omega$ x-ray diffraction (XRD) scan for STO//LSMO (50 UC)/SIO (25 UC) is shown in Fig. 3.1a, with clear LSMO (0 0 2) and SIO (0 0 2) peaks, showing that only single phase LSMO and SIO are present. RSMs around (0 0 3), (1 0 3), (2 0 3) diffraction peaks are shown in Fig. 3.1b, c, d, which indicate that LSMO and SIO are coherently strained to STO substrate.

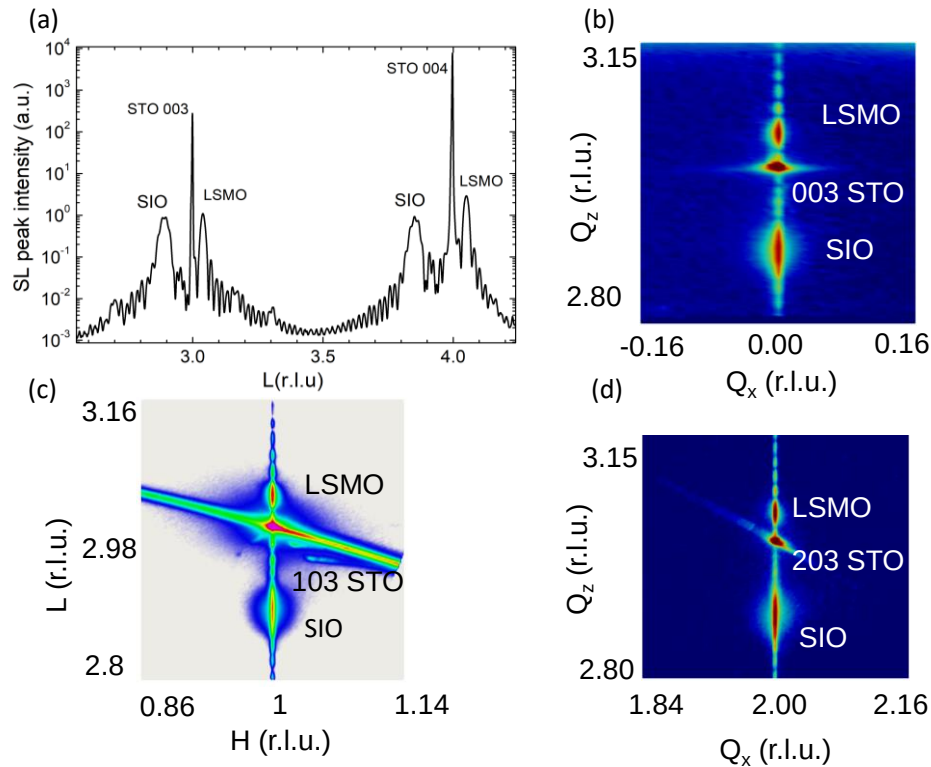


Figure 3.1: XRD and RSM for STO//LSMO (50 UC)/SIO (25 UC). a, $2\theta - \omega$ scan. b, RSM for STO (0 0 3) peak. c, RSM for STO (1 0 3) peak. d, RSM for STO (2 0 3) peak.

Half-ordering RSM peak around (0.5 0.5 1.5) for STO//LSMO (50 UC)/SIO (25 UC) is shown in Fig. 3.2a. RSMs of the STO//LSMO (50 UC)/SIO (25 UC) around (1 0.5 1.5), (0.5 1 1.5) and (0.5 1.5 1) diffraction peaks are shown in Fig. 3.2b, c, d, showing that the SIO layer presents domains with in-phase octahedral rotation axis aligned with both X and

Y directions. No domains with in-phase octahedral rotation axis along Z are detected and in-phase octahedral rotations in the LSMO layer are not detected either.

To examine the quality of the oxide interface at the microscopic level, I performed high angle annular dark field (HAADF) scanning transmission electron microscopy (STEM) experiments (Fig. 3.4a). The brighter contrast is from the SIO layer due to its higher Z_{eff} whereas the darker contrast is from the LSMO layer. The sharp interface quality is evident from Fig. 3.4(a) with MnO_2 termination in LSMO and SrO termination in SrIrO_3 .

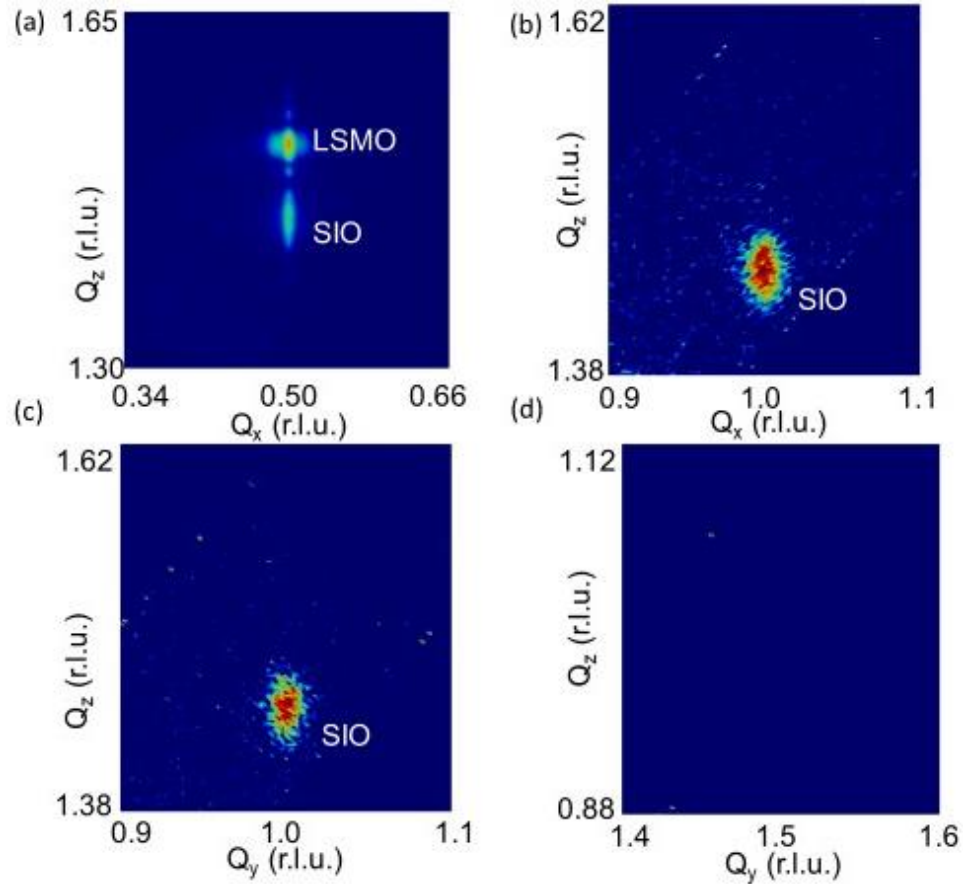


Figure 3.2: Half ordering RSM peaks for STO//LSMO (50 UC)/SIO (25 UC). a, RSM for STO (0.5 0.5 1.5) peak. b, RSM for STO (1 0.5 1.5) peak. c, RSM for STO (0.5 1 1.5) peak. d, RSM for STO (0.5 1.5 1) peak.

Fig. 3.3a shows X-ray diffraction patterns of LSMO(50 UC)/SIO(X UC), where X=10, 20 and 30 unit cells. The corresponding OOP lattice parameters are presented in the table in Fig. 3.3b, along with the in-plane and OOP lattice parameter data extracted from the HAADF-STEM images of the 10 and 30UC SIO samples.

There are three key observations from this data. First, from the XRD data in Fig. 3.3a, the LSMO OOP lattice parameter is essentially unchanged; second, the OOP lattice parameters of the SIO layer systematically change as a function of the SIO layer thickness, from 3.97 Å for the 10 UC sample to 4.06 Å for the 30 UC SIO. We note that the SIO

OOP lattice parameter (3.97 ± 0.02 Å) in the LSMO(50 UC)/SIO(10 UC) is smaller than what has been reported for 30 UC SIO grown on STO substrate (4.08 Å)[144]. Finally, most importantly, the OOP lattice parameter of the 30UC SIO on 50UC LSMO is essentially identical to that reported for the 30UC SIO on STO substrate[144]. Thus, pure relaxation of heteroepitaxial constraint imposed by the lattice mismatch between the in-plane dimensions of LSMO and SIO fails to explain the experimentally observed trend in OOP lattice dimensions in the SIO layer (i.e., the OOP lattice parameter of the SIO layer is changing counter to what would be expected from pure heteroepitaxy considerations). Fig. 3.3c-f capture the structural details of the STO/SIO(30 UC), STO/LSMO(50 UC) and the corresponding LSMO/SIO heterostructures with 10 and 30 UC of SIO.

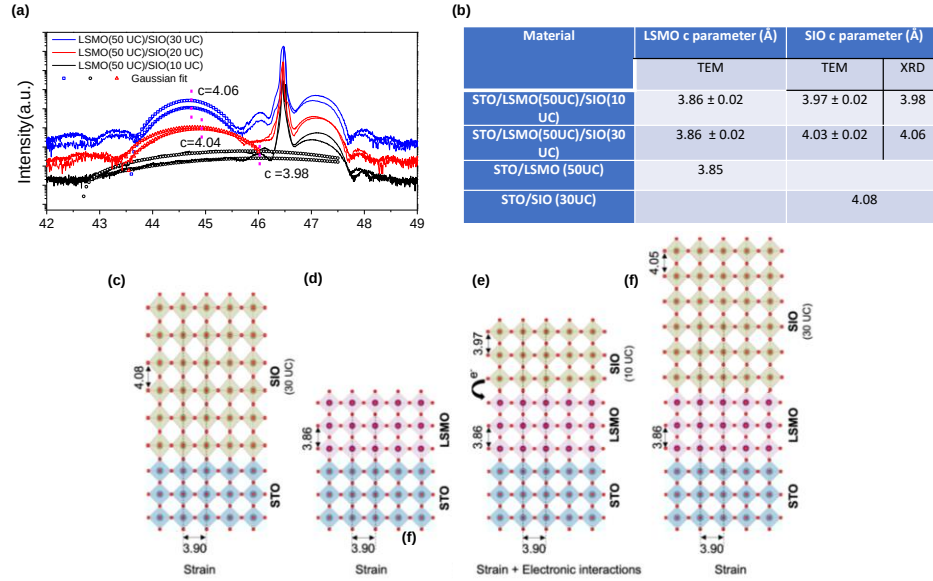


Figure 3.3: Lattice constants for LSMO/SIO systems. a, XRD line scans for STO/LSMO (50 UC)/SIO (10 UC), STO/LSMO (50 UC)/SIO (20 UC) and STO/LSMO (50UC)/SIO (30UC). b, A table of the lattice constants of LSMO and SIO obtained from TEM and XRD. The c parameter for 30-UC SIO on STO substrate is reported in Ref.[144] and the c parameter for 50-UC LSMO on STO substrate is reported in Ref.[145]. c, d, e, f, Structural schematics and corresponding lattice constants for STO/SIO (30 UC), STO/LSMO (50 UC), STO/LSMO (50 UC)/SIO (10 UC) and STO/LSMO (50 UC)/SIO (30 UC).

3.2.2 Electronic structure characterization

Therefore, in order to throw more light onto this puzzling structural evolution with SIO thickness, we performed atomic resolution electron-energy loss spectroscopy (EELS) measurements, which probes the empty density of states as presented in Fig. 3.4d. We observe a negative shift of ~1eV for the Mn L-edge in the 10UC SIO sample compared to the 30UC SIO sample (as well as the standard reference spectra[146] shown in Fig. 3.4d). These changes in the Mn L-edge point to the strong possibility of a charge transfer from

Ir to Mn[147-149] shown in Fig. 3.4b. We also observe a corresponding change in the O K-edge in the SIO layer (we note that we do not access the Ir L-edge, which appears at $\sim 12\text{KeV}$, and is not accessible in our EELS spectrometer). Since the EELS spectra essentially probes the empty density of states, the changes in the first 5eV of the O K-edge reflects changes in the electronic structure of the SIO layer, arising from changes in the Ir-O hybridization. Such a charge transfer can also be accompanied by orbital reconstruction in the SIO layer[139, 147]. Bulk SIO is a semimetal with strong spin-orbit coupling (SOC), which is comparable to the crystal field splitting (CFS)[150]. It has been recently reported that CFS can dominate over SOC in strained iridate systems[151]. An evidence for such an orbital re-construction due to CFS can be seen in O K edge in Fig. 3.4d. In LSMO (50 UC)/SIO (10 UC), the density of states for Ir-O related levels are broader than that those in LSMO (50 UC)/SIO (30 UC). We believe that due to the dominance of CFS over the SOC, the $S=1$ state is stabilized in the LSMO (50 UC)/SIO (10 UC) as is shown in Fig. 3.4c. Our present results agree with previously reported results on $\text{SrIrO}_3/\text{LaNiO}_3$ system[147]. We note that for LSMO (50 UC)/SIO (30 UC), where the charge transfer is negligible, there is no orbital reconstruction in Ir 5d levels shown in Fig. 3.4c.

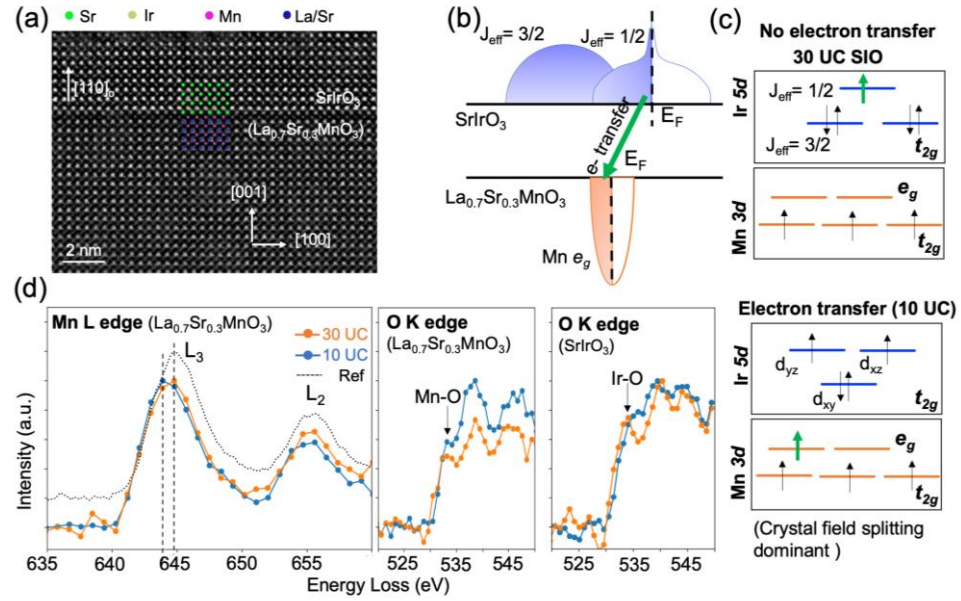


Figure 3.4: Local crystal and electronic structure of LSMO/SIO interface. **a**, High angle annular dark field (HAADF) image of LSMO/SIO layer displaying the atomically clean interface. **b**, The electronic arrangement of LSMO/SIO showing the electron charge transfer from Ir^{4+} (SrIrO_3) to Mn^{4+} ($\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$). **c**, Orbital reconstruction of Mn 3d and Ir 5d levels for LSMO (50 UC)/SIO (10 UC) after the electron transfer. **d**, Mn L edge and O K edge EELS spectrum extracted from the LSMO/SIO interface for different thicknesses.

3.3 Magnetism in the $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{SrIrO}_3$ heterostructure

3.3.1 Experimental observation of effective magnetization for $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{SrIrO}_3$ heterostructure

The magnetism of the heterostructure (LSMO (50 UC)/SIO (5 UC)) was checked with vibrating sample magnetometry (VSM) at room temperature as shown in Fig. 3.5a. The saturation magnetization was determined to be ~ 300 emu/cc and the magnetic coercive field is ~ 25 Oe, confirming a good ferromagnetism of the heterostructure. An effective magnetization around 448 ± 7 kA/m is obtained from Fig. 3.5b.

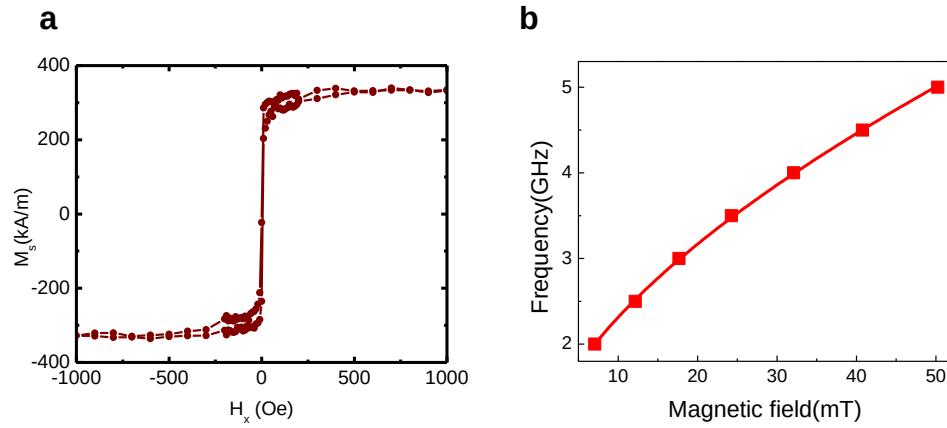


Figure 3.5: Magnetic characterization for LSMO/SIO heterostructures. a, In-plane magnetization hysteresis loop (M-H) measurement at 300 K for LSMO (50 UC)/SIO (5 UC). b, ST-FMR measurement for sample LSMO (50 UC)/SIO (21 UC) at 18dBm. Frequency as a function of magnetic field. The red line indicates a fit to Eq. (2) in main text.

To understand the potential spintronic manifestations of the clean epitaxial LSMO/SIO interface and the resulting charge transfer and orbital reconstruction, ST-FMR measurements were performed on LSMO/SIO heterostructures with varying thicknesses. Fig. 3.6a shows an example ST-FMR measurement for LSMO (50 UC)/SIO (22 UC) at a frequency of 4 GHz and input power 18 dBm, with an external magnetic field oriented 45° to the current direction. The measured ST-FMR signal fits well to Eq. (2.1), yielding the symmetric and anti-symmetric components. The resonance fields are summarized in Fig. 3.5b, and agree well with the Kittel equation:

$$f = \frac{\gamma}{2\pi} \sqrt{(\mu_0 H_{\text{ext}} + \mu_0 H_k)(\mu_0 H_{\text{ext}} + \mu_0 H_k + \mu_0 M_{\text{eff}})}, \quad (3.1)$$

where H_k is the in-plane magnetic field anisotropy. From the fit to Eq. (3.1), M_{eff} is extracted, showing a strong dependence on SIO thickness. As is shown in Fig. 3.6b, a $\sim 50\%$ enhancement of effective magnetization compared to the bulk saturation magnetization is seen for LSMO (50 UC)/SIO (5 UC) (Fig. 3.5a). The saturation magnetization M_s as measured by vibrating sample magnetometry does not show any such abrupt dependence

on the SIO thickness (Fig. 3.6b), indicating that the enhancement of M_{eff} is likely due to a change in magnetic anisotropy. The enhancement in M_{eff} is observed only for SIO thinner than ~ 25 UC and progressively decreases for the thicker SIO samples. The enhancement of M_{eff} is observed for all LSMO thicknesses, confirming the universal role of SIO in enhancing the M_{eff} of LSMO. The effective magnetization can be written as $\mu_0 M_{\text{eff}} = \mu_0 M_s - H_{k,\text{eff}}$ [152], where $H_{k,\text{eff}}$ is the effective anisotropy field. This enhancement in M_{eff} in the epitaxial LSMO/SIO is quite different from the SIO/CoFe systems, see Fig. 3.6c, which exhibits $M_{\text{eff}} < M_s$, and no strong dependence on SIO thickness, as typically seen in conventional metallic systems. This could be due to a difference in the sign of $H_{k,\text{eff}}$ between the epitaxial LSMO/SIO and non-epitaxial SIO/Py, which further accentuates the peculiarity of the LSMO/SIO system. In conventional systems, $H_{k,\text{eff}}$ can have contributions from a bulk component ($2K_u/M_s$) and an interface component ($2K_{\text{int}}/M_s t_{\text{LSMO}}$) [152], where K_u and K_{int} are the bulk and interfacial anisotropy constants, respectively. The difference between M_{eff} and M_s in the LSMO/SIO scales linearly with $1/M_s t_{\text{LSMO}}$, shown in the inset of Fig. 3.6d, as expected from an interfacial contribution. ($M_{\text{eff}} - M_s$) vs $1/M_s t_{\text{LSMO}}$ shows a large intercept, suggesting a stronger contribution from bulk anisotropy; however, the trend suggests that the bulk and the interfacial anisotropies have opposite signs.

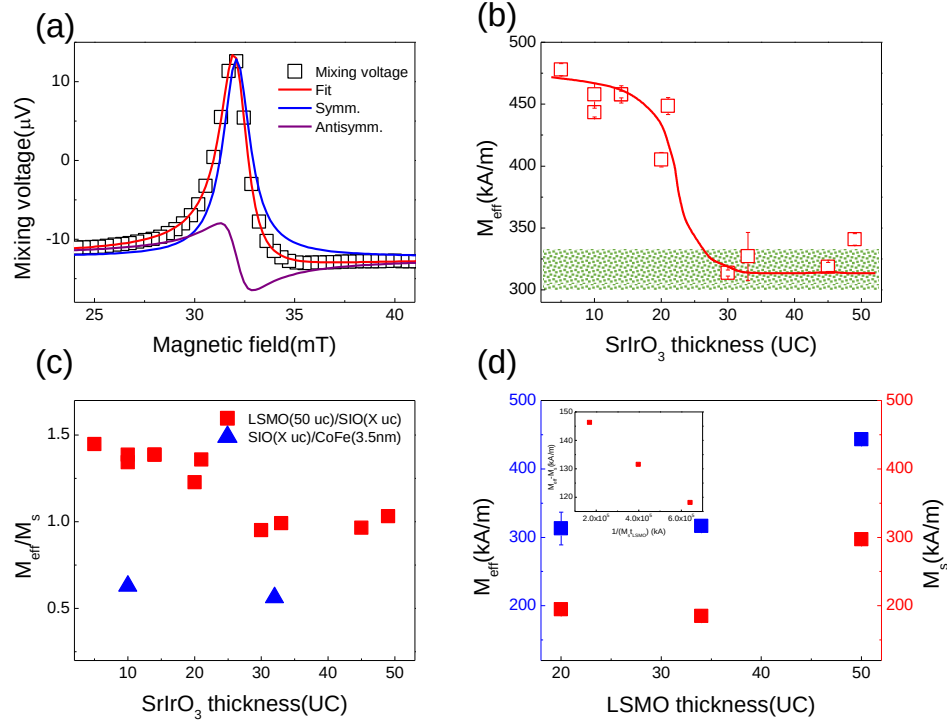


Figure 3.6: ST-FMR and Effective Magnetization for LSMO/SIO bilayer. **a**, ST-FMR for sample LSMO (50 UC)/SIO (22 UC) at 4GHz. The external magnetic field is oriented at 45° with respect to the current direction. Open black squares indicate spin mixing voltage. The red line is a fit to the spin mixing voltage data while the blue and purple curves indicate extracted symmetric and anti-symmetric spin mixing

voltages, respectively. **b**, Effective magnetization, M_{eff} , extracted from ST-FMR measurements, for LSMO (50 UC)/SIO (X UC) as a function of SIO thickness. The green dotted band indicates the saturation magnetization, M_s (ranging from 300 to 330 kA/m), for 50 UC LSMO measured using vibrating sample magnetometry. The red solid line is a guide to illustrate the thickness dependent M_{eff} transition. **c**, The ratio of M_{eff}/M_s as a function of SIO thickness. The red solid squares and blue solid triangles indicate LSMO (50 UC)/SIO (X UC), SIO (X UC)/CoFe (3.5 nm)/Al systems, respectively. **d**, M_{eff} and M_s for LSMO (X UC)/SIO (10 UC) as a function of LSMO thickness. The blue and red solid squares indicate M_{eff} and M_s , respectively. The inset shows $M_{\text{eff}} - M_s$ as a function of $1/(M_s t_{\text{LSMO}})$.

3.3.2 *ab initio* study of effective magnetization

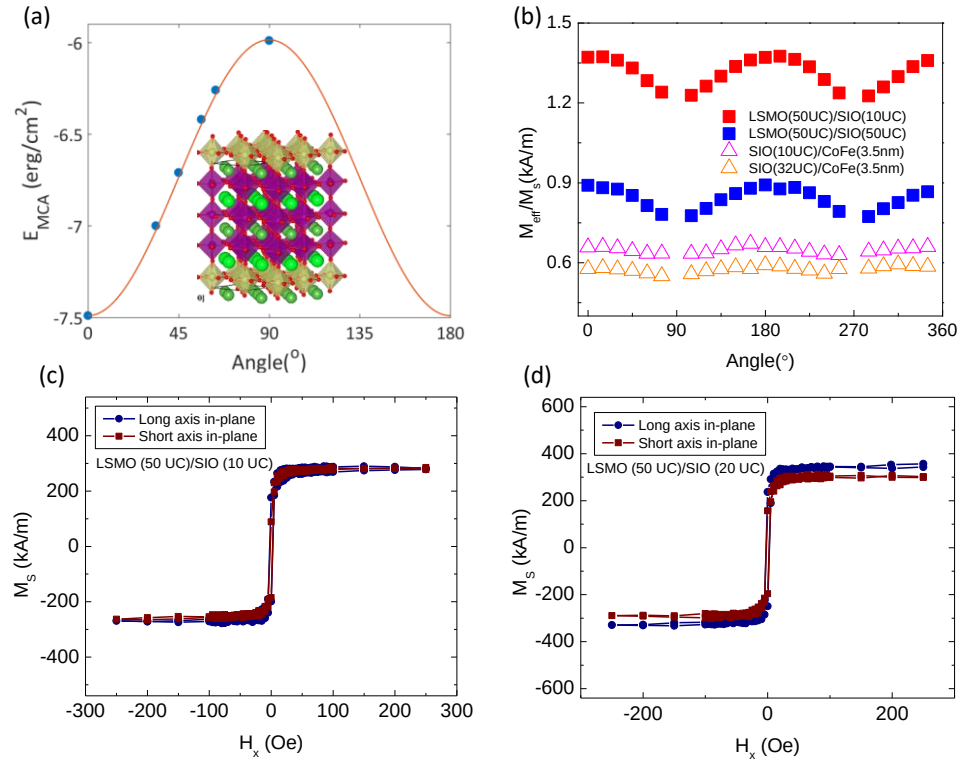


Figure 3.7: **a**, Angular dependence of the *ab initio* calculated MCA energy, $E_{\text{MCA}} = E(\theta = 90^\circ, \phi) - E(0^\circ, 0^\circ)$, per unit area for the LSMO (3 UC)/SIO (1 UC) superlattice, whose crystal structure is shown in the inset. The solid circles denote the *ab initio* MCA values and the red curve denotes the MCA calculated from the expression, $E_{\text{MCA}} = K_{xx}\sin^2(\phi) + K_{yy}\cos^2(\phi)$, where $K_{xx} = -6 \text{ erg/cm}^2$ and $K_{yy} = -7.5 \text{ erg/cm}^2$, are the *ab initio* calculated MCA values along the [100] and [010] directions, respectively. **b**, M_{eff}/M_s as a function of magnetic field angle. The red and blue solid squares represent LSMO (50 UC)/SIO (10 UC) ($M_s=300 \text{ kA/m}$) and LSMO (50 UC)/SIO (50 UC) ($M_s=300 \text{ kA/m}$), respectively. The magenta and orange solid triangles indicate SIO (10 UC)/CoFe (3.5 nm) ($M_s=1110 \text{ kA/m}$) and SIO (32 UC)/

CoFe (3.5 nm) ($M_s=1220$ kA/m), respectively. **c**, Vibrating sample magnetometry (VSM) for sample LSMO (50 UC)/SIO (10 UC). Wine red solid squares and navy blue solid triangles indicate $H_x||$ long axis and $H_x||$ short axis, respectively. The ratio between long axis and short axis is 2:1. **d**, Vibrating sample magnetometry (VSM) for sample LSMO (50 UC)/SIO (20 UC). Wine red solid squares and navy blue solid triangles indicate $H_x||$ long axis and $H_x||$ short axis, respectively. The ratio between the long axis and short axis is 2:1.

The angular variation of M_{eff} is displayed in Fig. 3.7b along with the experimentally determined value of the saturation magnetization, M_s , can be used to determine the MCA value, K_{MCA} , of the system, which is given by[153]

$$K_{\text{MCA}} = -2\pi M_s M_{\text{eff}} + 2\pi M_s^2 \quad \text{or} \quad \frac{M_{\text{eff}}}{M_s} = 1 - \frac{K_{\text{MCA}}}{2\pi M_s^2}, \quad (3.2)$$

where $K_{\text{MCA}} = K_V + \frac{K_i}{t_{\text{LSMO}}}$ includes the strain-induced bulk LSMO, K_V , and the LSMO/SIO interface, K_i (t_{LSMO} is the LSMO thickness) contributions, respectively. The angular dependence of M_{eff} , shown in Fig. 3.7b for two SIO thicknesses, can be attributed to the orthorhombic crystal structure of the SIO which results in an anisotropic in-plane MCA of the LSMO/SIO bilayer. The larger (smaller) value of M_{eff} relative to M_s denoted with red (blue) symbols in Fig. 3.7b suggests an in-plane (out-of-plane) MCA with negative (positive) K_{MCA} . Using the experimentally determined angular variation of $\frac{M_{\text{eff}}}{M_s}$ in Fig. 3.7b and the above expression we find that (i) at $\phi = 0^\circ$ $t_{\text{LSMO}}K_{\text{MCA}}$ is -0.4 erg/cm² and -0.15 erg/cm² for the 10 UC and 50 UC SIO thickness, respectively; and (ii) the amplitude of oscillation of $t_{\text{LSMO}}K_{\text{MCA}}$ (where $t_{\text{LSMO}} = 20$ nm) is 0.25 and 0.15 erg/cm² for the 10 UC (red symbols) and 50 UC (blue symbols), respectively.

Earlier theoretical and experimental results suggest a negligible bulk MCA at zero strain[154]. This in turn suggests that the origin of MCA lies in either the interfacial MCA, K_i , or the strain-induced bulk MCA. In order to clarify the first mechanism, we have carried out ab initio calculations of the zero-temperature MCA energy for the SIO (1 ML)/LSMO (3 ML) superlattice crystal structure shown in the inset of Fig. 3.7a. Fig. 3.7a shows the azimuthal angular variation of the MCA energy, $E_{\text{MCA}} = E(\theta = 90^\circ, \phi) - E(0^\circ, 0^\circ)$, per unit area for the LSMO (3 UC)/SIO (1 UC) superlattice. The calculated zero-temperature values of K_i is -7.5 erg/cm² at $\phi = 0$ and the amplitude of oscillation is 1.5 erg/cm². However, in order to compare with experiment one needs to take into account the temperature dependence of K_i given by[155]:

$$\frac{K_i(T)}{K_i(0)} = \left(\frac{M_s(T)}{M_s(0)} \right)^3 = 0.2, \quad (3.3)$$

Hence, the room-temperature values of K_i and the amplitude of oscillation are -1.5 erg/cm² and 0.3 erg/cm², respectively, which are $\sim$ a factor of four larger than the corresponding experimental values. The discrepancy may be due to the ultrathin LSMO and SIO thickness in the calculations compared to experiment and also the emergence of magnetism in the SIO component. A second plausible mechanism for the enhancement of M_{eff} relative to M_s is the magnetoelastic effect of bulk LSMO associated with the lattice constant mismatch between LSMO and orthorhombic SIO (tensile strain of $< 2\%$ on

LSMO along the a-direction and <1.5% along the b-direction) which in turn gives rise to an in-plane uniaxial MCA[154].

3.4 Spin-orbit torque efficiency in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{SrIrO}_3$ heterostructure

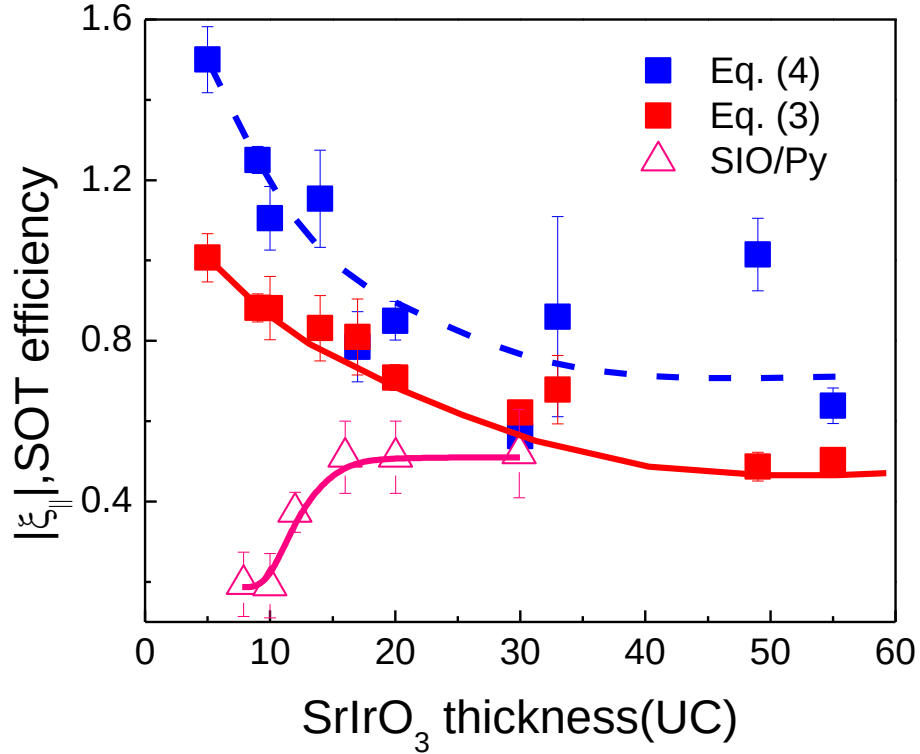


Figure 3.8: SOT Efficiency for LSMO/SIO. SOT efficiency as a function of SIO thickness is shown, with LSMO thickness fixed at 50 UC. The red solid squares represent SOT efficiency estimated by Eq. (2.2) for LSMO/SIO system. The red line is a guide to the eye. The blue solid squares indicate SOT efficiency for LSMO/SIO

estimated by the modified Eq. (2.2)- $\xi_{||} = \frac{s}{A} \left(\frac{e}{h} \right) \mu_0 M_{\text{eff}} t_{\text{FM}} d_{\text{NM}} \sqrt{1 + \mu_0 M_{\text{eff}} / H_0}$ (3.4).

The blue dashed line is a guide to the eye. The pink open triangles indicate SOT efficiency for SIO/Py[140]. The pink solid line is a guide for SIO/Py.

I now bring all the observed enhancements in M_{eff} to focus by calculating the charge to spin conversion efficiency (SOT efficiency) from the ST-FMR measurements. Fig. 3.8 summarizes the SOT efficiencies calculated with Eq. (2.2) for LSMO/SIO as a function of SIO thickness, in which a strong dependence of SOT efficiency on SIO thickness is observed. As the SIO thickness increases, the SOT efficiency decays, in direct contrast to its non-epitaxial counterpart SIO/Py[140], whose SOT efficiency increases with SIO thicknesses and eventually saturates at ~0.5. The SOT efficiency saturation with SOC

metal thickness is commonly attributed to spin diffusion length in the SOC material[156]. This contrasting thickness dependency suggests a distinctly different physics underlying the spin-charge conversion in these systems, resulting from the epitaxial interface. Intriguingly, unlike traditional heavy-metal systems, M_{eff} is always equal to or larger than M_s in LSMO/SIO heterostructures, which brings into question the use of M_s in Eq. (2.2). Therefore, as a comparison, I have overlayed data in Fig. 3.8 (blue solid squares) where I have calculated $\xi_{\parallel}$ using M_{eff} in the pre-factor rather than M_s . In order to shed lights on the origin of the SOT efficiency enhancement, further analyses on the effective interface spin mixing conductance and interface transparency were carried out.

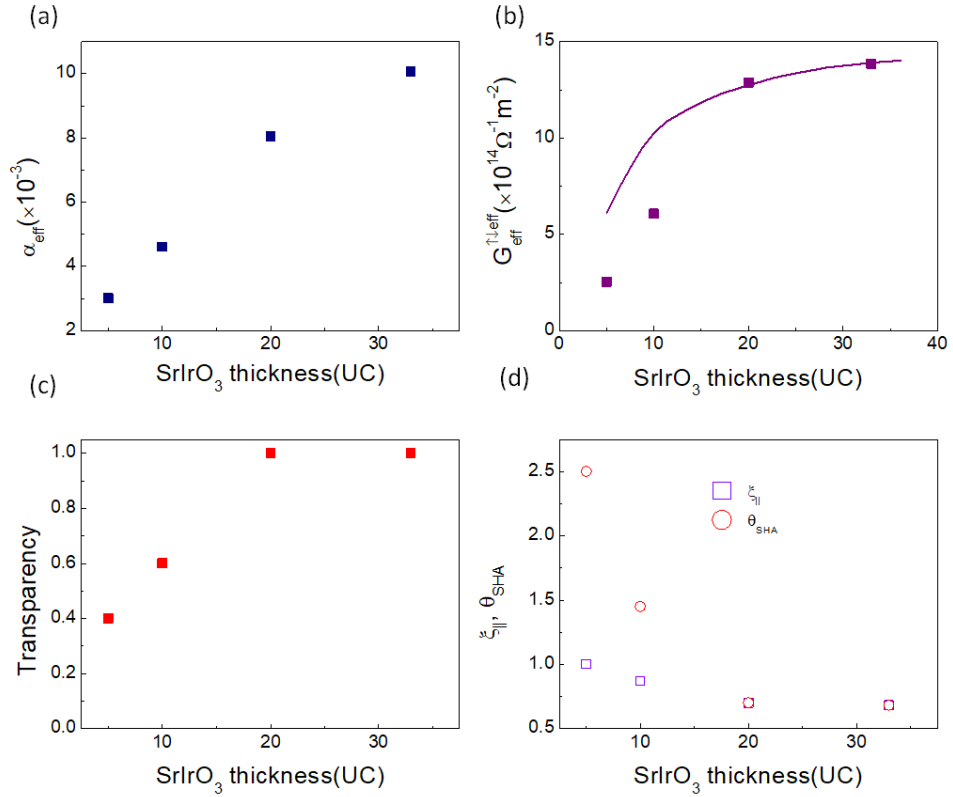


Figure 3.9: ST-FMR linewidth analysis. a, Effective Gilbert damping constant, α as a function of SIO thickness. b, Effective interface spin mixing conductance, $G_{\text{eff}}^{\uparrow\downarrow}$ as a function of SIO thickness; the purple line is a fit to the data. c, Interface transparency, T as a function of SIO thickness. d, SOT efficiency, $\xi_{\parallel}$ and spin Hall angle, θ_{SHA} as a function of SIO thickness. The violet open squares and pink open circles are the SOT efficiency and spin Hall angle, respectively.

In this section, I disentangle the interface transparency (T) and the internal spin Hall angle (θ_{SH}) from the measured spin-orbit torque (SOT) efficiency ($\xi_{\parallel}$), using the conventional semiclassical model[157, 158].

Interface spin mixing conductance: We start by estimating the real part of the effective spin mixing conductance ($G_{eff}^{\uparrow\downarrow} = \frac{q^2}{h} g_{eff}^{\uparrow\downarrow}$) using

$$g_{eff}^{\uparrow\downarrow} = \frac{4\pi M_s t_{LSMO}}{g \mu_B} (\alpha - \alpha_0) \quad (3.5),$$

where α is the effective Gilbert damping of the bilayer, which has been calculated using the FMR linewidth broadening $\Delta H = \Delta_0 + \frac{4\pi}{\sqrt{3}\gamma} \alpha f$. Here M_s , t_{LSMO} and α_0 are the saturation magnetization, thickness and intrinsic Gilbert damping of LSMO, respectively. g is the Landé factor, μ_B is the Bohr magneton, Δ_0 is the extrinsic linewidth contribution, γ is the gyromagnetic ratio, and f is the FMR frequency. The estimated $G_{eff}^{\uparrow\downarrow}$ is shown as data points in Fig. 3.9b.

$G_{eff}^{\uparrow\downarrow}$ is related to the bare spin mixing conductance ($G^{\uparrow\downarrow}$) and the source conductance of the spin Hall generator (G_{so})[159] as:

$$G_{eff}^{\uparrow\downarrow} = \frac{G^{\uparrow\downarrow} G_{so}}{G^{\uparrow\downarrow} + G_{so}} \quad (3.6),$$

where $G^{\uparrow\downarrow}$ is the bare spin mixing conductance, and G_{so} is given by[159]

$$G_{so} = \frac{\tanh\left(\frac{t_{SIO}}{\lambda_{sf}}\right)}{2\rho\lambda_{sf}} \quad (3.7),$$

Here, ρ is the resistivity of the sample which agrees with the data from[140]. We fit the data in Fig. 3.9b to Eq. (3.7) to estimate λ_{sf} .

The interface transparency factor: T is calculated using the following expression[159]:

$$T = \frac{G_{eff}^{\uparrow\downarrow}}{G_{so}} \tanh\left(\frac{t_{SIO}}{2\lambda_{sf}}\right) \tanh\left(\frac{t_{SIO}}{\lambda_{sf}}\right) = 2\rho\lambda_{sf} G_{eff}^{\uparrow\downarrow} \tanh\left(\frac{t_{SIO}}{2\lambda_{sf}}\right) \quad (3.8),$$

which we have shown in Fig. 3.9c. T is $\sim 40\%$ for thin samples (5 uc) which gradually increases and saturates to $\sim 100\%$ for thicker samples (≥ 20 uc). We extract the θ_{SH} from the measured $\xi_{||}$ using the following expression[159]:

$$\xi_{||} = T\theta_{SH} \quad (3.9),$$

Note that the enhancement we observe in $\xi_{||}$ for thinner samples is therefore not due to an enhancement in the interface transparency, T . Therefore, the enhancement in $\xi_{||}$ observed in the thin SIO regime corresponds to a substantial enhancement in θ_{SH} , as shown in Fig. 3.9d, suggesting that the enhancement of SOT efficiency is not simply due to the improvement of interface quality but is a result of the intrinsic electronic interaction (charge transfer + orbital reconstruction) between LSMO and SIO, supported by EELS mapping.

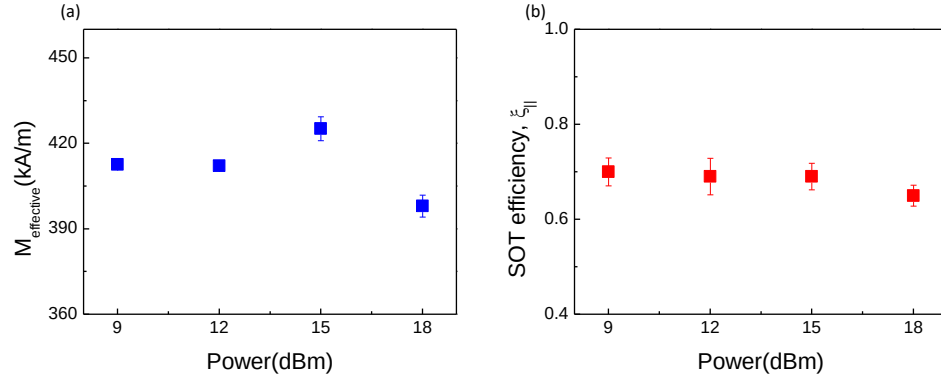


Figure 3.10: Power dependent ST-FMR measurement for LSMO (50 UC)/SIO (21 UC). a, Effective magnetization as a function of applied microwave power. b, SOT efficiency as a function of applied microwave power.

Effective magnetization as a function of applied microwave power is shown in Fig. 3.10a, showing negligible dependence on microwave power. Negligible dependence on applied microwave power for SOT efficiency (Fig. 3.10b) is seen, ruling out any heating effect that might cause spurious effects in ST-FMR.

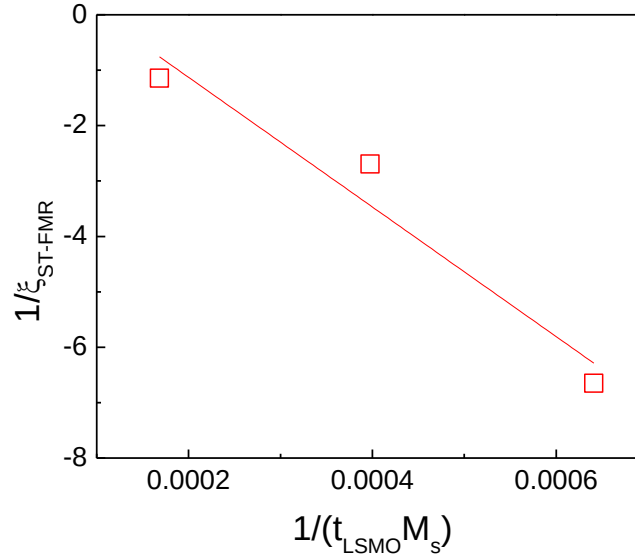


Figure 3.11: Field-like torque evaluation. $1/\xi_{\text{ST-FMR}}$ as a function of $1/(t_{\text{LSMO}} M_s)$ for sample LSMO (X UC)/SIO (10 UC). The thicknesses of LSMO shown in the figure are 20, 34 and 50 UC.

Eq. (2.2) is based on the assumption that the field-like torque is negligible in the estimation, which introduces some uncertainty in SOT efficiency estimation. To separate out the contribution from damping-like torque and field-like torque, a LSMO thickness series study was carried out. $1/\xi_{\text{ST-FMR}}$ as a function of $1/(t_{\text{LSMO}} M_s)$ is shown in Fig. 3.11. $1/\xi_{\text{ST-FMR}}$ can be written as[160]:

$$\frac{1}{\xi_{\text{ST-FMR}}} = \frac{1}{\xi_{\text{DL}}} \left(1 + \frac{\hbar}{e \mu_0 M_s t_{\text{LSMO}} t_{\text{SIO}}} \xi_{\text{FL}} \right), \quad (3.10)$$

where $\xi_{\text{ST-FMR}}$ is the SOT efficiency estimated by Eq. (2.2) in the main text, ξ_{DL} is the SOT efficiency from damping-like SOT, ξ_{FL} is the SOT efficiency from field-like SOT, t_{LSMO} is the thickness of LSMO and t_{SIO} is the thickness of SIO. A linear fit of $1/\xi_{\text{ST-FMR}}$ vs $1/(t_{\text{LSMO}} M_s)$ can then be used to determine $1/\xi_{\text{DL}}$ (from the intercept) and ξ_{FL} (from the slope). Since ξ_{DL} is large in our system, extracting $1/\xi_{\text{DL}}$ from the intercept may introduce an unnecessary degree of uncertainty. Therefore, we use the slope to determine the ratio for $\xi_{\text{FL}}/\xi_{\text{DL}}$. From the linear fit in Fig. 3.11, a ratio for $\xi_{\text{FL}}/\xi_{\text{DL}}$ of ~ -0.09 was determined, indicating that field-like torque is present in LSMO/SIO system but negligible.

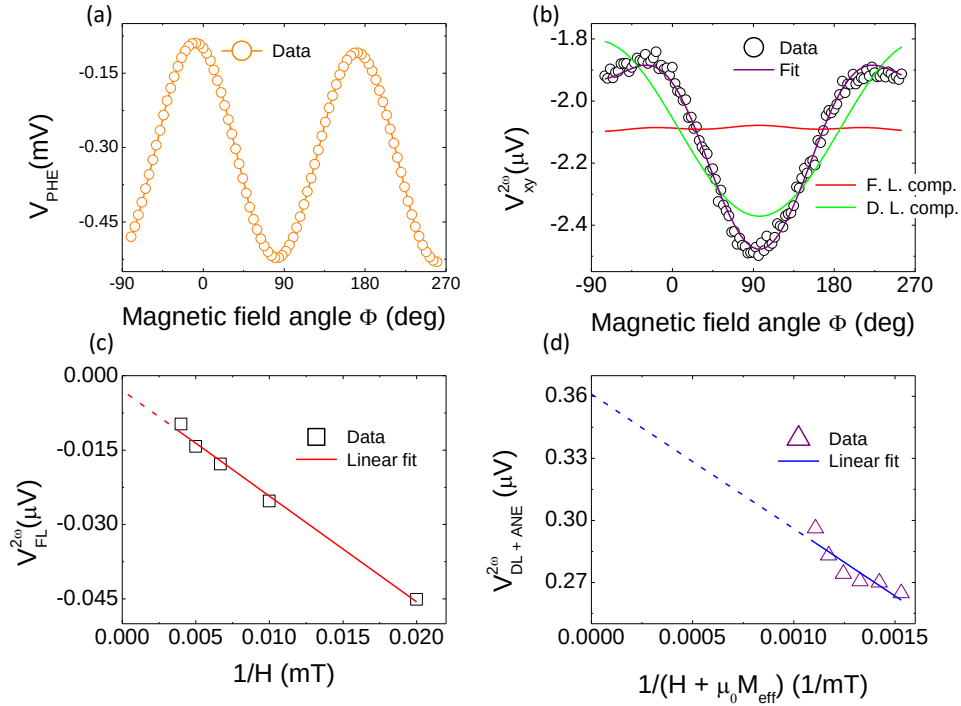


Figure 3.12: Second harmonic Hall for sample LSMO (50 UC)/SIO (18 UC). **a**, First harmonic Hall voltage resulting from the planar Hall magnetoresistance as a function of in-plane magnetic field angle Φ . An external field of 0.25 T and a sample voltage of 4.8 V were applied. **b**, Second harmonic Hall voltage as a function of in-plane magnetic field angle Φ , with an external field of 0.25T and a sample voltage of 4.8 V applied. The solid purple curve is a fit to Eq. (2.3). The red curve is the $\cos(2\Phi)\cos(\Phi)$ term (the field-like component). The green curve is the $\cos(\Phi)$ term (the damping-like component + thermal electric component). **c**, Dependence of the extracted field-like voltage component. The solid curve is a fit to the data using Eq. (2.3). **d**, Dependence of the extracted damping-like and thermal electric voltage component. The blue curve is a fit to the data using Eq. (2.3).

To further validate the SOT efficiency estimated by ST-FMR, second harmonic Hall measurement for an exemplar LSMO (50 UC)/SIO (18 UC) sample was performed. Fig.

3.12a shows the first harmonic Hall signal as a function of magnetic field, illustrating the characteristic $\sin(2\Phi)$ behavior. Fig. 3.12b shows the magnetic field-dependence of the second harmonic Hall signal fit to Eq. (2.3), which allows us to separate out the FL and DL contributions to the Hall signal. Separate curves depict the FL torque, combined in-plane DL torque and Nernst effect in Fig. 3.12b. The $1/H_{\text{ext}}$ -dependence of the FL-Hall voltage is shown in Fig. 3.12c, with an expected linear trend given by Eq. (2.3). To separate the DL torque contribution to the $\cos(\Phi_M)$ term from ANE contribution, $V_{\text{DL+ANE}}^{2\omega}$ is plotted as a function of $1/(H_{\text{ext}} + \mu_0 M_{\text{eff}})$. Since the ANE contribution depends only on the magnetization and is independent of the magnitude of the external magnetic field as long as the magnet is saturated, the ANE signal is given by the y-axis intercept of the linear fit, while the contribution from DL torque τ_{DL} is extracted from the slope of the linear fit. Using the extracted τ_{DL} , the SOT efficiency is calculated by $\xi_{\parallel} = \frac{\tau_{\text{DL}} M_{\text{stFM}}}{\gamma J_{\text{c}} \hbar/2e}$, yielding a value of 1.1, which is an upper bound for the results of the ST-FMR measurements carried out on heterostructures with the same LSMO/SIO thicknesses.

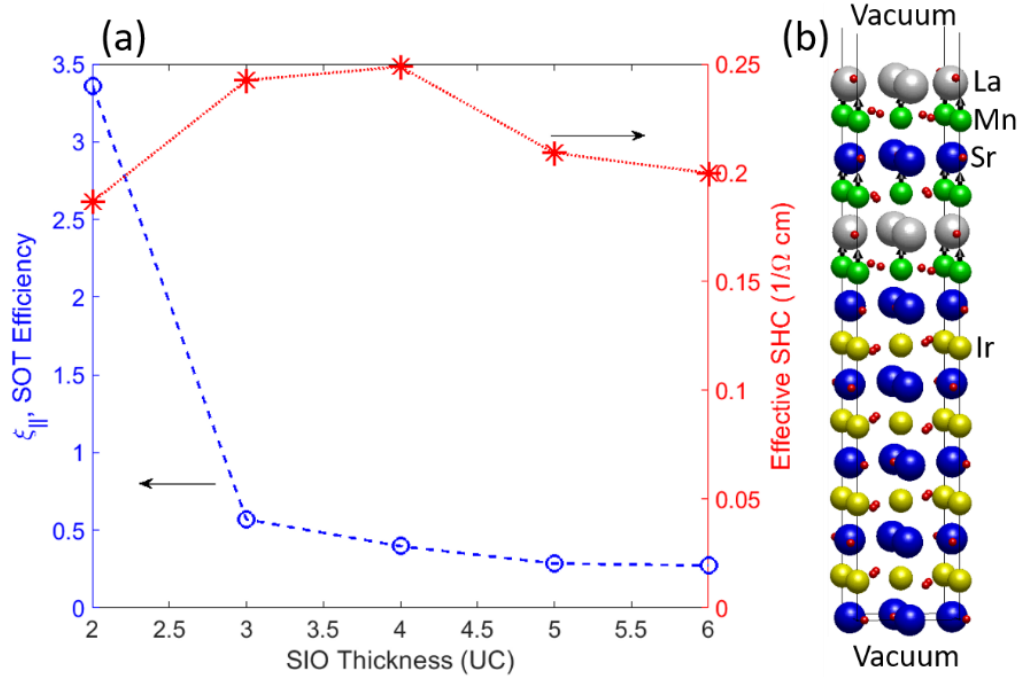


Figure 3.13: First principles calculation of spin orbit torque. **a**, Calculated effective spin-Hall conductivity (right-hand ordinate) versus SIO thickness in the (001) LSMO (3 UC)/SIO (n UC) (n=2-6) bilayer slab. SOT efficiency or Spin Hall Angle (left-hand ordinate), $\xi_{\parallel} = \sigma_{\text{SH}} \rho_{\text{SIO}}$, versus SIO thickness, where σ_{SH} is the ab initio calculated effective SHC and ρ_{SIO} is the experimentally measured thickness-dependent resistivity of SIO from Ref.[161]. **b**, Atomic structure of the (001) LSMO (3 UC)/SIO (4 UC) slab bilayer with Mn-O (Sr-O) terminations for the LSMO (SIO) ultrathin films. The blue, yellow, red, green, and grey spheres denote the Sr, Ir, O,

and La atoms, respectively, and the red arrows denote the non-collinear spin moments of the Mn and Ir atoms.

Theoretical calculations of the damping-like (DL) SOT were carried out using the Kubo formalism[156, 162] in response to an electric field along the a-axis (corresponding to the smaller in-plane lattice constant) for various magnetization orientations. The calculated DL-SOT versus magnetization orientation, $\vec{m}$, are then fitted to $\vec{\tau}_{DL} = \tau_{DL} \vec{m} \times \vec{m} \times (\vec{e}_z \times \vec{E}_{ext})$, in order to determine the DL-SOT strength, τ_{DL} , where $\vec{e}_z$ is a unit vector normal to the interface. Fig. 3.13a displays the results of the first principles calculations for the DL-SOT in the LSMO (3 UC)/SIO (n UC) (n=2-5) bilayer versus the SIO thickness. The atomic structure of the (001) LSMO (3 UC)/SIO (4 UC) slab is shown in Fig. 3.13b, with Mn-O (Sr-O) termination for the LSMO (SIO) ultrathin films. The right-hand ordinate (red symbols) in Fig. 3.13a displays the effective spin-Hall conductivity, $\sigma_{SH} = \frac{2e}{h} \tau_{DL} M_s / 2A\mu_B$, of the LSMO/SIO bilayer versus the SIO thickness. Here, M_s and A are the total magnetic moment and in-plane area per unit cell, respectively. In addition, the left-hand ordinate shows the dimensionless SOT efficiency, $\xi_{||} = \sigma_{SH} \rho_{SIO}$, as a function of SIO thickness, where σ_{SH} is the *ab initio* calculated spin Hall conductivity and ρ_{SIO} is the experimentally measured [161] thickness-dependent room-temperature resistivity of ultrathin SIO. We find the DL-SOT initially increases and then decreases with the SIO thickness. On the other hand, the spin-Hall angle shows a monotonic increase with decreasing SIO thickness, which is in qualitative agreement with the experimental results shown in in Fig. 3.8.

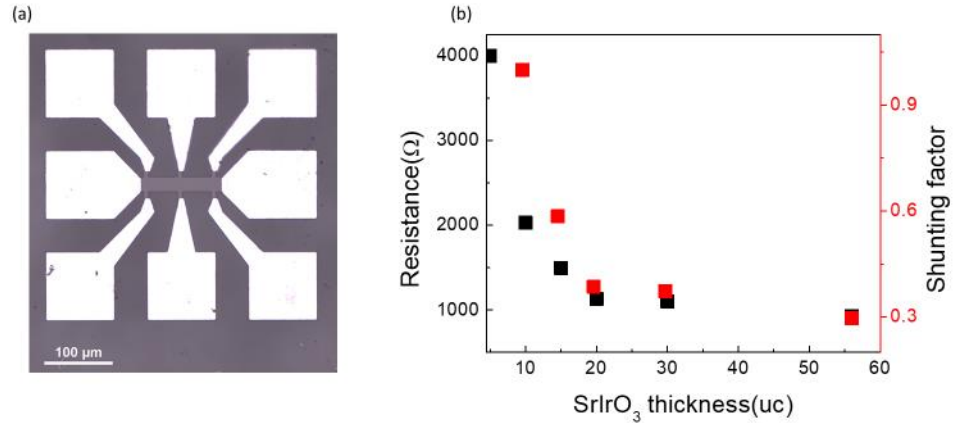


Figure 3.14: Sheet resistance for LSMO/SIO. **a**, A schematic for a Hall device used in the bilayer resistance measurement. **b**, Bilayer LSMO (50 UC)/SIO (X UC) resistance and shunting factor (defined as R_{SIO}/R_{LSMO}) as a function of SIO thickness at room temperature (300 K). The solid black(red) squares indicate bilayer sheet resistance (shunting factor).

We estimate the SOT conductivity ($\sigma_{||}$) of our all-oxide device using $\xi_{||} = \sigma_{||}/\sigma$ to be roughly $1 \sim 3 \times 10^5 (\frac{h}{2e}) \Omega^{-1} \cdot m^{-1}$ [56, 163, 164]. Note that the $\sigma_{||}$ in Pt is of the same order of magnitude, but it is achieved due to the product of a higher charge conductivity

and a lower SOT efficiency. A high SOT efficiency in the epitaxial oxide heterostructure, such as LSMO/SIO is promising for a lower magnetization switching current with much lower current shunting (Fig. 3.14). The most important observations in the epitaxial heterostructure are the key role of the electronic structure at the heteroepitaxial interface (Fig. 3.4) in enhancing the SOT efficiency, and how that electronic structure also induces a change in the anisotropy as measured in through the ST-FMR measurement through the enhancement in the M_{eff} relative to M_s . Our preliminary theoretical studies (Fig. 3.7) confirm that this enhancement in M_{eff} is a consequence of additional anisotropy terms at thin SIO thicknesses. As also observed in the Pt/Co/Oxide heterostructures[165-167], we hypothesize that this is likely a result of electronic orbital hybridization and the associated charge transfer across the LSMO/SIO interface seen by the EELS data (Fig. 3.4). We also recognize that the electronic band structure can be influenced by the lattice parameter and crystal symmetry in addition to charge transfer effects. The key difference between the results presented in Fig. 3.8 for the epitaxial LSMO/SIO interface and the SIO/Py interface[140] is the role of epitaxial constraints imposed on the crystal and electronic structure. We note that the SOT efficiency in our measurements for SIO layer thickness larger than ~ 25 -30UC is essentially the same as that reported by T. Nan et al[140]. The significant enhancement occurs for SIO thicknesses smaller than ~ 25 UC, for which the consequences of epitaxial constraint on the electronic and crystal structure are strongly manifested in the enhancement of the SOT efficiency.

3.5 Summary

In summary, I use heteroepitaxial perovskite oxides driven by the intrinsic similarity in crystal chemistry and structure to design and synthesize epitaxial oxide FM/SOC metal heterostructures as model systems to study the SOT efficiency and novel interface-driven spin-orbitronics. Using ST-FMR, I show heterostructures with $M_{\text{eff}} > M_s$ and $\xi_{||}=1$, taking advantage of intrinsic band structure and correlated electron phenomena. I show that precise control over crystallinity, crystal orientation, interface quality, and strain states of the bilayer, allows us to achieve high SOT efficiency with lower current shunting factor. The critical role of the interface electronic structure strongly suggests future work on manipulating the interface electronic structure through the insertion of “surfactant” layers that can locally manipulate the electronic structure. Furthermore, this work suggests a broader set of FM/SOC heterostructures that can be created using the principles of heteroepitaxy to yield large SOT efficiencies, given the rich diversity of oxide materials.

Chapter 4

Spin transport manipulation: Interface termination

A collective excitation of the spin structure in a magnetic insulator can transmit spin-angular momentum with negligible dissipation[168-172]. This quantum of a spin wave, introduced more than nine decades ago, has always been interpreted and manipulated in terms of magnetic dipoles[173-175], (i.e., time-reversal symmetry). Here, I show the experimental observation of chiral-spin transport in multiferroic BiFeO_3 , where the asymmetric magnon dispersion is instead controlled via inversion symmetry the spin transport is controlled by reversing the ferroelectric polarization (i.e., spatial inversion symmetry). The ferroelectrically controlled chiral magnons produce an unprecedented ratio of up to 40% rectification at room temperature. The spin torque that the magnons in BiFeO_3 carry can be used to efficiently switch the magnetization of adjacent magnets, with a spin-torque efficiency being comparable to the spin Hall effect in heavy metals. Utilizing these controllable chiral magnons in BiFeO_3 , an all-oxide, energy-scalable logic is demonstrated composed of spin-orbit injection, detection, and magnetoelectric control. This observation opens a new chapter of multiferroic magnons and paves an alternative pathway towards low-dissipation nanoelectronics.

4.1 Introduction

Intense studies of spin-current transport in magnetic insulators[82, 86, 87, 171] have opened various avenues for research on spintronics[83, 84, 88, 89, 176, 177] and their potential use in low-power applications. This trend has also triggered renewed interest in magnon physics, which can be controlled via time-reversal-symmetry design (e.g., magnetic domain walls[173], spin-Hall currents[174], static magnetic fields[175]). On the other hand, recent works have shown that broken inversion symmetry and the resulting chiral Dzyaloshinskii-Moriya interaction (DMI) can also influence magnon propagation in non-centrosymmetric magnets[178-180]. In this spirit, multiferroics are potentially of great interest, since they display broken inversion symmetry and a ferroelectric polarization (ferroelectric order) which, in turn, couples to the magnetism such that it might transform the magnons polarization that can be switched by application of an electric field, which in turn allows for control of the magnetism and, potentially, the magnons. The idea that multiferroics could be promising candidates for magnonic manipulation has been considered for some time[66, 67], but despite this early interest, observation and manipulation of chiral magnon propagation in multiferroics - that is, the capability to manipulate chiral spin transport via ferroelectric polarization reversal - remains elusive.

Here, I demonstrate non-volatile, bistable chiral-spin transport in multiferroic BiFeO_3 upon reversing the ferroelectric polarization. BiFeO_3 is a model multiferroic that exhibits multiple order parameters (i.e., antiferromagnetism and ferroelectricity) and intrinsic magnetoelectric coupling of these coexisting order parameters[91, 92]. In the bulk, BiFeO_3 possesses a rhombohedrally distorted perovskite structure in which a spin cycloid is formed with period length of ~ 64 nm[104] and a Néel temperature of ~ 643 K[105]. Strain in BiFeO_3 thin films can break the spin cycloid and favors a G-type antiferromagnetic order wherein canting of the moments can produce a weak, long-range net magnetization ($\sim 0.1 \mu_B$ per Fe)[66, 106], arising from the DMI. At the same time, BiFeO_3 thin films possess robust ferroelectricity, with a Curie temperature ~ 1100 K and large polarization $> 90 \mu\text{C}/\text{cm}^2$ [107]. In turn, BiFeO_3 exhibits outstanding magnetoelectric properties which have made it the focus of numerous studies hoping to utilize it as a platform to achieve low-energy (low-voltage) memory devices (e.g., deterministic switching of adjacent ferromagnets[93]). Specifically, achieving control over changing the magnetization states by means of ferroelectric polarization switching has always been the keen pursuit in these studies. A new mechanism is proposed herein, that magnons in this system can serve as a measure of the magnetization states subject to the control of ferroelectric polarization.

4.2 Concept of spin transport manipulation in multiferroics

As illustrated (Fig. 4.1a and Fig. 4.1b), a 180° reversal of ferroelectric polarization $\mathbf{P}$ leaves the Néel vector $\mathbf{L}$ invariant[93]. The polarization switch is accompanied by the

reversal of the octahedral antiphase tilts $\mathbf{A}$, which in turn controls the sign of the chiral interaction corresponding to the antisymmetric exchange coupling (DMI)[106, 181]. Therefore, the magnon chirality defined in terms of the magnon propagation wave vector $\mathbf{K}$, the vector $\mathbf{A}$, and the Néel vector $\mathbf{L}$ (taken to be along $[\bar{1} 0 1]_{pc}$) changes from left-handed to right-handed. This change in chirality can produce rectification, i.e. a difference in magnon propagation depending on the sign of the direction of travel[178, 182]. To explore this possibility, we undertook coupled phase-field and micromagnetic simulations of BiFeO_3 to test whether multiferroic switching indeed allows for control of the dc component of a spin wave excitation. The switching of ferroelectric polarization from $[\bar{1} \bar{1} \bar{1}]$ to $[1 1 1]$ alters the chirality of the preferred magnons and indeed switches the sign of the spin current excited by a given magnetic-field excitation Fig. 4.1c.

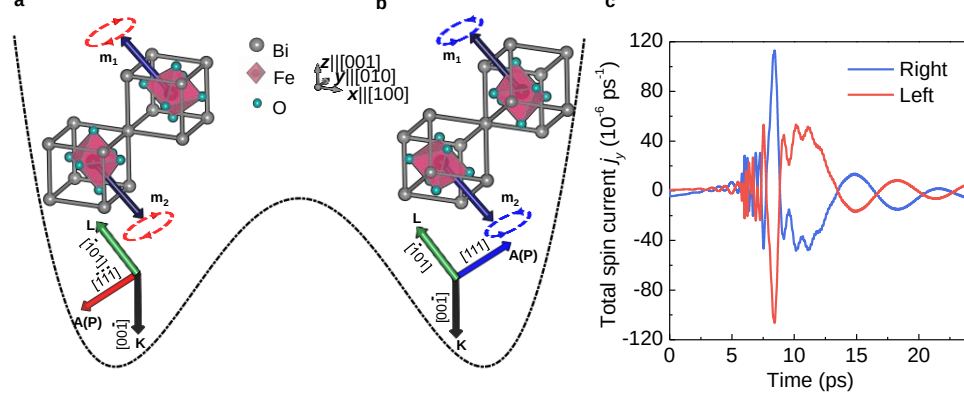


Figure 4.1: Concept of chiral magnon propagation in multiferroics. **a, b**, Sketches of magnon propagation in BiFeO_3 lattice. The double well denotes the bi-stable states of ferroelectric polarizations. The magnon propagation wave vector $\mathbf{K}$, octahedral anti-phase tilt field $\mathbf{A}$ (parallel to $\mathbf{P}$) and Néel vector $\mathbf{L}$ form a left-handed chirality and right-handed chirality for a and b respectively. **c**, Calculated y component of the total spin current j as a function of time depending on the chirality. The applied magnetic field H to generate the spin wave excitation is set along $[\bar{1} 0 1]$ or parallel to the Néel vector $\mathbf{L}$. The ratio of dc components of right-handedness (upward polarization) and left-handedness (downward polarization) is -1.06. Similar behavior is seen in the other components of the spin current.

4.3 Ferroelectric polarization manipulation by interface termination

4.3.1 Previous study on bulk ferroelectric polarization manipulation by interface termination

Previously, it has been shown that the interface termination in the LSMO/BFO heterostructure can directly determine the bulk ferroelectric polarization of BFO[183]. Due to the ABO_3 perovskite structure exhibited by both LSMO and BFO, two kinds of

distinct interfaces can be created in the [0 0 1]-oriented LSMO/BFO heterostructure. If the bottom electrode LSMO is terminated with MnO_2 (B site termination), an interface with stacking sequence of $\text{La}_{0.7}\text{Sr}_{0.3}\text{O}-\text{MnO}_2-\text{BiO}-\text{FeO}_2$ can be created (Fig. 4.2a). On the other hand, if LSMO is terminated with $\text{La}_{0.7}\text{Sr}_{0.3}\text{O}$ (A site termination), the interface stacking sequence will be changed to $\text{MnO}_2-\text{La}_{0.7}\text{Sr}_{0.3}\text{O}-\text{FeO}_2-\text{BiO}$. The distinctly different interfacial stacking sequences can directly impact the electronic interaction at these heterointerfaces, which ultimately determines the bulk polarization of BFO. The ferroelectric manifestation of different interface terminations was checked with piezoresponse microscopy (PFM). As displayed in Fig. 4.2c and Fig. 4.2d, the contrast of the out-of-plane PFM images is an indication of the ferroelectric polarization orientations in the out-of-plane direction, with polarization in the dark area pointing upward (from bottom electrode to top electrode) and polarization in the light area pointing downward (from top electrode to bottom electrode). Intriguingly, the out-of-plane ferroelectric polarization orientation is strongly correlated with the interface termination of the LSMO/BFO heterostructures. Specifically, an upward as-grown ferroelectric polarization is seen for samples with $\text{La}_{0.7}\text{Sr}_{0.3}\text{O}-\text{MnO}_2-\text{BiO}-\text{FeO}_2$ stacking sequence, while a downward as-grown ferroelectric polarization is associated with a $\text{MnO}_2-\text{La}_{0.7}\text{Sr}_{0.3}\text{O}-\text{FeO}_2-\text{BiO}$ interfacial atomic sequence, as shown in Fig. 4.2c and Fig. 4.2d. The as-grown ferroelectric polarization states are further revealed by the systematic shifting of the hysteresis loops in the piezoelectric (or ferroelectric) switching measurements (Fig. 4.2e and Fig. 4.2f). For heterostructure with $\text{La}_{0.7}\text{Sr}_{0.3}\text{O}-\text{MnO}_2-\text{BiO}-\text{FeO}_2$ interfacial sequence, a positive shift with respect to zero volt is observed (Fig. 4.2e), indicating a positive internal voltage (Fig. 4.2g) and an upward as-grown polarization state. The opposite shift (negative) is seen for $\text{MnO}_2-\text{La}_{0.7}\text{Sr}_{0.3}\text{O}-\text{FeO}_2-\text{BiO}$ interface, suggesting a negative internal voltage (Fig. 4.2h) and a downward as-grown polarization state. Thus, by manipulating the interfacial sequence of the heterostructure, the as-grown ferroelectric polarization states can be precisely controlled.

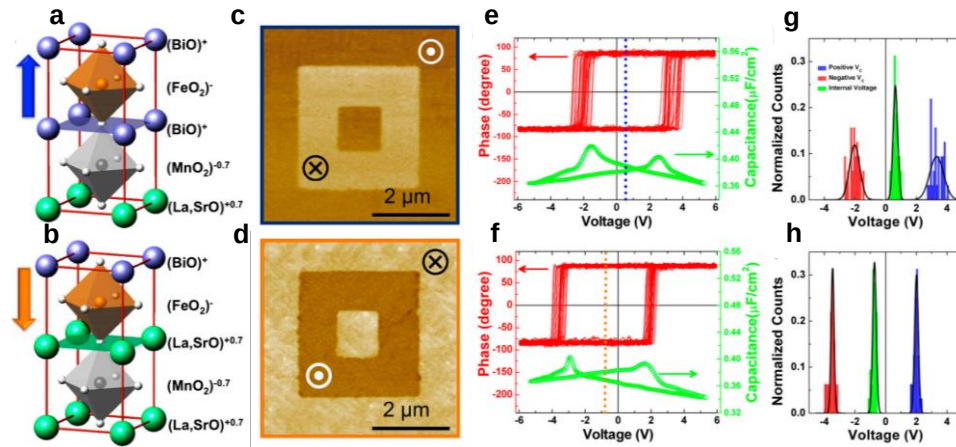


Figure 4.2: Interface termination of bulk polarization. **a, b**, Schematic illustrations of LSMO/BFO heterostructures with $\text{La}_{0.7}\text{Sr}_{0.3}\text{O}-\text{MnO}_2-\text{BiO}-\text{FeO}_2$ and $\text{MnO}_2-\text{La}_{0.7}\text{Sr}_{0.3}\text{O}-\text{FeO}_2-\text{BiO}$ interfaces respectively. **c, d**, Typical out-of-plane PFM images of LSMO/BFO heterostructures with $\text{La}_{0.7}\text{Sr}_{0.3}\text{O}-\text{MnO}_2-\text{BiO}-\text{FeO}_2$ and MnO_2-

$\text{La}_{0.7}\text{Sr}_{0.3}\text{O}-\text{FeO}_2-\text{BiO}$ interfaces respectively, and corresponding quasistatic piezoresponse (Top, red) and capacitance-voltage (Bottom, green) hysteresis loops (e and f). g, h, Histogram analysis of obtained coercive voltage and biased voltage from the piezoresponse measurements in e and f. Adapted from Ref.[183].

4.3.2 Interface termination control by RHEED assisted PLD

The key to achieving difference interfacial sequences for the heterostructure lies in the ABO_3 perovskite crystal structure of the SrTiO_3 substrate, which enables two kinds of terminations (A site termination and B site termination) for the (0 0 1)-oriented substrate. The SrTiO_3 substrates treated with buffered HF etch and thermal anneal procedures are terminated with TiO_2 atomic plane (B site termination)[184]. Therefore, the MnO_2 (B site) termination LSMO can be achieved by growing on the treated SrTiO_3 by direct. The RHEED intensity oscillation for growing LSMO on SrTiO_3 by direct is shown in Fig. 4.3a, and clear intensity oscillations during the growth is seen, indicating a layer-by-layer growth mode. The resulting $\text{La}_{0.7}\text{Sr}_{0.3}\text{O}-\text{MnO}_2-\text{BiO}-\text{FeO}_2$ is displayed in Fig. 4.3b. To grow $\text{La}_{0.7}\text{Sr}_{0.3}\text{O}$ (A site) terminated LSMO, we need to modify the B site terminated substrate to be A site terminated, which can be achieved by growing 2.5-unit cells of SrRuO_3 on the B site terminated SrTiO_3 substrates (Fig. 4.3c). Because of the high volatility of RuO_2 [185], SrRuO_3 at high temperature (700 °C) will be self-terminated with SrO (A site), which leads to the modification of the substrate termination from TiO_2 to SrO . The subsequent deposition of 50 unit-cell of LSMO results in $\text{La}_{0.7}\text{Sr}_{0.3}\text{O}$ terminated LSMO (Fig. 4.3d).

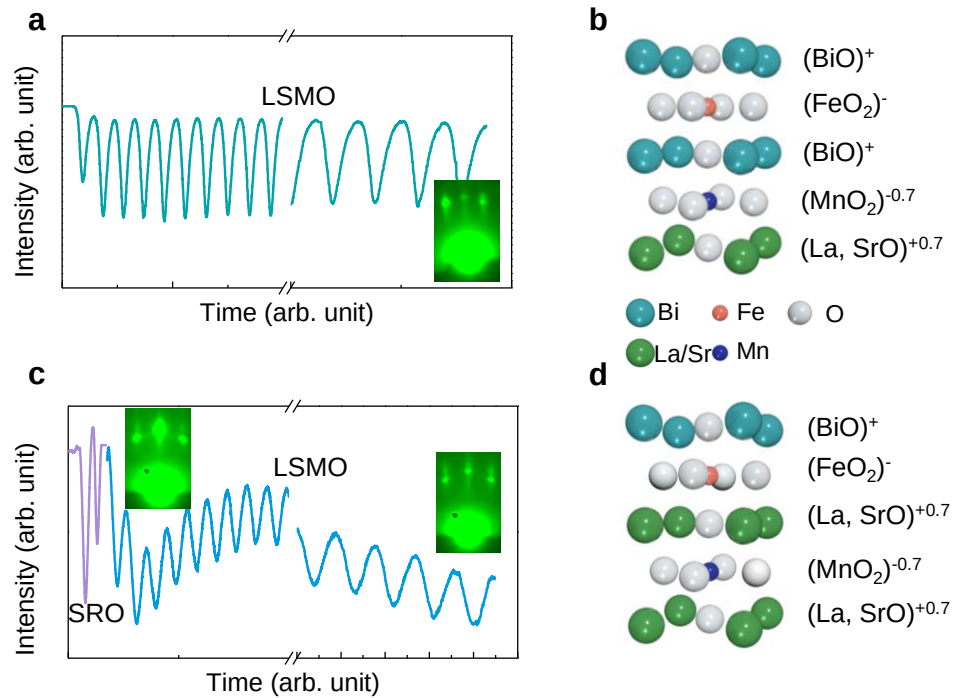


Figure 4.3: Atomic scale control of interface termination by pulsed laser deposition. a, RHEED intensity oscillation of the specular beam during the growth of

$\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ by direct on TiO_2 -terminated SrTiO_3 (0 0 1) surface. **b**, A schematic of the resulting MnO_2 -BiO interface. **c**, RHEED intensity oscillation of the specular reflected beam during the growth of SrRuO_3 and $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ on TiO_2 -terminated SrTiO_3 (0 0 1) surface. A growth of 2.5 unit-cell of SrRuO_3 on TiO_2 terminated SrTiO_3 substrate is required in order to achieve the SrO terminated substrate, due to the fact that RuO is highly volatile at high temperature and SrRuO_3 is therefore self-terminated with SrO layer. **d**, A schematic of the resulting $\text{La}_{0.7}\text{Sr}_{0.3}\text{O}$ - FeO_2 interface.

Following growth, X-ray diffraction $2\theta - \omega$ line scan (Fig. 4.4e) reveals that the STO/LSMO/BFO/SIO heterostructure grows epitaxially and a clear peak for LSMO (0 0 2) is observed. Due to the lattice constants match between BFO and SIO, the BFO (0 0 2) peak and SIO (0 0 2) peak are highly convoluted and cannot be readily distinguished. However, Kiessig fringes accompanying BFO and/or SIO (0 0 2) peak with two distinct periodicities were observed, suggesting that two distinct layers of different thicknesses are present. To examine the quality of the oxide interface at the microscopic level, we performed high angle annular dark field (HAADF) scanning transmission electron microscopy (STEM) experiments (Fig. 4.4a). The HAADF image reveals a high quality heterostructure consisting of sequential LSMO, BFO and SIO, with atomically sharp interfaces. The surface topography of the (0 0 1)-oriented SrTiO_3 substrate was checked with AFM before growths, showing the presence of atomically flat terraces as large as $1\ \mu\text{m}$ (Fig. 4.4b). Another crucial property for the detecting of spin transmission is the room temperature ferromagnetism of the heterostructure (or loosely speaking LSMO). Magnetometry measurements at room temperature were performed for the tri-layer (Fig. 4.4c), in which a hysteresis develops when the strength of the applied magnetic field in the plane exceeds that of the coercive field. A saturation magnetization approximately 320 emu/cc and a coercive field 25 Oe were determined, suggesting a high quality LSMO layer. Ferroelectric domains of BFO on LSMO were imaged with PFM, revealing the presence of stripe-like ferroelectric domain structures separated by 71° domain walls.

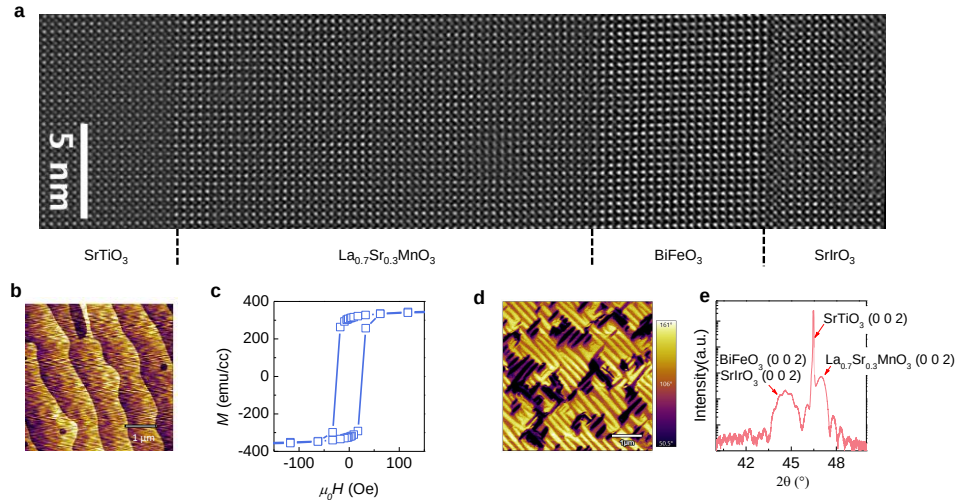


Figure 4.4: Structural, magnetic, and ferroelectric characterization for heterostructure LSMO/BFO/SIO. **a**, HAADF image of LSMO/BFO/SIO tri-layer, displaying

atomically sharp interfaces and high crystal quality. **b**, HF etched and thermally annealed SrTiO₃ substrate with atomic steps and terraces. **c**, Magnetization vs applied magnetic field (MH) measurement for LSMO/BFO/SIO heterostructure, showing a coercivity of ~ 25 Oe and a saturation magnetization ~ 320 emu/cc. **d**, In-plane piezoresponse microscopy (PFM) image of the BiFeO₃ layer. **e**, High resolution XRD $2\theta - \omega$ scan of the tri-layer.

4.3.3 As-grown ferroelectric polarization states probed by piezoelectric switching

In this section, I am going to investigate how the different interfacial structures introduced in the previous sections can impact the electronic interactions at the interface. To verify the successful control of bulk polarization of BFO by interface termination, piezoresponse force microscopy (PFM) is employed to probe the ferroelectric polarization states of the ferroelectric layer BiFeO₃ in the LSMO/BFO heterostructure with different interfacial terminations (La_{0.7}Sr_{0.3}O-MnO₂-BiO-FeO₂ in Fig. 4.5a and MnO₂-La_{0.7}Sr_{0.3}O-FeO₂-BiO in Fig. 4.5b). The PFM measurements were carried out with an AFM-based setup under ambient environment using a commercially acquired TiPt-coated Si tip. The piezoelectric hysteresis loops were obtained by measuring the surface displacement as a function of applied DC voltage (up to ± 9 V). Strikingly, the as-grown ferroelectric polarization states are strongly correlated with the interface terminations. The local switching behavior for a La_{0.7}Sr_{0.3}MnO₃/BiFeO₃ bilayer with La_{0.7}Sr_{0.3}O-MnO₂-BiO-FeO₂ interface stacking is shown in Fig. 4.5c and a right shift of the hysteresis loop with respect to zero volt is unambiguously observed, indicating an upward as-grown polarization. On the contrast, a left shift of hysteresis loop with respect to zero volt for a La_{0.7}Sr_{0.3}MnO₃/BiFeO₃ bilayer with MnO₂-La_{0.7}Sr_{0.3}O-FeO₂-BiO interface stacking is clearly seen (Fig. 4.5d), showing a downward as-grown polarization.

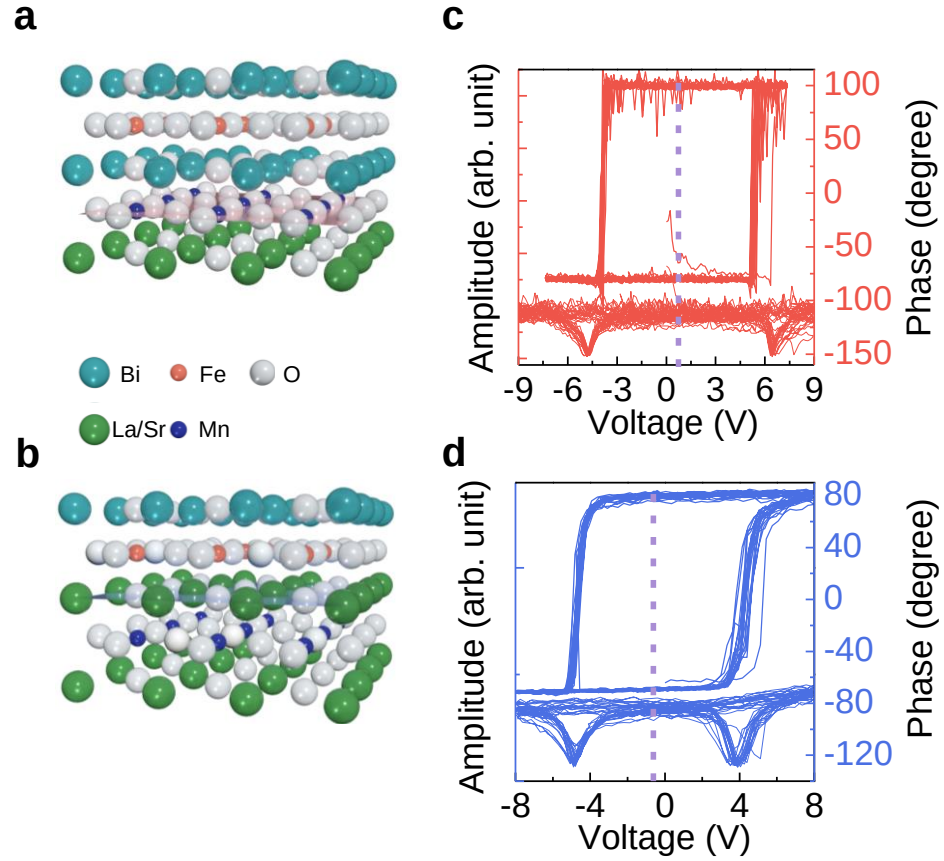


Figure 4.5: Piezoelectric hysteresis loops for LSMO/BFO with different interface terminations. **a**, A schematic for LSMO/BFO atomic stacking, where $\text{La}_{0.7}\text{Sr}_{0.3}\text{O-MnO}_2\text{-BiO-FeO}_2$ is stacked at the interface. **b**, A schematic for LSMO/BFO atomic stacking, where $\text{MnO}_2\text{-La}_{0.7}\text{Sr}_{0.3}\text{O-FeO}_2\text{-BiO}$ is stacked at the interface. **c**, Corresponding piezoresponse for $\text{La}_{0.7}\text{Sr}_{0.3}\text{O-MnO}_2\text{-BiO-FeO}_2$ stacking (LSMO (12 nm)/BFO (50 nm)), with top and bottom being phase and amplitude respectively. The solid line denotes the shifting of the piezoresponse curves to zero volts, indicating an upward ferroelectric polarization for $\text{La}_{0.7}\text{Sr}_{0.3}\text{O-MnO}_2\text{-BiO-FeO}_2$ stacking. **d**, Corresponding piezoresponse for $\text{MnO}_2\text{-La}_{0.7}\text{Sr}_{0.3}\text{O-FeO}_2\text{-BiO}$ stacking (LSMO (12 nm)/BFO (50 nm)), with the top and bottom being phase and amplitude respectively. The solid line denotes the shifting of the piezoresponse curves to zero volts, indicating a downward ferroelectric polarization for $\text{MnO}_2\text{-La}_{0.7}\text{Sr}_{0.3}\text{O-FeO}_2\text{-BiO}$ stacking.

4.4 Spin transport in multiferroic BiFeO₃ controlled by interface termination

4.4.1 BiFeO₃ thickness dependent study with ST-FMR technique

The ST-FMR signal, V_{mix} , is produced across the LSMO ST-FMR device as a rectification of anisotropic magnetoresistance that oscillates at the same frequency as the input microwave current I_{RF} . A microwave current at frequencies 4-6 GHz and power 12 dBm is applied to the device, with an in-plane external magnetic field oriented 45° with respect to the current direction. Finally[117],

$$V_{\text{mix}} = -\frac{1}{4} \frac{dR}{d\theta} \frac{\gamma I_{\text{RF}} \cos\theta}{2\pi\Delta \left(\frac{df}{dH}\right)_{H_{\text{ext}}=H_0}} (\tau_{\text{DL}} F_{\text{S}}(H_{\text{ext}}) + \tau_{\text{FL}} F_{\text{A}}(H_{\text{ext}})), \quad (4.1)$$

where $dR/d\theta$ is the angle dependent magnetoresistance at θ , γ is the gyromagnetic ratio, Δ is the linewidth of the ST-FMR signal, $\left(\frac{df}{dH}\right)_{H_{\text{ext}}=H_0}$ is the field gradient of the resonance frequency, τ_{DL} is the damping-like torque, τ_{FL} is the field-like torque, $F_{\text{S}}(H_{\text{ext}})$ is the symmetric Lorentzian function and $F_{\text{A}}(H_{\text{ext}})$ is the anti-symmetric Lorentzian function respectively, is detected by Keithley 2182A nanovoltmeter, from the fits of which the symmetric component V_{S} and the anti-symmetric component V_{A} , which are associated with damping-like torque (τ_{DL}) and field-like torque (τ_{FL}) respectively, can be extracted. As a result, spin orbit torque efficiency- ξ_{SOT} is evaluated using $\xi_{\text{SOT}} = \frac{2e\tau_{\text{DL}}M_{\text{S}}t_{\text{FM}}}{J_{\text{C}}\hbar}$, where

$$\tau_{\text{DL}} = \frac{4V_{\text{S}}\Delta}{I_{\text{RF}}\cos\theta \frac{dR}{d\theta} \left(\frac{1+\mu_0 M_{\text{eff}}}{B_{\text{res}}}\right)^{0.5}} [59, 186].$$

The SOT efficiency for the samples in this work was measured from 4 GHz to 6 GHz and the error bar in Fig. 4.6c is a result of SOT efficiencies distribution across different devices.

To investigate the critical role that ferroelectric polarization plays in the spin transport in BiFeO₃, I first engineer the interface termination between La_{0.7}Sr_{0.3}MnO₃ and BiFeO₃ to be either La_{0.7}Sr_{0.3}O-MnO₂-BiO-FeO₂ or MnO₂-La_{0.7}Sr_{0.3}O-FeO₂-BiO (4.3.3). Specifically, the as-grown state of samples possessing MnO₂-terminated interface is upward (Fig. 4.6a), whereas the as-grown state of samples possessing La_{0.7}Sr_{0.3}O-terminated interface is downward (Fig. 4.6b). To understand the potential magnonic manifestations of the opposite as-grown polarizations, the same ST-FMR measurements are performed on the two sets of samples with opposite polarization states depicted in Fig. 4.6a and Fig. 4.6b. The SOT efficiency as a function of BiFeO₃ thickness is summarized in Fig. 4.6c, in which open squares and open circles represent SOT efficiencies for samples with upward polarization and downward polarization respectively. Strikingly, the SOT efficiency for the trilayer with upwardly poled BiFeO₃ increases at small BiFeO₃ thickness then decays exponentially at larger BiFeO₃ thickness, while the SOT efficiency for the trilayer with downwardly poled BiFeO₃ monotonically decays. Noteworthily, the SOT efficiency for system with upward polarization is consistently larger than that for system with downward polarization, signifying the existence of a bi-stable on and off spin transmission states in BiFeO₃ governed by ferroelectric polarization.

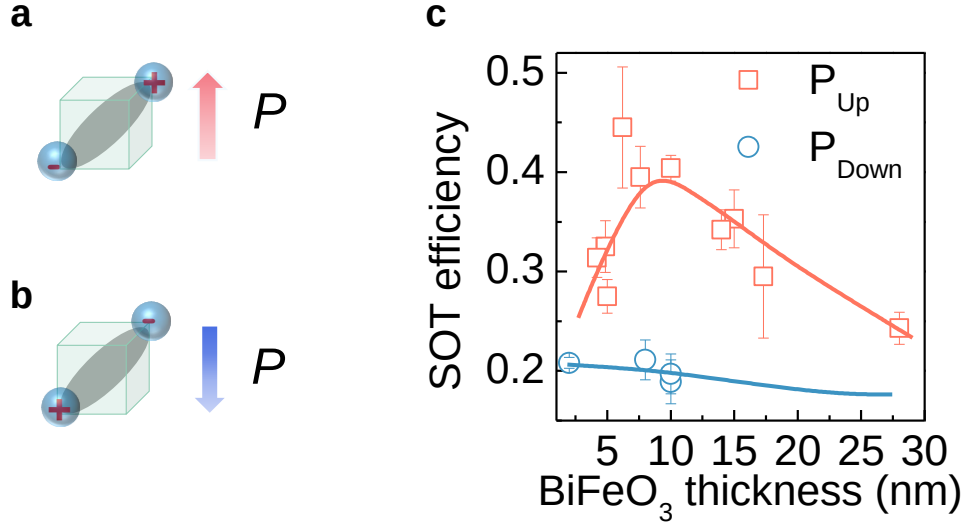


Figure 4.6: Ferroelectric control of spin transport enabled by the interface chemistry. **a, b**, An artistic illustration for upward and downward ferroelectric polarization. **c**, Spin orbit torque efficiency for BiFeO₃ with different polarization states as a function of BiFeO₃ thicknesses. The red (blue) data points represent the spin orbit torque efficiency for LSMO, where BiFeO₃ has an upward (downward) polarization.

4.4.2 Direct magnetization switching

The magneto transport experiments, including longitudinal resistance and switching measurements, were conducted in Physical Property Measurement System (Cryogenic Ltd). The devices used for current-induced magnetization switching measurements consist of two detection channels ($10 \mu\text{m} \times 80 \mu\text{m}$) and two writing ($5 \mu\text{m} \times 80 \mu\text{m}$) channels. The current channel for detection is denoted as I^+ , I^- and the corresponding perpendicular Hall-voltage channel is denoted as V_{PHE}^+ , V_{PHE}^- , forming a planar Hall measurement configuration. The two writing channels $J_{\text{write } 1}$ and $J_{\text{write } 2}$ are at 135° and 225° with respect to the current channel used for detection. Initially, I saturated the magnetization of the LSMO layer with an external magnetic field of 1000 Oe along $J_{\text{write } 1}$ that is at 135° to the current channel for detection, resulting in a maximum value of planar Hall resistance as indicated in Fig. 4.7b. Then 1 ms current pulses were applied in the $J_{\text{write } 1}$ channel after the removal of the external field. The resultant anti-damping like torque ($\tau_{AD} \sim m \times (\sigma \times m)$) denoted by the green arrow in Fig. 4.7c is pointed perpendicular to the magnetic moment m (red arrow in Fig. 4.7c), which eventually reorients the magnetic moment to the τ_{AD} direction with large enough pulsing currents. I find that the magnetic moment is always switched by 90° from its original orientation, thus causing the planar Hall resistance to decrease as shown in Fig. 4.7c. Similarly, when the magnetic moment is initially aligned along the $J_{\text{write } 2}$ channel at 225° relative to the current channel for detection (red arrow in Fig. 4.7d), current pulses in the $J_{\text{write } 2}$ channel switch the magnetic

moment away from the $J_{\text{write } 2}$ channel towards 315° orientation, thereby increasing the planar Hall resistance as displayed in Fig. 4.7d.

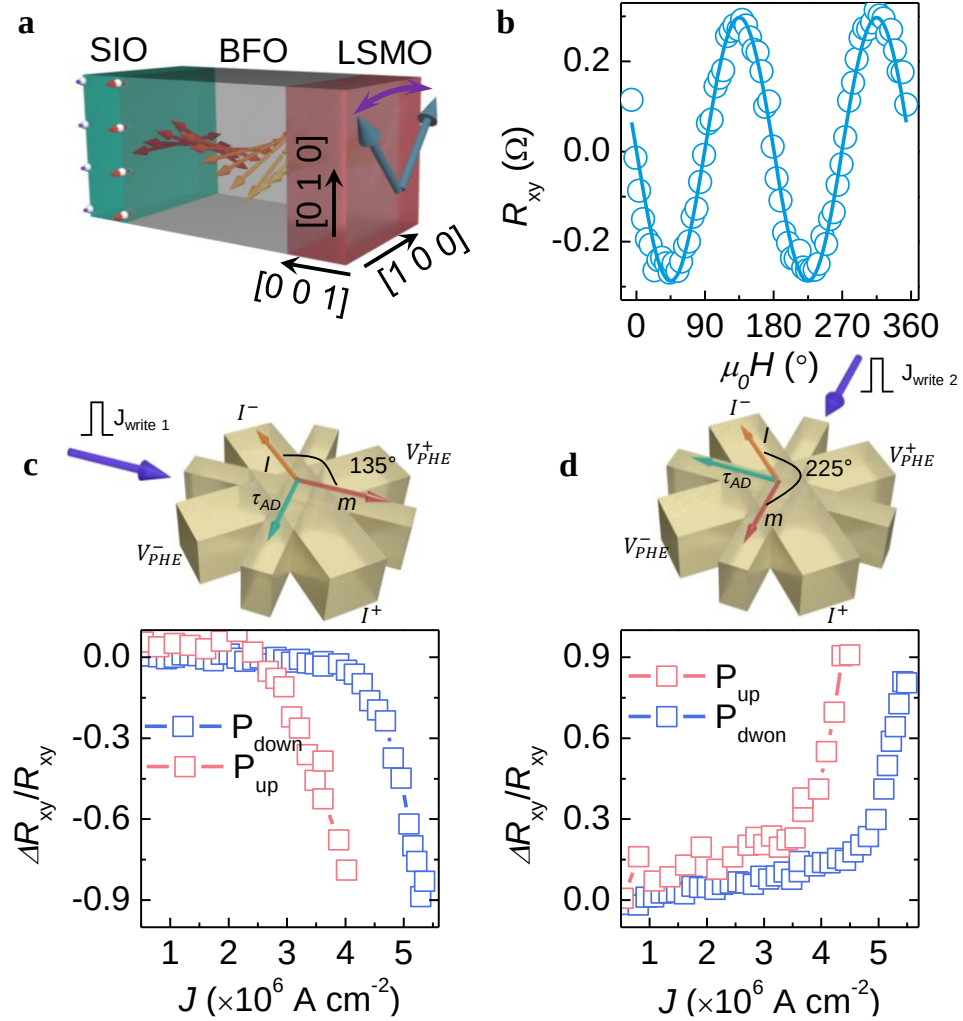


Figure 4.7: Magnetization switching induced by the spin torque in BiFeO₃ under the control of ferroelectric-polarization at room temperature. a, A schematic demonstrating the magnetization switching of LSMO by the current-induced torques borne by the magnons transmitted through BiFeO₃ for a sample LSMO (20 nm)/BFO (10 nm)/SrIrO₃ (10 nm). The oscillating arrows represent the magnon in BFO excited by the spin accumulation at the BFO/SIO interface. The torque carried by the transmitted magnons switches the in-plane magnetization (dark blue arrows) of the neighboring LSMO layer. b, The planar Hall resistance (R_{xy}) as a function of the in-plane magnetic field angle, showing a $\sin 2\theta$ dependence. The strength of the magnetic field is fixed at 50 Oe. c, d, The top panels are schematics for the switching measurements with current pulses along the 135° and 45° directions with respect to the probing current channel, respectively. The normalized planar Hall resistance change as a function of the progressively increasing pulsing current density along the 135° and 45° direction with respect to the detection current channel (I^+ , I^-). The

current pulses are 1 ms long. A constant current of 100 μA is applied on I^+, I^- channel for probing the planar Hall resistance change.

To corroborate the presence of chiral magnon propagation in BiFeO_3 , magnetization switching experiments were carried out to serve as a direct demonstration of the strength of spin torques under ferroelectric-polarization modulation. Previously, it has been reported that the spin torque that arises from the magnons in magnetic insulators can be used to switch an adjacent ferromagnet[87, 88]. There is, however, a lack of experimental observations to this effect in multiferroic materials. Here, I investigate 90° magnetization switching of an in-plane-oriented LSMO layer by the spin torque carried by the BiFeO_3 . Current-induced magnetization switching measurements were carried out for LSMO (20 nm) /BFO (10 nm) /SIO (10 nm) heterostructures with upward- and downward-pointing ferroelectric polarization at room temperature (Fig. 4.3a). The switching schematic is also demonstrated (Fig. 4.3a), where the magnetic moment of the ferromagnetic manganite is switched by 90° consistently (e.g., from $[1\ 1\ 0]$ to $[1\ \bar{1}\ 0]$). The magnetic switching was detected by the planar Hall effect in the LSMO. By rotating an external magnetic field (50 Oe) larger than the coercive field of LSMO, the planar-Hall resistance (R_{xy}) is clearly observed to show a $\sin 2\theta$ angular dependence (Fig. 4.7b). The maxima and minima of the planar-Hall resistance are unambiguously observed when the magnetic moments orient 135° (315°) and 45° (225°) with respect to the probing current direction, indicating that the magnetic moments on the aforementioned orientations can be precisely identified with the planar-Hall measurement configuration. For the magnetization switching measurements as described thoroughly in the last paragraph, I first used an in-plane magnetic field to saturate the magnetization of the LSMO along the current pulse channel, which is oriented at 135° relative to the current direction. Then, after the removal of the magnetic field, progressively increasing current pulses were applied in the channel in order to reorient the magnetization to the perpendicular direction (225°). The magnetization of the LSMO was switched towards the 225° direction upon reaching the critical switching current density, as indicated by the planar Hall resistance (Fig. 4.7c). Similarly, after the magnetization of LSMO was initialized along the 225° direction, pulsed currents were applied in this direction could switch the magnetization to the 315° direction (Fig. 4.7d). This observation verifies that the spin torque carried by the BiFeO_3 is sufficient to enable magnetization switching of an adjacent ferromagnet. The critical switching current densities were averaged to be approximately $3.2 \times 10^6\text{ A/cm}^2$ and $4.2 \times 10^6\text{ A/cm}^2$ for upward and downward ferroelectric polarizations, respectively, comparable to the critical switching current density observed in the LSMO/SIO system[116]. I calculated the magnitude of Oersted field generated by the pulsing current and compared it with the magnetic coercive field of LSMO. By using the geometry described in Fig. 4.7, the pulsing current generated Oersted was calculated as[34] $B_{\text{Oersted}} = \mu_0 J \frac{d_1 + d_2}{2}$ with $J = 3.2 \times 10^6\text{ A/cm}^2$ (corresponding to 1.6 mA), $d_1 = 10\text{ nm}$ (SrIrO_3 thickness) and $d_2 = 30\text{ nm}$ (BiFeO_3 10 nm and LSMO 20 nm). B_{Oersted} was estimated to be around 8 Oe, way smaller than the magnetic coercivity of LSMO (around 25 Oe). The Oersted field generated by the pulsing current can be ruled out because magnetization switching induced by Oersted field is not dependent on the ferroelectric polarization and the Oersted field is small (8 Oersted

at 1.6 mA). More importantly, the smaller critical switching current density for the upward polarization corresponds to a higher spin transmission, in good agreement with the ferroelectric polarization controlled-SOT efficiency study with the ST-FMR technique. The current-induced magnetization switching measurements reinforce the idea that the upward polarization, which corresponds to a right-handed magnon propagation, is favorable for magnon transport.

4.5 Concluding remarks

In summary, I have demonstrated that LSMO/BFO/SIO heterostructures with opposite ferroelectric polarizations can be fabricated by precisely controlling the interface stacking sequence or the termination of the substrate. Such an abrupt change in ferroelectric polarization is shown to have profound influence on the spin transport of BiFeO₃ via antiferromagnetic magnons, due to the magnetoelectric coupling between the ferroelectric ordering and antiferromagnetic ordering. The spintronic manifestation of the ferroelectricity controlled antiferromagnetism is investigated with various techniques (e.g., ST-FMR and direct magnetization switching). An existence of high/low bistable spin transmission states was proven by ST-FMR measurements on samples with opposite ferroelectric polarization, which was further corroborated by the direct magnetization switching measurements.

Chapter 5

Spin transport manipulation: Electric-field

5.1 Background and motivation

Spin current, a flow of spin angular momentum, is the essence in the field of spintronics that exploits the interplay between spin and charge to achieve the next generation beyond complimentary metal-oxide-semiconductor devices[1]. In particular, the efficient spin current generation in various material systems[34, 54, 187] has been an everlasting pursuit to generate efficient spin orbit torques (SOT). Noteworthily, recent progress in low temperature ($< 50\text{K}$) two-dimensional electron gas system[52] and ferroelectric semiconductor GeTe[188] where an extra order parameter is present, has shown successful manipulation of spin current by the applying of an electric field, advancing the realization of spintronic based logic devices one step further. Spin current carried by electrons, however, is associated with electron flow, thus making Joule heating and energy dissipation inevitable. To make matters worse, the efficient spin current generation by large spin Hall effect is usually accompanied by strong spin orbit coupling[189] that confines the spin diffusion length to be several nanometers[164], impeding the delivery of spin information over long distance.

Recently, insulating antiferromagnets have shown to have the capability of carry spin angular momentum by the propagation of spin waves[84-86], by which magnetization switching has been made possible[87, 88, 173], without the flowing of electrons in the antiferromagnets. Moreover, spin transmission in some antiferromagnets with easy-plane[89] or easy-axis[83, 90] anisotropy can occur for the distance up to several micrometers, greatly preserving the spin coherence over long distances. However, despite the advantages magnon current has over spin current, the most effective way employed so

far to modulate the spin transport in antiferromagnets is to use a constant magnetic field or magnetic field pulses, of which up to several tesla might be required depending on the strength of anisotropy of the antiferromagnet[83, 89, 90]. The application of utilizing the aforementioned advantages exhibited by conventional antiferromagnets to practical spintronic devices is hindered by the requirement of energetically prohibitive magnetic fields to control the spin dynamics. The controlling of ferroelectric order by an electric field, however, costs energy typically thousands of times smaller[190] than that of magnetic order, indicating that switching the ferroelectric order as the primary order parameter[91-93, 191] could be a viable path to energy efficient spintronic devices. The use of ferroelectric order as the primary order parameter is allowed by the magnetoelectric mechanism[192], especially in multiferroic materials. Multiferroic materials such as BiFeO_3 [66], in which multiple order parameters are present, shows robust antiferromagnetism, making them potential candidates for spin transport via AFM magnons[193-195]. What makes multiferroic materials stand out for spintronic application is the feasibility of controlling the spin transport with electric fields rather than magnetic fields, due to the intrinsic coupling between the ferroelectric and antiferromagnetic orders. Here I show that beyond antiferromagnets, multiferroic BiFeO_3 exhibits a tunable spin current transmission realized with the switching of the ferroelectric order.

5.2 Fabrication process for polarization dependent spin transmission study

To make ST-FMR and Hall devices, the samples were first spin coated with photoresist (MiR 701), then devices with a $15\ \mu\text{m} \times 10\ \mu\text{m}$ rectangular dimensions ($5\ \mu\text{m} \times 80\ \mu\text{m}$ for the direct magnetization switching experiments) were made by photolithography (Heidelberg MLA 150). A Pi scientific ion miller was used to mill the sample that stopped at the LSMO layer (Fig. 5.1b). The remaining LSMO layer was exposed and used as the bottom electrode in the ferroelectric switching experiments. A voltage was applied between SIO and LSMO, which poles the ferroelectric polarization of BFO to the desired states. Right after this, the remaining LSMO was removed using a H_3PO_4 wet etch ($\text{H}_3\text{PO}_4\text{: H}_2\text{O} = 1\text{: }3$). Finally, electrical contacts were made from 100 nm of Pt with a ground-signal-ground (GSG) geometry for ST-FMR devices, so that when a GSG high frequency probe made into contact with the samples the current travelling through the Pt contacts did not produce a net Oersted field at the sample.



Figure 5.1: General device fabrication process for polarization dependent spin transmission study. **a**, The tri-layer heterostructure consists of LSMO, BFO, and SIO. **b**, Ferroelectric switching configuration. V : voltage applied. **c**, The heterostructure before electrode deposit. **d**, The profile of the device for spin transmission study.

Ferroelectric polarization hysteresis loops and ferroelectric current loops were performed using a commercial Precision multiferroic tester (Radiant technologies) with SrIrO_3 ($\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$) serving as the top (bottom) electrode (Fig. 5.2a). The reversal of the applied electric field on the trilayer is achieved by selectively grounding the top and bottom electrodes. The switching current was recorded as a function of applied voltages. A successful switching of ferroelectric polarization is associated with switching current reaching a maximum (or minimum) at the coercive fields. For 10 nm BFO, a large leaking current was observed, which hinders consistent ferroelectric polarization reversals. As the thickness of the BFO increases, the leakage is mitigated (Fig. 5.2c and Fig. 5.2d). Consistent ferroelectric polarization switching can be achieved for 15 nm and 20 nm BFO, as suggested by the switching current peaks at negative and positive coercive fields.

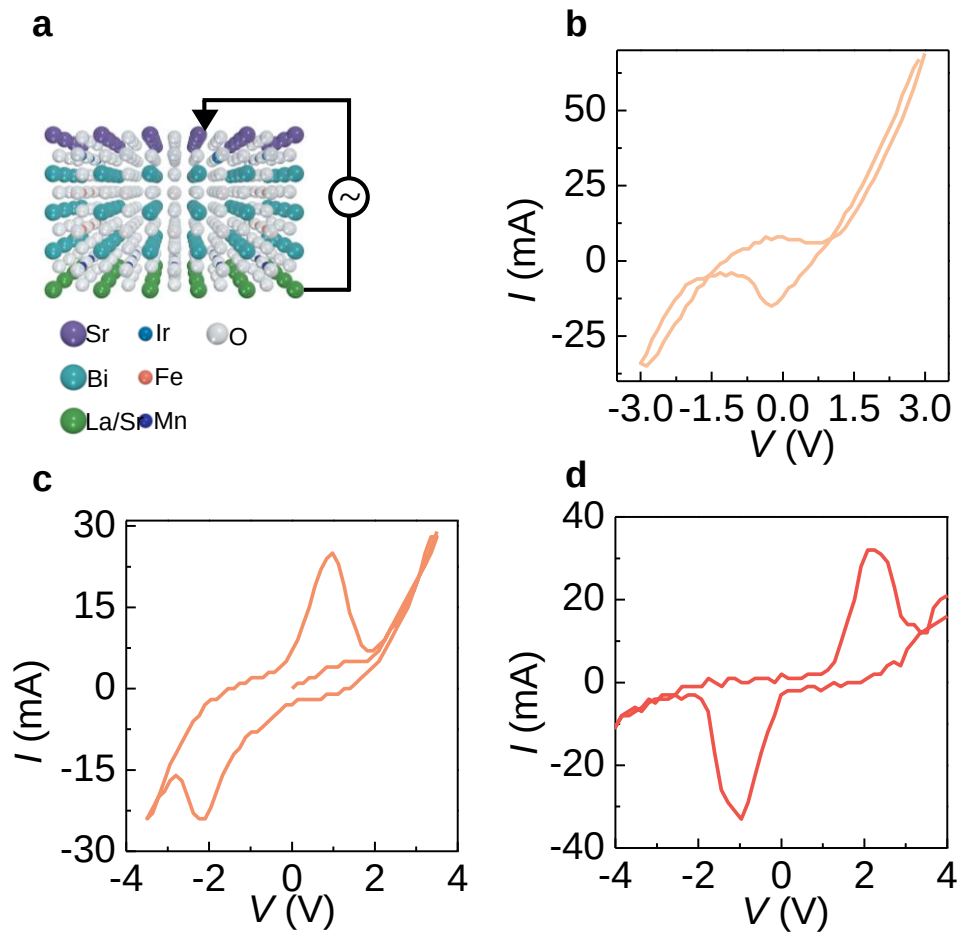


Figure 5.2: Ferroelectric switching of BFO in LSMO/BFO/SIO heterostructure. **a**, A schematic illustration of the switching experiment setup. **b**, **c**, **d**, Switching current as a function of applied voltages for 10 nm, 15 nm and 20 nm BFO respectively.

5.3 Electric-field control of spin transport with ST-FMR

To experimentally explore the controllable chiral-spin transport enabled by the ferroelectric switch in BiFeO₃, I first characterized the strength of spin transmission via magnons in heterostructures of the form LSMO/BFO/SIO. I perform the measurement using a well-established technique, spin torque ferromagnetic resonance (ST-FMR) (Fig. 5.3a), launching the spin current using the large spin Hall effect of SrIrO₃ and detecting the spin torque applied to the magnetic LSMO layer. An example ST-FMR spectrum, composed of both symmetric and anti-symmetric components, is shown (Fig. 5.3b) and indicates the successful detection of the spin currents carried by magnons through the BiFeO₃. To manipulate the orientation of the ferroelectric polarization, an external electric field was applied along the out-of-plane $[0\ 0\ 1]$ to set the ferroelectric polarization of the BiFeO₃ as illustrated (top panel, Fig. 5.3c, in which the top SrIrO₃ and bottom LSMO

layers serve as the two electrodes sandwiching the ferroelectric). Specifically, upward-(downward-) pointing polarization (180° switching) can be achieved by applying negative (positive) voltage larger than the coercivity of the BiFeO_3 on the top SrIrO_3 layer (bottom panel, Fig. 5.3c). After poling the BiFeO_3 in each individual device to the desired polarization state, I then deposited platinum contacts onto the devices (contacting both the SIO and LSMO layers) and performed ST-FMR measurements on the poled devices. By adopting an established analysis method discussed in section 4.4.1, the SOT efficiency for the damping-like (DL) torque (symmetric component) is evaluated and summarized (Fig. 5.3d). I observe that the SOT efficiency can be set at a high value by negative (upward polarization) or at a low value by positive (downward polarization) voltages, indicating the presence of bistable high/low spin transport in BiFeO_3 . It is worthwhile to note that the stability of the ferroelectric polarization reveals another crucial advantage associated with spintronics produced in this manner – non-volatility. By setting the polarization direction, high and low SOT efficiencies of 0.33 and 0.23 can be achieved, showing a high/low ratio of approximately 40%. Furthermore, the ferroelectric polarization-controlled spin transmission in BiFeO_3 is consistent with ST-FMR measurements on samples with different interface terminations in section 4.4.

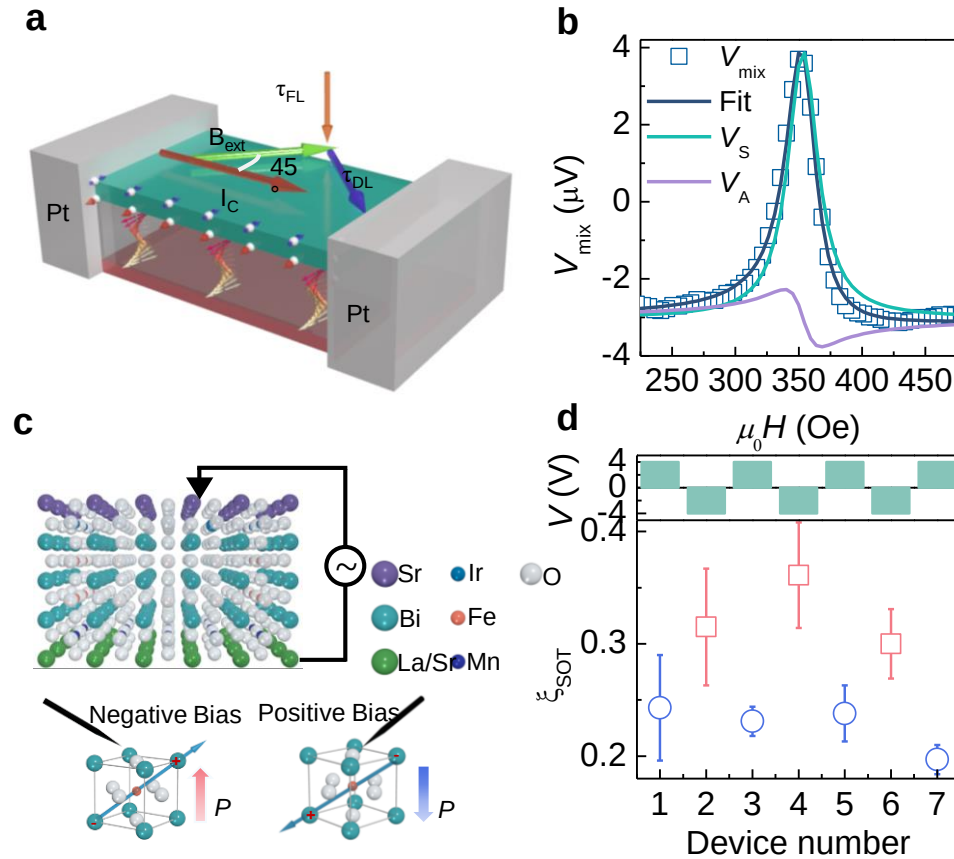


Figure 5.3: Spin transport controlled by ferroelectric polarization in BiFeO_3 . a, A 3D schematic illustration for the ST-FMR measurement. Top: SrIrO_3 , middle: BiFeO_3 , bottom: $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$. I_C represents the radio frequency microwave current, which

flows at a 45° angle with respect to the in-plane external magnetic field represented by B_{ext} . The rotating arrows are a representation of the magnon propagation driven by spin accumulation at the $\text{BiFeO}_3/\text{SrIrO}_3$ interface. b, Measured ST-FMR signal at room temperature for 12 dBm of applied microwave power at 4 GHz for a sample LSMO (20 nm)/BFO (4 nm)/SIO (10 nm) of dimension $25\mu\text{m} \times 50\mu\text{m}$. The lines are Lorentzian fits to the mixing voltage as described in methods (ST-FMR analysis section), showing both symmetric and anti-symmetric components. c, A schematic of a LSMO (20 nm) /BFO (25 nm) /SIO (10 nm) heterostructure subject to an external electric field, along with schematics of the BiFeO_3 ferroelectric polarization under negative and positive biases. d, The SOT efficiency measured with the ST-FMR technique as a function of bias voltages applied beforehand for samples of LSMO (20 nm)/BFO (25 nm)/SIO (10 nm). The open squares (circles) represent the SOT efficiency for the tri-layer with negative (positive) bias voltages applied on the top electrode – SrIrO_3 .

5.4 Switching of a ferromagnet with magnon spin-torque

Previously, it has been shown that the spin-torque carried by magnon current in antiferromagnets is sufficiently large to switch the magnetization of adjacent ferromagnets with perpendicular magnetic anisotropy (PMA). In the previous sections, I have proven that multiferroic BiFeO_3 , where antiferromagnetic order and ferroelectric order coexist, is able to transmit spin current via antiferromagnet magnons; and the resulting spin-torque carried by the antiferromagnet magnons is sufficiently efficient to switch the magnetization of a ferromagnet (LSMO) by 90° in the plane. However, it still remains elusive that whether a deterministic switching of an adjacent ferromagnet with PMA can be realized by this spin-torque at the multiferroics and ferromagnet interface. In this section, the magnetization switching of a PMA ferromagnet (SrRuO_3) by spin-torque in BiFeO_3 is investigated in detail, with an introduction to the previous work relevant to this topic.

5.4.1 Previous work

NiO is an antiferromagnet with biaxial anisotropy and has been shown to be able to transmission spin current generated by either spin Seebeck effect[196] or ferromagnetic resonance[84]. Theoretically, it has been proven that spin transmission in NiO is enabled by the interference of two eigen modes of evanescent spin waves[197]. It became pressing to investigate if the spin-torque arising from NiO is able to achieve magnetization switching of a ferromagnet that is in close proximity to the antiferromagnet. In 2019, Yi Wang et al.[87] reported the first experimental demonstration of magnetization switching with the assistance of spin-torque carried by the magnon current in NiO . They fabricated a $\text{Bi}_2\text{Se}_3/\text{NiO}/\text{Py}$ heterostructure (Fig. 5.4a), in which the magnetization of Py is switched by the spin current that passes through NiO . In their MOKE measurements (Fig. 5.4b), the magnetization of the Py dot by injecting current pulses in the $\text{Bi}_2\text{Se}_3/\text{NiO}$ channel as indicated by the contrast change in the MOKE signal, and the magnetization of Py can be

reversibly switched by positive and negative currents. It has proven that the switching is caused by current induced spin-torque arising from NiO and the spin-torque is sufficiently large to switch the magnetization consistently.

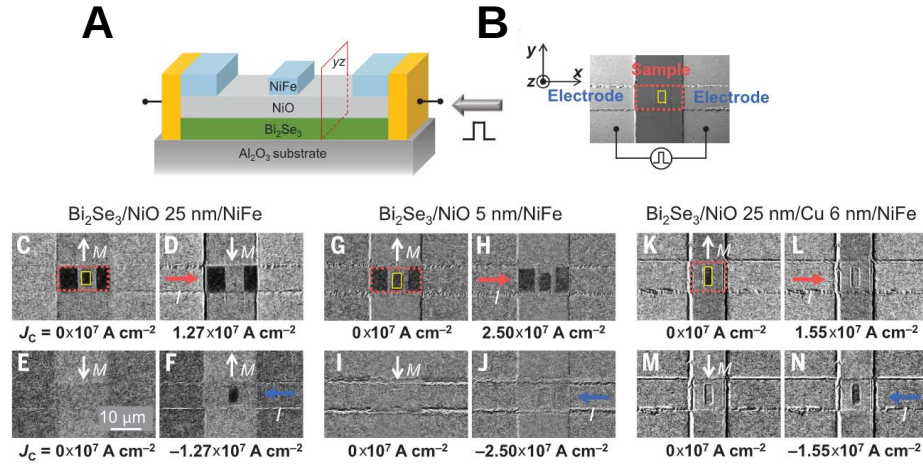


Figure 5.4: Magnetization switching by spin torque carried antiferromagnet magnons in $\text{Bi}_2\text{Se}_3/\text{NiO}/\text{NiFe}$ heterostructure. A, A schematic illustration of $\text{Bi}_2\text{Se}_3/\text{NiO}/\text{NiFe}$ heterostructure for magnetization switching. The island on the center is a small NiFe dot. B, An optical image of the device for magnetization switching study. C to F, MOKE images for magnetization switching driven by magnon torque in $\text{Bi}_2\text{Se}_3/\text{NiO}$ (25 nm)/NiFe by injecting current pulses along +x direction (C and D) or -x direction (E and F) at room temperature. G to J, MOKE images for magnetization switching in $\text{Bi}_2\text{Se}_3/\text{NiO}$ (5 nm)/NiFe. G, H: current pulses along +x direction; I, J: current pulses along -x direction. K to N, MOKE images for magnetization switching in $\text{Bi}_2\text{Se}_3/\text{NiO}$ (5 nm)/Cu (6 nm)/NiFe. K, L: current pulses along +x direction; M, N: current pulses along -x direction. For all MOKE images, dark contrast represents magnetization along +y direction and light contrast represents magnetization along -y direction. Adapted from Ref.[87].

Another important piece of work is reported by Hailong Wang et al.[88], in which they demonstrated the magnetization switching of ferromagnet with PMA by NiO. A heterostructure consisting of Pt/NiO/CoTb, in which CoTb is the ferromagnet with PMA, was fabricated (Fig. 5.5a). The magnetization switching of CoTb is consistently switched as shown in Fig. 5.5c to f. The chirality of the Hall resistance loops is switched upon the switching of the polarity of the external magnetic fields, suggesting that the switching is caused by current induced spin-torque not by either Oersted field or Joule heating. By increasing the thickness of the NiO layer (Fig. 5.5c to f), an increase for critical switching currents and a decrease for the switching percentage are observed, consistent with magnetization switching caused by spin-torque arising from NiO. The demonstration of magnetization switching in antiferromagnetic NiO in turn motivates us to investigate the possibility of the magnetization switching of ferromagnets with PMA by spin-torque arising from multiferroic BiFeO_3 .

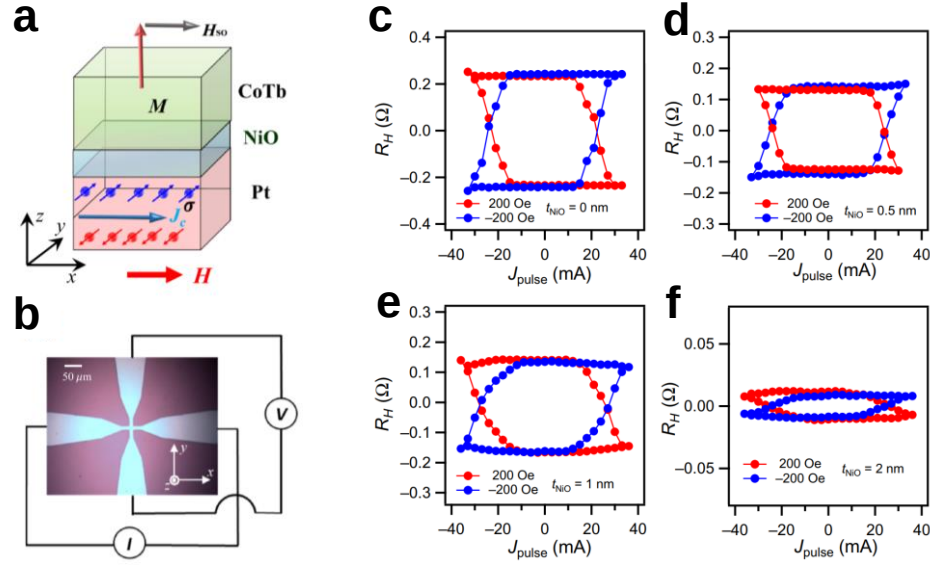


Figure 5.5: Magnetization switching in Pt/NiO/CoTb heterostructure. a, A schematic illustration of the heterostructure for magnetization switching. M is the magnetization along the out-of-plane direction. A small magnetic field is applied in the x direction in order to achieve deterministic switching. b, An optical image of the Hall device for switching measurements. c to f, Anomalous Hall resistance loops as a function of pulsing currents under ± 200 Oe bias field for sample Pt/NiO (x nm)/CoTb, with x being 0, 0.5, 1, and 2 nm. Adapted from Ref.[88].

5.4.2 Magnetization switching with spin-torque in BiFeO₃

A heterostructure consisting of SRO/BFO/SIO was grown on (0 0 1)-oriented SrTiO₃ substrate with TiO₂ termination by RHEED assisted PLD, using the optimized growth conditions summarized in Table 5. Then the magnetization property of the heterostructure was examined with anomalous Hall effect (AHE) measurements. The single Hall cross device consists of two orthogonal channels with the dimension $5 \mu\text{m} \times 10 \mu\text{m}$. During the measurement, a dc current of $100 \mu\text{A}$ was applied and the transverse Hall voltage was measured as a function of the external magnetic field swept in the out-of-plane direction. The dc current flowing through SRO will be spin polarized by the magnetization of SRO and distinctively different Hall resistances can be detected for different magnetization states due to spin dependent scattering of the electrons. Thus, the magnetization orientation in the switching measurements can be unambiguously identified with the anomalous Hall resistance measurements. The AHE at different temperatures is displayed in Fig. 5.4. Sharp hysteresis loops were observed for AHE measurements throughout the temperatures (20 K, 50 K, 70 K, 90 K) (Fig. 5.4) that the measurements were performed at, suggesting good ferromagnetism with PMA at these temperatures. Noteworthy, multi-step magnetization switching behavior was pronounced at these temperatures, indicating the possible existence of magnetization multi-domains of SRO in the SRO/BFO/SIO heterostructure.

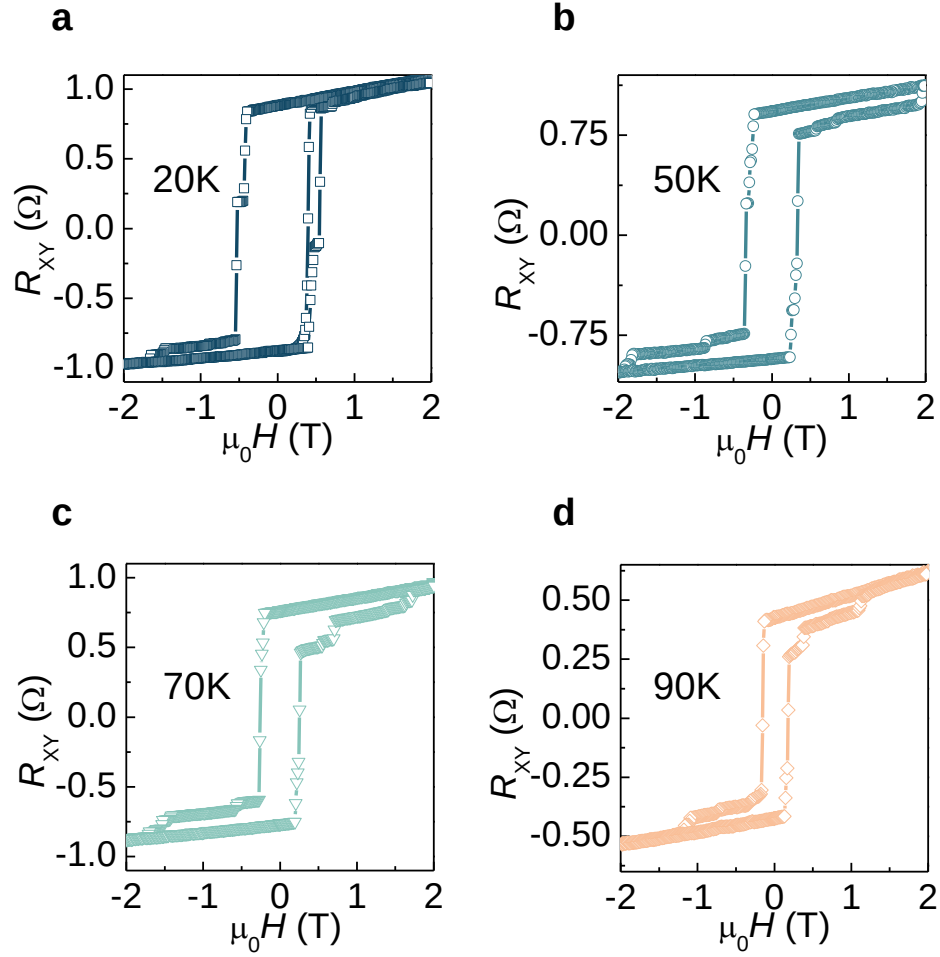


Figure 5.4: Temperature dependent anomalous Hall effect for the sample STO/SRO (7 nm)/BFO (14 nm)/SIO (14 nm). **a, b, c, d**, Anomalous Hall resistance as a function of magnetic field in the out-of-plane direction at 20 K, 50 K, 70 K, and 90 K respectively. 100 μ A of dc current was applied during the measurements.

The spin torque that arises from BiFeO_3 can be directly observed by switching the magnetization of an adjacent ferromagnet with PMA. To investigate this idea, a heterostructure consisting of SRO (7 nm)/BFO (15 nm)/ SIO (14 nm) was prepared for the magnetization switching study (Fig. 5.5a). Pulsing currents at 1000 Hz with 1 ms duration were applied in the y direction, which generates a damping-like effective field B_{DL} in the y direction. To achieve deterministic switching of the magnetization, an assisting magnetic field was applied along the pulsing current direction (y direction) (Fig. 5.5b) to induce a small tilting of the magnetization off the z direction (Fig. 5.5a). Progressively increasing current pulses were applied in the y direction in order to reorient the magnetization to the opposite direction. AHE resistance (R_{XY}) loops for SRO were observed when sweeping current pulses under the application of +1000 Oe and -1000 Oe magnetic fields (Fig. 5.5c and Fig. 5.5d). The magnetization switching occurs at approximately $\pm 2.5 \times 10^6 \text{ A/cm}^2$ (Fig. 5.5c and Fig. 5.5d), comparable to the critical current density observed for SRO/SIO

heterostructure ([116]). The deterministic switching (bidirectional) depending on the pulsing current polarity excludes the possibility that the switching is dominated by Joule heating effect. The switching chirality is changed upon the switching of the polarity of the assisting field (from -1000 Oe to +1000 Oe), consistent with the previous reports. The chirality change is because that upon the switching of the polarity of the external field, the magnetization will be tilted to the opposite direction, which requires currents with opposite polarity to switch it to the other stable state. Importantly, the chirality change by the switching of assisting field polarity confirms that Oersted field that is also generated by the charge current is not the origin of the magnetization switching of SRO.

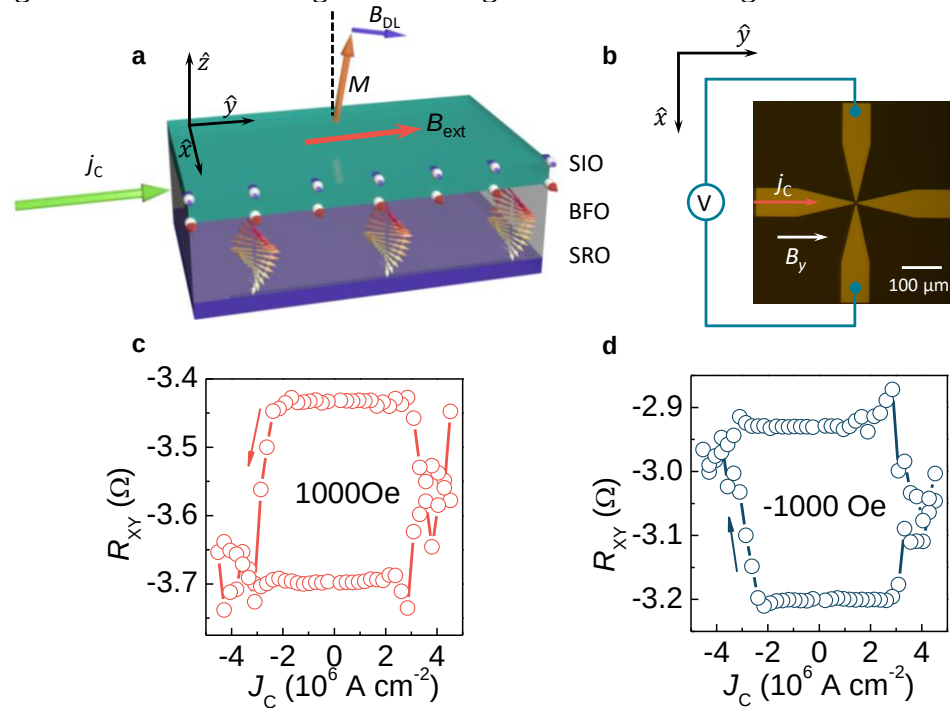


Figure 5.5: Deterministic magnetization switching in the perpendicular direction with spin torque in multiferroic BiFeO₃ for a sample STO/SRO (7 nm)/BFO (14 nm)/SIO (14 nm). **a**, A schematic illustration of the sample stack for magnetization switching. M : magnetic moment of SrRuO₃; B_{DL} : damping like effective field; B_{ext} : in-plane external magnetic field applied to assist the magnetization switching; j_c : pulsing charge current. The rotating arrows are a representation of the magnon current in BiFeO₃. **b**, The device geometry (Hall configuration) for the magnetization switching measurements. An assisting external magnetic field is applied along the pulsing current direction. **c**, **d**, The transverse resistance (anomalous Hall resistance) as a function of pulsing current magnitudes with $B_{ext} = 1000$ Oe and -1000 Oe respectively. During the experiment, pulsing currents of 1000 Hz with 1 ms duration were applied.

5.5 Non-local spin transport controlled by electric-field

I further investigated the experimental demonstration of MESO via electrically controllable chiral-spin transport. A schematic of the nonlocal measurement is provided (Fig. 5.6a); it consists of spin-orbit input and output via parallel source and detector wires, as well as magnetoelectric control. Fig. 5.6b illustrates well-ordered 71° ferroelectric domains, showing the high quality of the BiFeO_3 films. A nonlocal, quasi-static measurement was performed (Fig. 5.6c), varying the magnitude of the electric-field pulse applied in-plane across the channel from negative to positive and back again, and after each pulse measuring (over 100 seconds) non-local voltage signals due to magnon transport between the source and detector wires. I observe a butterfly-shaped and hysteretic response in the magnon current launched by the spin Hall effect (first-harmonic voltage, Fig. 5.6c) and the magnon current launched thermally by the spin Seebeck effect, respectively (second-harmonic voltage, Fig. 5.6d), in which the coercive fields match each other. Figure 5.6e presents measurements of the magnitude of these spin-transport signals as a function of changing the spacing between the source and detector wires from 500 nm to 4 μm . The open squares reflect the magnitude of the signals associated with spin currents launched by the spin Hall effect (first harmonic voltage) and the red circles reflect signals launched thermally by the spin Seebeck effect (second harmonic voltage). Remarkably, the output voltage detected in $\text{BiFeO}_3/\text{SrIrO}_3$ stacks ($\sim 1\mu\text{V}$) is approximately one order of magnitude larger than the counterparts in BiFeO_3/Pt ($\sim 0.1\mu\text{V}$)[198], which is supported by the large spin-torque efficiency of SrIrO_3 previously observed with the ST-FMR measurement[187]. The distance dependence for both types of signals in BiFeO_3 fits well to the expectation for diffusive magnon propagation[199]

$$R_{\text{nl}} = \frac{c}{\lambda} \frac{\exp(d/\lambda)}{1 - \exp(d/2\lambda)}, \quad (5.1)$$

with effective diffusion lengths $\sim 0.17\mu\text{m}$ (first harmonic) and $\sim 0.25\mu\text{m}$ (second harmonic).

Using this result and the spin-torque efficiencies obtained above, I can calculate the inverse-spin-Hall output voltage that would be obtained for a vertical geometry with BiFeO_3 thicknesses of 10 and 15 nm using[200]

$$V_{\text{ISHE}} = \frac{\theta_{\text{SOT}} \lambda_{\text{SD}} \tanh\left(\frac{t_{\text{SIO}}}{2\lambda_{\text{SD}}}\right) L}{t_{\text{SIO}} \sigma_{\text{SIO}}} J_{\text{S}}, \quad (5.2)$$

where I use the values $\theta_{\text{SOT}} = 0.4$ is the spin-torque efficiency, $\lambda_{\text{SD}} = 1.4\text{ nm}$ [140] is the spin-diffusion length of SrIrO_3 , $t_{\text{SIO}} = 10\text{ nm}$ is the thickness, $\sigma_{\text{SIO}} = 1 \times 10^5 \Omega^{-1}\text{m}^{-1}$ is the conductivity, $L = 10\text{ nm}$ is the wire length. The output voltage is on the order of 10 mV, which almost reaches the threshold switching voltage for $\text{BiFeO}_3 \sim 100\text{ mV}$. Our work of spin transport in multiferroics reveals the unexplored chiral-magnon propagation in the presence of ferroelectricity. This observation sheds lights on implementing multiferroics as the potential spin carriers and opens new possibilities of energy efficient magnonic spin-logic devices.

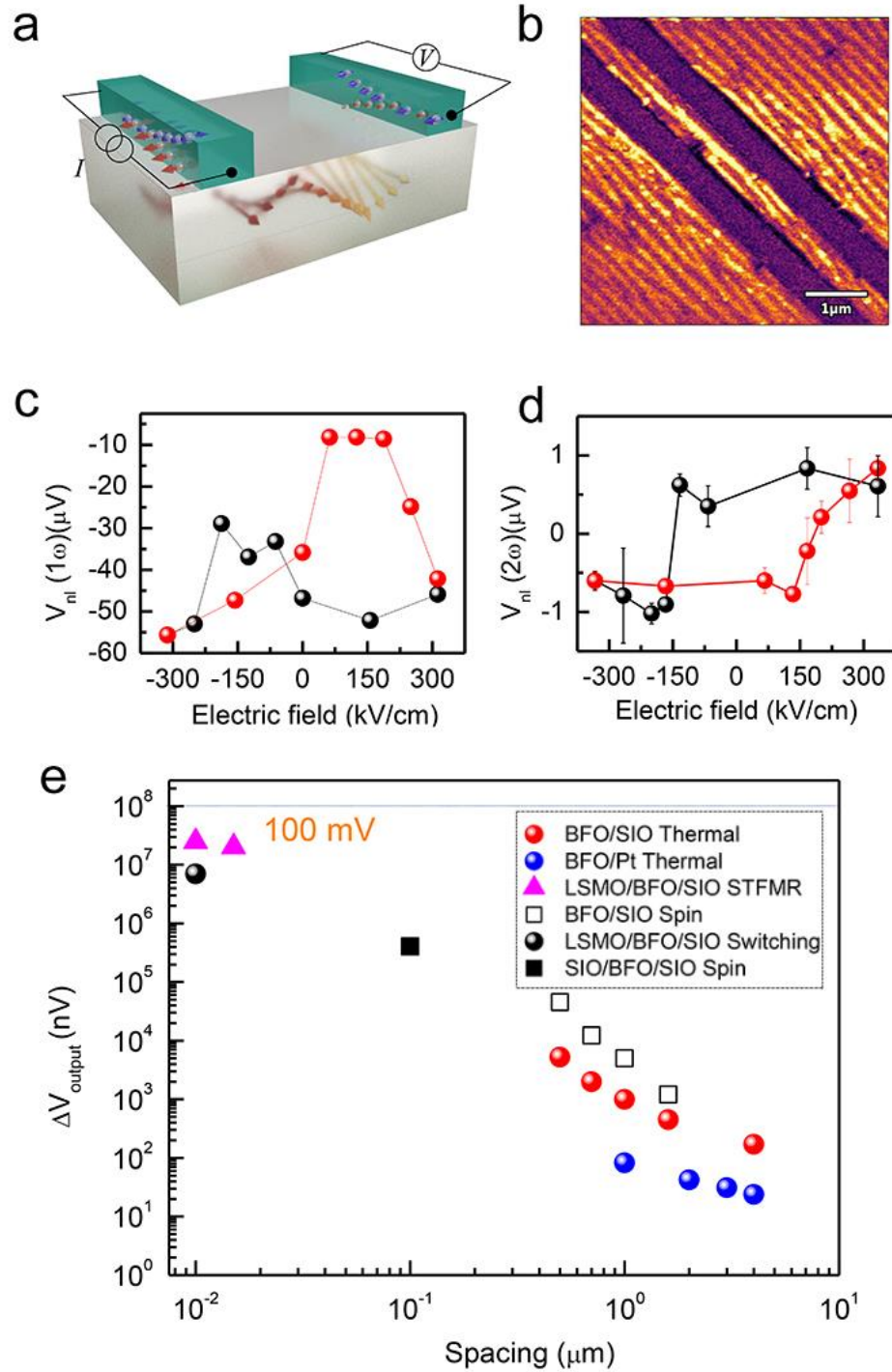


Figure 5.6: Magnetolectric spin-orbit logic based on chiral magnon transport in BiFeO₃. **a**, Schematic of the non-local device formed with a magnetoelectric capacitor on a BiFeO₃ thin film. **b**, Piezo-force microscopy image of the non-local

device structure. The channel width is 0.5 μm . **c**, Hysteresis loop from spin magnon readout in SrIrO_3 with first harmonic measurements for a device with a 500 nm channel width. **d**, Hysteresis loop from thermal magnon readout in SrIrO_3 with second harmonic measurements in a device with a 1 μm channel width. **e**, Output voltage scaling with channel spacing for measurements in nonlocal devices (points for spacings 0.5 μm and above), vertical structure of SIO/BFO/SIO when BFO is 100 nm, and estimates of voltages that could be obtained for vertical transport of magnons through 10 and 15 nm of BiFeO_3 . The power is fixed at 3 mW.

5.6 Conclusion

To summarize, I have shown the ferroelectric polarization-controlled spin transport by electrically manipulating the ferroelectric polarization orientations. The electric field-controlled spin transport in BiFeO_3 is another piece of evidence supporting the existence of bistable high/low spin transmission states controlled by polarization. It is worthwhile to note that the spin torque arising from BiFeO_3 is sufficiently large to deterministically switch the magnetization of a ferromagnet with perpendicular magnetic anisotropy.

Chapter 6

Conclusion and future prospects

With Moore's law approaching its physical limit, the need to design next generation beyond complementary metal-oxide-semiconductor (CMOS) memory and logic devices becomes pressing. Spin-transfer-torque (STT) and spin-orbit-torque (SOT) magnetic-random-access-memory (MRAM) are promising candidates for next generation devices with dimension miniaturization. However, a relative high writing current is required for these devices, hindering the immediate application of spintronics-based memory device into memory technology. In this dissertation I present a systematic study on spin generation and manipulation in complex oxide heterostructures consisting of ferromagnet $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ and SrRuO_3 , multiferroic BiFeO_3 and spin-orbit coupled SrIrO_3 . Reflection high energy electron diffraction assisted pulsed laser deposition was employed for the synthesis of high-quality heterostructures with atomic scale precision. The quality of the heterostructures was examined with various techniques, showing single crystallinity with atomically smooth surface and sharp interfaces. Further spin transport measurements were conducted on the all-oxide heterostructures to reveal superior performance as well as unique physics hosted by these heterostructures.

I first investigate efficient spin orbit torque generation by using heteroepitaxial perovskite oxides driven by the intrinsic similarity in crystal chemistry and structure to design and synthesize epitaxial oxide FM/SOC metal heterostructures as model systems to study the SOT efficiency and novel interface-driven spin-orbitronics. Using ST-FMR, I show that LSMO/SIO heterostructure displays $M_{\text{eff}} > M_s$ and $\xi_{||}=1$, taking advantage of intrinsic band structure and correlated electron phenomena that is revealed with local electronic structure study. I show that precise control over crystallinity, crystal orientation, interface quality, and strain states of the bilayer, allows us to achieve high SOT efficiency with lower current shunting factor. The finding of high SOT efficiency in this all-oxide epitaxial system emphasizes the significant role oxide epitaxy can play as a tool in spintronics. Furthermore, this work suggests a broader set of FM/SOC heterostructures

that can be created using the principles of heteroepitaxy to yield large SOT efficiencies, given the rich diversity of oxide materials.

In addition to efficient spin current generation, the capability of tuning spin current propagation is of equal importance. There are two effective pathways to tune the propagation of spin in BiFeO_3 , which are essentially resorting to the multiferroic nature of BiFeO_3 . One is to precisely control the interface termination with atomic scale precision to achieve opposite spontaneous ferroelectric polarizations. ST-FMR measurements performed on samples with opposite ferroelectric polarization showed distinct spin transmission efficiencies. This observation was later confirmed with the direction switching measurements for samples with opposite ferroelectric polarization. The direct switching measurement not only showed that the spin torque is sufficiently efficient to switch the magnetization of a ferromagnet with in plane anisotropy but also proved the existence of bistable high/low spin transmission states in BiFeO_3 as suggested by the ST-FMR measurements.

The other pathway commonly used to consistently manipulate ferroelectric polarization is to apply electric fields larger than the coercivity of the material to fully reverse the polarization. Here I demonstrated that by careful device design and fabrication ST-FMR measurements were successfully carried out on devices with distinct history of electrical poling. Full reversal of ferroelectric polarization was observed in the ferroelectric switching measurements. The subsequent ST-FMR measurements on those devices showed the same trend for polarization dependent spin transmission as the interface-controlled spin propagation. Additionally, it was shown that the spin torque arising from BiFeO_3 is sufficient to switch the magnetization of a ferromagnet with perpendicular magnetic anisotropy, which is promising for high-density magnetic storage.

Obviously, the role that epitaxy plays in achieving high SOT efficiency is not unique to the $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{SrIrO}_3$ system. Utilizing epitaxy as a powerful tool in spintronics could be a research endeavor that can be pursued to achieve efficient spin current generation. Taking into account epitaxy, careful design of all-oxide heterostructures consisting of spin-orbit coupled materials and ferromagnets can lead to the discovery of all-oxide epitaxial systems with record high SOT efficiency. In addition, more efficient control of spin current with smaller electric field and high rectification ratio is another interesting research direction to pursue. Also, the material candidates for spin current manipulation should be expanded and not limited to multiferroic BiFeO_3 .

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