
Synthesis and Characterization of Oxide Thin Films for Spintronics

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Akashdeep Akashdeep
May 2026

Dedicated to my family and teachers

Abstract

The pursuit of energy-efficient computing platforms requires breakthroughs in spintronics, where information is encoded in electron spin rather than charge. This thesis focuses on novel oxide systems, specifically ruthenium dioxide (RuO_2), a conductive metallic candidate for the emerging altermagnetic phase, which promises compensated magnetic order and strong intrinsic spin splitting, alongside yttrium iron garnet (YIG), the benchmark ferrimagnetic insulator renowned for its ultralow damping.

The central objective is to achieve a verifiable and quantitative understanding of spin-current generation, propagation, and electrical detection in these oxide heterostructures. A fundamental scientific challenge addressed is the stringent requirement to distinguish potential intrinsic phenomena, such as non-relativistic spin splitting effects stemming from crystal symmetry, from pervasive extrinsic contributions arising from interfaces, defects, or dimensionality.

First, foundational structural quality was established through controlled physical vapor deposition, monitored *in situ* using reflection high-energy electron diffraction, while *ex situ* x-ray diffraction provided further verification of crystallographic phase purity and structural coherence. Second, quantitative interface-sensitive magnetotransport experiments (ADMR in RuO_2/Py bilayers) were performed, confirming the dominance of interface effects. Third, localized magnetic order was probed using depth-resolved low-energy muon spin rotation ($\text{LE-}\mu\text{SR}$), enabling nanometer-scale distinction between surface and bulk properties. The optimized epitaxial $\text{RuO}_2/\text{TiO}_2$ films also served as a platform for collaborative ARPES, XMCD/XPS, and optical experiments. These studies reported signatures interpreted as consistent with altermagnetic order in RuO_2 . Finally, the YIG/Pt(Ta) magnetotransport experiments successfully verified the sign dependence of the Hall signal, validating the reliability of the applied analysis framework for extracting spin transport parameters.

The integrated experimental data yield critical insights. $\text{LE-}\mu\text{SR}$ measurements revealed that static, inhomogeneous magnetic order in RuO_2 thin films is confined predominantly to the near-surface region, suggesting an extrinsic origin, likely due to

dimensionality or defect effects, rather than a bulk altermagnetic phase. Correspondingly, transport analysis demonstrated that interface-generated spin currents, arising from localized spin-orbit scattering at the RuO_2/Py interface, predominantly govern the macroscopic magnetoresistance response, effectively masking potential subtle bulk non-relativistic spin splitting signals.

These findings clarify that, in the present generation of oxide heterostructures, macroscopic spintronic functionality is chiefly determined by extrinsic interfacial spin dynamics. While RuO_2 is confirmed as a robust, metallic platform for spin injection, its utility hinges on meticulous interface and stoichiometry control to either optimize these dominant interface-generated spin currents or successfully stabilize the intrinsic altermagnetic phase. The challenge is underscored by evidence suggesting the lack of magnetic moment in bulk RuO_2 . The analytical methodologies and conceptual framework developed in this work pave the way for future studies focusing on epitaxial strain engineering and ultrafast dynamics, accelerating the path toward energy-efficient, oxide-based spintronics devices.

Zusammenfassung

Das Streben nach energieeffizienten Rechenplattformen erfordert grundlegende Durchbrüche in der Spintronik, bei der Informationen nicht in der elektrischen Ladung, sondern im Elektronenspin kodiert werden. Diese Arbeit konzentriert sich auf neuartige Oxid-Systeme, insbesondere Rutheniumdioxid (RuO_2), ein leitfähiges metallisches Materialsystem für die neu entstehende altermagnetische Phase, das eine kompensierte magnetische Ordnung und ein starkes intrinsisches Spin-Splitting verspricht, sowie Yttrium-Eisen-Granat (YIG), den ferrimagnetischen Isolator, der für seine extrem geringe Dämpfung bekannt ist.

Das zentrale Ziel besteht darin, ein überprüfbares und quantitatives Verständnis der Erzeugung, Ausbreitung und elektrischen Detektion von Spinströmen in diesen Oxid-Heterostrukturen zu erlangen. Eine zentrale wissenschaftliche Herausforderung besteht in der eindeutigen Unterscheidung potenziell intrinsischer Phänomene, wie nichtrelativistische Spinaufspaltungseffekte aufgrund der Kristallsymmetrie, von allgegenwärtigen extrinsischen Beiträgen zu unterscheiden, die durch Grenzflächen, Defekte oder Dimensionalität entstehen.

Zunächst wurde die grundlegende strukturelle Qualität durch kontrollierte physikalische Gasphasenabscheidung hergestellt, die *in situ* mittels Reflektions-Hochenergie-Elektronenbeugung überwacht wurde, während *ex situ* Röntgenbeugung eine weitere Überprüfung der kristallographischen Phasenreinheit und strukturellen Kohärenz ermöglichte. Zweitens wurden quantitative, grenzflächenempfindliche Magnetotransportexperimente (ADMR in RuO_2/Py -Doppelschichten) durchgeführt, die eindeutig die Dominanz von Grenzflächeneffekten bestätigten. Drittens wurde die lokalisierte magnetische Ordnung mit Hilfe der tiefenaufgelösten Niedrigenergie-Myonen-Spinrotation ($\text{LE-}\mu\text{SR}$) untersucht, wodurch eine Unterscheidung zwischen Oberflächen- und Volumeneigenschaften im Nanometerbereich möglich war. Die optimierten epitaktischen $\text{RuO}_2/\text{TiO}_2$ -Filme dienten auch als Plattform für gemeinsame ARPES-, XMCD/XPS- und optische Experimente. Diese Studien berichteten über Signaturen, die als konsistent mit der altermagnetischen Ordnung in RuO_2 interpretiert wurden. Schließlich

konnten die Magnetotransportexperimente mit YIG/Pt(Ta) die Vorzeichenabhängigkeit des Hall-Signals erfolgreich verifizieren und damit die Zuverlässigkeit des angewandten Analyseframeworks zur Extraktion von Spintransportparametern bestätigen.

Die integrierten experimentellen Daten liefern wichtige Erkenntnisse. LE- μ SR-Messungen ergaben, dass die statische, inhomogene magnetische Ordnung in RuO₂-Dünnschichten überwiegend auf den oberflächennahen Bereich beschränkt ist, was auf einen extrinsischen Ursprung hindeutet, der wahrscheinlich auf Dimensionalitäts- oder Defekteffekte zurückzuführen ist und nicht auf eine antiferromagnetische Phase im Volumen. Entsprechend zeigte die Transportanalyse, dass durch die Grenzfläche erzeugte Spinströme, die aus lokalisierter Spin-Bahn-Streuung an der RuO₂/Py-Grenzfläche entstehen, die makroskopische Magnetowiderstandsreaktion überwiegend bestimmen und dadurch potenzielle subtile nichtrelativistische Spin-aufspaltungssignale aus dem Volumen effektiv überdecken.

Diese Ergebnisse verdeutlichen, dass in der aktuellen Generation von Oxid-Heterostrukturen die makroskopische spintronische Funktionalität hauptsächlich durch die extrinsische Spin-Dynamik an den Grenzflächen bestimmt wird. Während RuO₂ als robuste, metallische Plattform für die Spin-Injektion bestätigt ist, hängt seine Verwendbarkeit von einer sorgfältigen Kontrolle der Grenzflächen und der Stöchiometrie ab, um entweder diese dominanten, an den Grenzflächen erzeugten Spinströme zu optimieren oder die intrinsische altermagnetische Phase erfolgreich zu stabilisieren. Die Herausforderung wird durch Hinweise auf das Fehlen eines magnetischen Moments in RuO₂-Volumenmaterial unterstrichen. Die in dieser Arbeit entwickelten analytischen Methoden und konzeptionellen Rahmenbedingungen ebnen den Weg für zukünftige Studien, die sich auf kontrollierte epitaktische Verspannungen und ultraschnelle Dynamik konzentrieren und damit den Weg zu energieeffizienten, oxidbasierten Spintronik-Bauelementen ebnen.

Nomenclature

Acronyms / Abbreviations

<i>ADMR</i>	Angle-Dependent Magnetoresistance
<i>AFM</i>	Antiferromagnet
<i>AHE</i>	Anomalous Hall Effect
<i>AHMR</i>	Anomalous Hall Magnetoresistance
<i>AMR</i>	Anisotropic Magnetoresistance
<i>ARPES</i>	Angle-Resolved Photoemission Spectroscopy
<i>ASSE</i>	Altermagnetic Spin Splitting Effect
DFT+U	Density Functional Theory with Hubbard correction
DMFT	Dynamical Mean-Field Theory
<i>DSH-AHE</i>	Double Spin Hall Anomalous Hall Effect
<i>DSH-HE</i>	Double Spin Hall Hanle Effect
<i>FM</i>	Ferromagnet
GGG	Gadolinium Gallium Garnet
<i>GSE</i>	Geometric Size Effect
<i>HM</i>	Heavy Metal
<i>HMR</i>	Hanle Magnetoresistance
<i>iSHE/ISHE</i>	Inverse Spin Hall Effect

<i>LEM</i>	Low-Energy Muons
<i>LEM-μSR</i>	Low-Energy Muon Spin Rotation/Relaxation
<i>LM</i>	Light Metal
<i>MCD</i>	Magnetic Circular Dichroism
<i>MCDAD</i>	Magnetic Circular Dichroism in the Angular Distribution
<i>MI</i>	Magnetic Insulator
<i>MR</i>	Magnetoresistance
<i>μSR</i>	Muon Spin Rotation/Relaxation
<i>OHE</i>	Ordinary Hall Effect
<i>OMR</i>	Ordinary Magnetoresistance
<i>OREMR</i>	Orbital Rashba–Edelstein Magnetoresistance
<i>PCB</i>	Printed Circuit Board
<i>PLD</i>	Pulsed Laser Deposition
<i>PSI</i>	Paul Scherrer Institute
<i>PVD</i>	Physical Vapor Deposition
Py	Permalloy
<i>REMR</i>	Rashba–Edelstein Magnetoresistance
<i>RHEED</i>	Reflection High-Energy Electron Diffraction
SARPES	Spin- and Angle-Resolved Photoemission Spectroscopy
<i>SH-AHE</i>	Spin Hall Anomalous Hall Effect
<i>SHE</i>	Spin Hall Effect
<i>SH-HE</i>	Spin Hall Hanle Effect
<i>SMR</i>	Spin Hall Magnetoresistance

<i>SμS</i>	Swiss Muon Source
<i>SOC</i>	Spin-Orbit Coupling
<i>SOT</i>	Spin-Orbit Torque
<i>SP-STM</i>	Spin-Polarized Scanning Tunneling Microscopy
<i>SSE</i>	Spin Seebeck Effect
<i>SSH-AHE</i>	Single Spin Hall Anomalous Hall Effect
<i>SSH-HE</i>	Single Spin Hall Hanle Effect
<i>SST</i>	Spin-Splitter Torque
<i>TF</i>	Transverse Field
THz	Terahertz
<i>TR-MOKE</i>	Time-Resolved Magneto-Optic Kerr Effect
<i>UHV</i>	Ultra High Vacuum
<i>XMCD</i>	X-Ray Magnetic Circular Dichroism
<i>XRD</i>	X-Ray Diffraction
<i>XRR</i>	X-Ray Reflectivity
<i>YIG</i>	Yttrium Iron Garnet

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Chapter 1

Introduction

Modern information processing faces rising energy costs as device dimensions shrink and interconnect densities grow. Spintronics addresses this pressure by routing and manipulating information with spin currents, aiming to reduce dissipation while retaining electronic speed and scalability. A central enabler is the interconversion of charge and spin in conductors with strong spin–orbit coupling. The spin Hall effect (SHE) generates a transverse spin current from a longitudinal charge current, and the inverse spin Hall effect (ISHE) converts a spin current back into a measurable voltage; together, these processes underpin many spin-transport architectures [1, 2].

Beyond ferromagnets, the materials palette now includes antiferromagnets (AFMs) and symmetry-defined phases such as altermagnets (AMs) [3]. AFMs offer robustness to stray fields and ultrafast magnetic dynamics, but their staggered order parameter (Néel vector) often couples weakly to macroscopic probes, complicating electrical read-out and control. For background on antiferromagnetic spintronics and broader context, see [4, 5]. AMs, by contrast, have been proposed to host momentum-dependent spin splitting without a net magnetization, raising the possibility of ferromagnet-like transport signatures in compensated systems. These ideas sharpen a practical challenge: which observables are genuinely symmetry-selective, and how can measurements avoid confounding factors that are ubiquitous in thin films?

Ruthenium dioxide (RuO_2) provides a focused setting for these questions. It is a correlated, conductive oxide in the rutile structure, placing it at the intersection of symmetry arguments, interfacial engineering, and oxide stability. Bulk indicators of itinerant antiferromagnetism shifted perceptions of RuO_2 beyond a simple Pauli paramagnet [6], yet thin-film investigations have yielded mixed conclusions about its magnetic ground state. Surface- and depth-sensitive spectroscopies, including spin- and angle-resolved photoemission (SARPES), report band structures that can be compatible with a nonmagnetic state, without clear momentum-dependent spin splitting [7]. At the same time, some transport signatures in films exhibit symmetry patterns that can be interpreted within band-splitting frameworks [8]. Reconciling these views requires care, because measurement weight functions differ: techniques such as SARPES emphasize the near-surface region, whereas transport averages over the stack and interfaces.

Interfaces themselves complicate the attribution of spin signals. Broken inversion symmetry and interfacial spin–orbit coupling can generate interface-generated spin currents (IGSCs), which may be comparable in magnitude to bulk-generated spin currents and can exhibit distinct polarization rules [9]. In multilayers, the measured response therefore mixes bulk conversion, interfacial generation and filtering, and

proximity effects. Disentangling these contributions is central to any assignment of “bulk” properties in a thin-film context. This motivates two complementary needs: structural control that reduces extrinsic variability, and measurement strategies that provide depth selectivity or external benchmarks.

Depth-resolved probes can address the first need by isolating near-surface behavior from deeper regions. Low-energy muon spin rotation/relaxation (LE- μ SR) offers nanometer-scale control of implantation depth and high sensitivity to static or slowly fluctuating magnetic fields, enabling depth profiles of magnetic textures without relying on global transport proxies. Because thin films can host reconstructed terminations, vacancies, strain variations, and other defect landscapes, a depth-sensitive local probe helps distinguish surface-localized magnetism from genuine bulk order. Complementary transport measurements then test whether electrical signatures align with depth profiles under controlled structural conditions.

A second strand relies on established benchmarks that constrain sign and scaling conventions. The archetypal yttrium iron garnet/metal platform (YIG/Pt) remains a standard for calibrating spin pumping and ISHE and for modeling spin Hall magnetoresistance (SMR) [10, 11]. A 2016 theory further connected transverse voltage signals in such bilayers to a nonlocal anomalous Hall effect (nonlocal AHE), whose sign tracks the metal’s spin Hall angle θ_{SH} [12]. Comparing Pt and Ta, with opposite spin Hall angles, therefore provides a cross-check that is independent of YIG specifics and helps to constrain interpretations used elsewhere in this thesis.

In light of these considerations, the present work is organized around three guiding questions. First, in RuO₂/Permalloy (Py) heterostructures, do bulk conversion mechanisms or interface-generated spin currents dominate the observed magnetotransport responses? Second, do RuO₂ thin films host surface-localized magnetism, and how is any such magnetism distributed with depth as probed by LE- μ SR? Third, how do benchmarks based on YIG with Pt or Ta (YIG/Pt(Ta)) calibrate sign conventions and test the mechanisms invoked to interpret spin–charge interconversion and Hall signal? These questions define the scope and success criteria for the remaining chapters.

The scope of the thesis is limited to epitaxial thin films and bilayers where materials and interfaces can be systematically tuned. The primary stacks are RuO₂ films (thicknesses of tens of nanometers) on single-crystal substrates and RuO₂/Py bilayers, alongside reference YIG/Pt(Ta) bilayers. Measurements span room temperature to cryogenic regimes. The work focuses on magnetotransport and local magnetism rather than device engineering or CMOS integration. Outside the scope are ultrafast (THz) dynamics, atomistic theory beyond literature guidance, and claims regarding

bulk single-crystal RuO_2 beyond what is directly supported by cited studies. These boundaries manage expectations and guide interpretation throughout Chs. 4–7.

Within this scope, the thesis makes four contributions, mapped to the central chapters. First (Ch. 4), it reports RuO_2 growth windows and surface-quality control using in-situ reflection high-energy electron diffraction (RHEED) and ex-situ structural probes, establishing a reproducible baseline for subsequent measurements. Second (Ch. 5), it analyzes magnetotransport in RuO_2/Py using a framework that explicitly accounts for interface-generated spin currents and bulk conversion, building on theoretical descriptions of interfacial spin–orbit effects [9]. Third (Ch. 6), it employs LE- μSR to obtain depth profiles of magnetic fields in RuO_2 films, with the goal of distinguishing surface-localized magnetism from deeper responses. Fourth (Ch. 7), it tests, in YIG/Pt(Ta), a sign prediction for nonlocal AHE [12], providing an external yardstick that constrains interpretation elsewhere.

Methodologically, the approach combines thin-film synthesis with in-situ and ex-situ characterization and complementary electrical and local magnetic measurements. RuO_2 and YIG/Pt(Ta) heterostructures are fabricated by sputtering or pulsed-laser deposition on lattice-matched substrates. In-situ RHEED tracks epitaxial growth modes; ex-situ probes assess crystallinity and surface morphology. Lithographed Hall bars enable dc magnetotransport, including angular protocols and thickness series. LE- μSR employs tunable implantation energies to achieve nanometer-scale depth resolution. Data interpretation uses drift–diffusion modeling that incorporates boundary conditions appropriate to interface-generated spin currents and SMR formalisms.

Because interpretations depend on disentangling multiple contributions, a brief note on interfaces is warranted. In the presence of strong interfacial spin–orbit coupling, broken inversion symmetry at a boundary can generate spin currents whose magnitude and polarization do not simply mirror bulk conversion. Neglecting such terms risks misattributing interface-driven phenomena to exotic bulk mechanisms, especially in conductive oxides where correlated behavior and structural defects can modulate near-surface electronic states [9, 13–17]. The combined strategy, structural control, depth selectivity, and external benchmarks, seeks to minimize such ambiguities without presupposing a specific magnetic ground state for RuO_2 .

The remainder of the thesis is organized as follows. Chapter 2 introduces essential concepts in magnetism and spintronics, and summarizes materials considerations relevant to oxides. Chapter 3 details film growth, structural characterization, transport protocols, LE- μSR methodology, and modeling approaches. Chapter 4 reports RHEED-guided epitaxy of RuO_2 and establishes growth/strain windows. Chapter 5 ex-

amines magnetotransport in RuO_2/Py in a framework that weighs interface-generated and bulk mechanisms [9]. Chapter 6 presents LE- μSR depth profiles of RuO_2 thin films and discusses implications for surface versus bulk magnetism. Chapter 7 uses YIG/Pt(Ta) to test a sign prediction for nonlocal AHE and to calibrate spin–charge interconversion models [10–12]. Chapter 8 summarises insights and outlines directions for future interface engineering in oxide spintronics.

Chapter 2

Description of magnetism, spintronics and materials

This chapter starts with a brief discussion of the origin of magnetism, characterization of magnetic materials, and magnetic energy terms, which dictate the magnetic state of the materials. Much of the foundational understanding of magnetism presented in this thesis is based on the comprehensive treatments found in standard solid-state physics and magnetism texts, such as those by Ashcroft and Mermin [18], Kittel [19], Blundell [20], and Cullity [21]. While the original insights and developments belong to the primary literature, these textbooks serve as well-accepted compilations of the theoretical framework, and are cited here collectively to avoid fragmentary referencing of every individual contribution. We will also discuss the material of particular interest and its exciting properties.

2.1 Overview of magnetism

Magnetism has always found its way to us either due to its fundamental intricacies or the applicability of devices in day-to-day life. This interest initiates with discovering the first known magnetic material, magnetite (Fe_3O_4), which attracts iron. The first applicability of magnetic materials as a needle to navigate with the help of the earth's magnetic field was back in the 12th century. Research on the magneto-static in the 19th century was led by Ampere Law, Biot-Savart Law, and Gauss's law for magnetism which helped to understand the current induced magnetic field and its dipolar nature.

Otherwise, magnetism in materials has always been under extensive research. This ample research leads to an understanding that magnetism in materials results from charge particles carrying angular momentum. This angular momentum could either be orbital or intrinsic quantum-mechanical spin momentum. The equation 2.1 gives the orbital magnetic moment (μ_l) of a charged particle moving around the nucleus with orbital angular momentum (l), where e is the charge of the electron, m_e is the mass of the electron. The proportionality factor g relating the angular momentum to the magnetic moment is known as gyromagnetic ratio. Due to its heavier mass than the electrons, the nucleus contributes very little to matter's magnetism, which is also explained by equation 2.1.

$$\mu_l = (\text{current})(\text{area of loop}) = \mathbf{I} \cdot \mathbf{A} = -\frac{e}{2m_e} \mathbf{l} \quad (2.1)$$

The orbital angular momentum of the electron is quantized in the units $h/2\pi$ in a particular direction (say z-axis), where h is Planck's constant. Thus a quanta value of the orbital moment in a particular direction is given by equation 2.2,

$$\mu_{z(l)} = \frac{eh}{4\pi m} = \mu_{z(s)} = \mu_B \quad (2.2)$$

The quantum mechanical description of the electron's intrinsic angular momentum is called *spin*, which gives rise to the electron's spin magnetic moment (μ_s). The electron's spin angular momentum has a z -component of $\pm h/4\pi$, which is half of the quanta of orbital angular momentum. The g -factor (gyromagnetic ratio) relating magnetic moment to angular momentum is $g \approx 2$ for spin angular momentum and $g = 1$ for orbital angular momentum. Thus, despite the spin angular momentum being half the orbital quanta, the magnitude of the fundamental spin magnetic moment equals the quanta of orbital magnetic moment (2.2). This fundamental magnitude of magnetic moment is known as the Bohr magneton (μ_B).

The effective angular momentum is the vector sum of the orbital and spin momentum of electron clouds in the atom and it decides the net magnetic moment and, depending on how these magnetic moments of individual atom interacts, decides the magnetic state of a material. The macroscopic magnetic property of any material is then decided by the relative orientation of the total magnetic moment of different domains. Magnetization ($\mathbf{M}$) as the net magnetic moment (μ) per unit volume (V) is the macroscopic parameter to characterize the magnetic materials. The response of the magnetic material in the presence of an applied magnetic field ($\mathbf{H}$) is called the magnetic flux or magnetic induction ($\mathbf{B}$) and is given by

$$\mathbf{B} = \mu_0(\mathbf{H} + \mathbf{M}) = \tilde{\mu}\mathbf{H} \Rightarrow \tilde{\mu} = \mu_0(1 + \tilde{\chi}) \quad (2.3)$$

where $\tilde{\chi}$ and $\tilde{\mu}$ are the magnetic susceptibility tensor and the magnetic permeability tensor, respectively.

2.2 Types of magnetism

All the materials can be classified magnetically. It is called diamagnetic in the case of a material with no permanent magnetic moment. The material with a permanent magnetic moment can be differentiated by the type of interaction between magnetic moment or no interaction. Here this section describes the diamagnetism, paramagnetism, ferromagnetism, antiferromagnetism and altermagnetism.

2.2.1 Diamagnetism

The net magnetic moment of electrons in an atom nullifies due to an equally opposite balance in spin angular momentum and orbital angular momentum, a case of atoms with filled shells. Hence, the net effective magnetic moment of the material is zero. When we put such material under a magnetic field, the angular momentum of each atom will precess around the field with Larmor frequency, which means each atom has effective angular momentum along the magnetic field. This induced angular momentum will give us a net magnetic moment from each atom in the opposite direction to the field. Hence, the negative magnetic susceptibility. This magnetism is the most ubiquitous and dwarfed form of magnetism. Hence becomes negligible in the presence of other magnetism forms.

2.2.2 Paramagnetism

The atom's net magnetic moment is maintained due to an imbalance between its spin and orbital angular momentum in the case of partially filled shells. Two energy terms in this material determine the specimen's net magnetization or its relationship to temperature when subjected to a magnetic field. First is the Zeeman energy, which tries to align the individual magnetic moment along the field to reduce the system's energy, and the second is the thermal energy, which tries to randomize the individual magnetic moment. Hence, effectively a positive magnetic susceptibility. So, the Boltzmann distribution of magnetic moments with respect to the applied field at equilibrium temperature helps us calculate the system's magnetization. Quantum theory of paramagnetism by considering the quantization of total angular momentum of atom about the axis of external field fits better to the results of the experiments than Langevin theory which considers the continuous distribution of the angular momentum about the axis of the external field. The magnetic susceptibility of such a system is inversely proportional to the system's temperature T or $(T - \theta)$, called Curie Law (eq. 2.4) or Curie-Weiss law (eq. 2.5), where value of θ is proportional to strength of interaction between magnetic moments due to the molecular-Weiss field and C is the Curie constant.

$$\chi = \frac{C}{T} \quad (2.4)$$

$$\chi = \frac{C}{T - \theta} \quad (2.5)$$

2.2.3 Ferromagnetism

The presence of exchange interaction between the magnetic atoms mediated by the electrons gives a favorable energy state for the relative orientation of magnetic moments. The exchange interaction is a quantum mechanical phenomenon arising due to the symmetry properties of wavefunctions of electrons and the Pauli exclusion principle. Specifically, in ferromagnets, the exchange interaction leads to parallel alignment of neighboring electron spins, which minimizes the energy of the system. Ferromagnetism explicitly breaks time-reversal symmetry because the state with aligned spins and net magnetization is not invariant under reversal of spin direction. This symmetry breaking is crucial, leading to unique physical properties such as hysteresis and remanence observed in ferromagnetic materials.

Weiss molecular field theory: This theory goes by the idea of the presence of an internal molecular magnetic field (H_m) in the solid, which is proportional to the magnetization of the solid itself. M being a consequence of H and H_m being a consequence of M . Here, H and H_m work together to saturate the material. This theory explains the temperature dependence of saturation magnetization and the paramagnetism with the difference of magnitude of the molecular field. Above a critical temperature known as the Curie temperature (T_C), thermal energy disrupts the alignment, causing ferromagnetic order to vanish, transitioning the material to a paramagnetic state. Below T_C , the molecular field stabilizes the spontaneous magnetization, enabling long-range ferromagnetic order. The limitation to this theory is that it cannot match the experimentally observed magnetic moment of atoms and account for its different values above and below T_C .

Band theory: Band theory provides a deeper quantum mechanical understanding of ferromagnetism by considering electron bands in momentum space (k-space). This theory considers the occupation of electrons in the partially filled valence band with a high density of states. This high density of states makes exchange interaction strong enough to create an imbalance of spins in the band. Hence, spin-dependent splitting of electronic bands. This spin-dependent energy splitting facilitates spontaneous spin polarization and sustained ferromagnetic order. Band theory thus complements Weiss theory by providing a microscopic foundation for understanding electronic and magnetic phenomena in ferromagnets and gives an excellent match for magnetic moment per atom for Fe, Co, and Ni.

2.2.4 Antiferromagnetism

Antiferromagnetism is characterized by magnetic moments that align antiparallel to magnetic atoms, resulting in no net macroscopic magnetization. Like ferromagnetism, antiferromagnetism arises from quantum mechanical exchange interactions, but exchange interactions promote antiparallel alignment to lower the total energy. Antiferromagnetism preserves macroscopic time-reversal symmetry because reversing all spins leaves the magnetic state unchanged due to the symmetric spin configuration.

Weiss Molecular Field Theory for Antiferromagnetism: Analogous to ferromagnetism, antiferromagnetism can be described using Weiss theory by introducing two sublattices with opposing magnetizations. If the two magnetic sublattices have unequal magnetic moments, the resulting ordering is called ferrimagnetism rather than antiferromagnetism. Each sublattice generates an internal molecular field that stabilizes the antiparallel spin arrangement. Antiferromagnets exhibit a critical temperature known as the Néel temperature (T_N). Above T_N , thermal fluctuations overcome the molecular field, transitioning the material into a paramagnetic state. Below T_N , the sublattice magnetizations stabilize the antiferromagnetic order, maintaining zero net magnetization.

Band Theory for Antiferromagnetism: Band theory offers additional insight into antiferromagnetic ordering by examining electronic bands in k-space. Unlike ferromagnetic materials, antiferromagnetic systems exhibit symmetric band splitting with equal occupancy of spin-up and spin-down electrons, resulting in no net spin polarization. Thus, antiferromagnetic materials possess distinctive electronic properties, including symmetrical and spin-degenerate electronic bands. The symmetry of antiferromagnetic materials' band structure sets the stage for exploring phenomena where subtle alterations to symmetry conditions lead to new magnetic states.

2.2.5 Altermagnetism

Altermagnetism represents a novel magnetic state distinct from traditional ferromagnetic and antiferromagnetic orderings. Unlike antiferromagnets, altermagnets exhibit significant spin splitting and anisotropy within their electronic band structure despite having no net macroscopic magnetization. While antiferromagnetism preserves macroscopic time-reversal symmetry due to symmetric spin arrangements, altermagnetism explicitly breaks this symmetry in momentum space (k-space) through highly anisotropic and direction-dependent spin-split band structures [22].

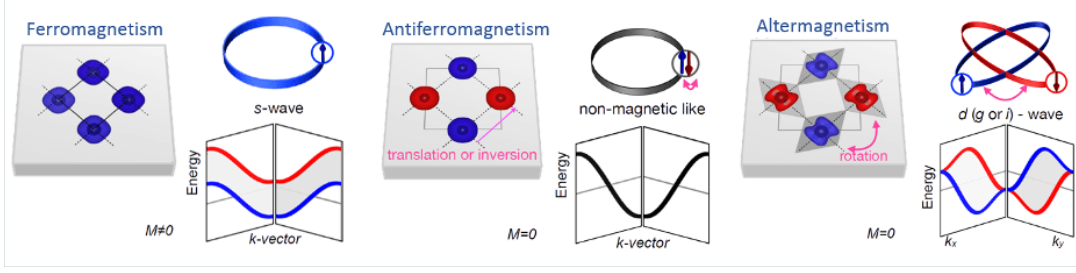


Figure 2.1 Schematic representation comparing ferromagnetism, altermagnetism, and antiferromagnetism. The diagrams illustrate differences in spin orientation (real-space symmetry), spin-dependent electronic band structures (k-space), and time-reversal symmetry properties [22].

Figure 2.1 schematically illustrates the fundamental differences between ferromagnetic, antiferromagnetic, and altermagnetic ordering, highlighting how spin arrangements and electronic band structures vary across these magnetic states. The fundamental distinction of altermagnetism arises from specific crystal symmetries that allow spin-dependent anisotropy in electron dispersion relations. Unlike ferromagnetic and antiferromagnetic materials, altermagnets possess symmetry conditions that selectively lift spin degeneracy in certain crystallographic directions while maintaining zero net magnetization in real space. This unique symmetry-breaking in momentum space leads to novel electronic properties and potential applications in spintronics, such as anisotropic spin transport and spin-polarized currents without net magnetization [23, 22].

2.3 Relevant energies in magnetic systems

The ground state energy describes a physical system in equilibrium. The total free energy (E_t) contains several contributions to a magnetic thin film sample to determine its magnetic state. The fundamental energy terms are exchange energy (E_{ex}), dipolar energy (E_d), anisotropy energy (E_a) and Zeeman energy (E_{Zee}) which couple the atomic magnetic moments.

$$E_t = E_{ex} + E_d + E_a + E_{Zee} + \dots \quad (2.6)$$

Exchange energy

Exchange interaction is a consequence of Coulomb and quantum mechanical exchange between the interacting electrons. It stems from the constraint on the symmetry of composite wave function of the electrons system as electrons are fermions. Consider two spin vectors S_i and S_j , the exchange energy is defined in Heisenberg model as,

$$E_{ex} = -2 \sum_{i \neq j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j \quad (2.7)$$

Where J_{ij} is the exchange integral, which determines how magnetic moments couple. J_{ij} determines ferromagnetic (antiferromagnetic) coupling as parallel (antiparallel) alignment of moments when it is positive (negative).

Dipolar energy

Dipolar energy is a consequence of discontinuous magnetization at the surface of the magnetic sample. Conserving magnetic flux (Gauss's law for magnetism) due to the magnetization and magnetic field at the sample surface builds up a demagnetization field in the sample. The demagnetizing field (H_d) is proportional to the M and depends on the shape of the sample. Which is also responsible for the shape anisotropy of the sample. The dipolar energy is written as,

$$E_d = -\frac{1}{2} \mu_0 \int \mathbf{H}_d \cdot \mathbf{M} dV \quad (2.8)$$

Magnetic-anisotropy energy

Magnetic anisotropic energy has contributions from shape anisotropy, a consequence of demagnetizing field, and magneto-crystalline anisotropy, a consequence of intrinsic spin-orbit coupling. The latter contribution is discussed here. The interaction of the spin magnetic moment of electrons to the orbital magnetic moment, which is strongly bound to the crystal field of the atom's environment, gives a more energetic favorable direction to the spin moment in the crystal. Hence, the easy axis is a preferential magnetization direction in the crystalline sample. The general form of uniaxial anisotropy is given by

$$E_a = V K_1^{\text{uni}} \sin^2 \theta + V K_2^{\text{uni}} \sin^4 \theta + \dots \quad (2.9)$$

with K_i^{uni} is the anisotropy constants and here θ is the angle between the magnetization and the easy axis.

Zeeman Interaction

Zeeman energy is the potential energy of the magnetic moment in the presence of an external magnetic field (H_{ext}). The magnetic moment tends to align with the field to minimize the energy. This energy term is written as

$$E_{Zee} = -\mu_0 \int \mathbf{H}_{\text{ext}} \cdot \mathbf{M} dV \quad (2.10)$$

2.4 Spin Currents and Hall Phenomena

Spin current refers to the flow of electron spin angular momentum, distinct from the conventional charge current, which involves the net movement of electric charge. While charge current arises from electrons moving collectively in response to electric fields, spin currents occur due to the directed flow of spin-polarized electrons. Crucially, spin currents can exist even in the absence of net charge transport, as electrons of opposite spins moving in opposite directions produce no overall charge flow or via magnons in magnetic insulators, but still carry spin information. Generation of spin currents can occur through various physical mechanisms, including spin-orbit coupling [24–26], spin pumping from magnetic precession [26], thermal gradients via the spin Seebeck effect (SSE) [27], and through interfacial interactions in multilayered structures [9].

The interaction of spin and charge degrees of freedom, mediated primarily through spin-orbit coupling, leads naturally to several Hall effects. Spin currents, particularly when influenced by spin-orbit interactions, strongly connect to Hall phenomena—namely, the ordinary Hall effect (OHE), anomalous Hall effect (AHE), and spin Hall effect (SHE). These phenomena share the common theme of transverse responses arising due to the interplay between magnetic fields, magnetization, and spin-orbit coupling.

The OHE is one of the earliest discovered manifestations of magnetotransport phenomena. It occurs when charge carriers, moving under the influence of an electric field, are deflected by the Lorentz force due to an externally applied magnetic field. This deflection leads to an accumulation of charges on one side of the conductor, thereby creating a measurable transverse voltage known as the Hall voltage. Mathematically,

the Hall voltage V_H can be expressed as:

$$V_H = \frac{IB}{ned}, \quad (2.11)$$

where I represents the current flowing through the conductor, B is the applied magnetic field perpendicular to the current direction, n denotes the carrier density, e the electronic charge, and d the thickness of the sample.

In contrast to the ordinary Hall effect, the AHE does not require an external magnetic field to arise but instead emerges intrinsically from spin-orbit coupling within magnetized materials. The AHE results from electrons experiencing asymmetric scattering, driven primarily by the spin-dependent interactions induced by the material's magnetization. As such, AHE is strongly linked to the magnetic order of the material. The anomalous Hall resistivity ρ_{xy}^{AHE} typically depends directly on the perpendicular component of the magnetization M_z and can be described by:

$$\rho_{xy}^{\text{AHE}} = R_s M_z, \quad (2.12)$$

where R_s is a material-dependent parameter.

Closely related to the AHE is the SHE, observed in non-magnetic materials where spin-orbit coupling induces a transverse spin current from an applied charge current, even in the absence of net magnetization. The spin Hall effect can be viewed as an extension of the AHE, occurring without the requirement of magnetic order. In SHE, electrons with opposite spin orientations are deflected in opposite directions, generating a spin accumulation at lateral boundaries, and can be mathematically represented as:

$$J_s = \theta_{\text{SH}} \frac{h}{4e\pi} J_c \quad (2.13)$$

where J_s denotes the generated spin current, J_c is the applied charge current density, θ_{SH} is the spin Hall angle characterizing the efficiency of the conversion from charge to spin current, and h and e represent Planck's constant and the electron charge, respectively.

The SHE and the AHE share a common physical origin in spin-orbit coupling, which governs spin-dependent scattering of charge carriers. The SHE can be understood as the non-magnetic counterpart of the AHE—both produce transverse responses, but whereas AHE requires intrinsic magnetization, SHE arises purely from spin-orbit interactions in non-magnetic systems. In this context, interface-generated spin currents emerge as a particularly important class, where spin-orbit coupling at material

interfaces induces spin accumulation and transport. These interfacial spin currents are crucial for understanding spin-charge conversion and spin transport in modern heterostructures..

2.5 Magnetoresistance

Magnetoresistance (MR) refers to the change in a material's electrical resistivity in response to an applied magnetic field. The magnitude and nature of this change depend on the material's intrinsic properties and the relative orientation of magnetization, current, and magnetic field. Key types of magnetoresistance include ordinary magnetoresistance (OMR), anisotropic magnetoresistance (AMR), and spin Hall magnetoresistance (SMR).

OMR results from the deflection of charge carriers due to the Lorentz force when a magnetic field is applied perpendicular to the direction of current flow. This curvature of the carrier trajectory increases the effective path length, leading to higher resistivity. In a simplified form, this classical behavior is expressed as:

$$\rho_{\text{long}} \propto \Delta\rho B^2. \quad (2.14)$$

AMR, observed in ferromagnetic materials, originates from the spin-dependent scattering of conduction electrons [28]. The resistivity depends on the angle between the current and the magnetization direction. When the current is aligned parallel to the magnetization, the resistivity differs from when it is perpendicular. This angular dependence is given by:

$$\rho_{\text{long}}(\theta) = \rho_{\parallel} + (\rho_{\perp} - \rho_{\parallel}) \sin^2 \theta, \quad (2.15)$$

where $\rho_{\parallel}$ and $\rho_{\perp}$ denote the resistivity with magnetization parallel and perpendicular to the current, respectively.

SMR arises in bilayer systems composed of a ferromagnetic insulator or metal and a non-magnetic heavy metal with significant spin-orbit coupling, such as Pt, Ta, or W. In these systems, the spin Hall effect in the heavy metal layer generates a transverse spin current from an applied charge current. This spin current accumulates at the interface with the magnetic layer, where it is either absorbed or reflected depending on the orientation of the magnetization. This interplay modifies the longitudinal and transverse resistivity of the heavy metal layer [29].

The longitudinal resistivity in SMR can be expressed as:

$$\rho_{\text{long}} \approx \rho + \Delta\rho_0 + \Delta\rho_1(1 - m_y^2), \quad (2.16)$$

while the transverse resistivity, associated with the spin Hall effect and its modulation by the magnetization, is given by:

$$\rho_{\text{trans}} \approx \Delta\rho_1 m_x m_y + \Delta\rho_2 m_z, \quad (2.17)$$

where $\Delta\rho_0$, $\Delta\rho_1$, and $\Delta\rho_2$ are material-dependent coefficients, m_x , m_y , and m_z are the components of the unit magnetization vector.

These magnetoresistance effects are not isolated phenomena but are intrinsically connected through spin-orbit coupling and spin transport mechanisms. The spin Hall effect, for instance, generates spin currents that directly contribute to SMR, while both the AHEs and SHEs arise from the same underlying spin-orbit interactions. Together, these effects underscore the centrality of spin-orbit coupling in mediating and linking transverse transport and magnetoresistive responses in spintronic systems.

2.6 Magnetic-material of interest

2.6.1 Ruthenium dioxide

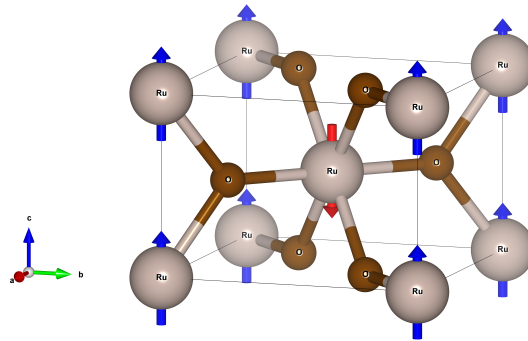


Figure 2.2 RuO₂ unit cell with the magnetic moment of each Ru atom [30].

Ruthenium dioxide (RuO₂) is a binary transition metal oxide that crystallizes in the tetragonal rutile structure. In this structure, each ruthenium (Ru) atom is located at the center of a distorted octahedron coordinated by six oxygen (O) atoms [31, 32]. The unit cell contains two formula units with Ru atoms at distinct sublattices,

often referred to as Ru_A and Ru_B , related by a C4 rotation combined with a translation [22, 31, 33]. Historically, RuO_2 has been considered a classic example of paramagnetic metal, exhibiting high electrical conductivity comparable to that of typical metals [6]. Its robustness, chemical stability, and excellent electrical conductivity have led to its widespread use in various technological applications, including heterogeneous catalysis, electrocatalysis, electrode materials in electrolytic cells, supercapacitors, batteries, fuel cells, and as diffusion barriers in microelectronic devices [34, 33]. Its metallic nature and band structure have been subjects of theoretical and experimental investigation [22, 33].

Despite the long-standing view of RuO_2 as a paramagnet, recent experimental findings have indicated the presence of itinerant antiferromagnetism with a Néel temperature (T_N) exceeding 300 K [6, 33, 35–38]. This discovery, primarily supported by neutron diffraction and resonant x-ray scattering experiments, reported small Ru magnetic moments of approximately 0.05 μ_B at room temperature. The Néel vector was observed to align along the c-axis [6, 33]. Theoretical studies employing Density Functional Theory (DFT) with various approaches such as DFT+U and Dynamical Mean-Field Theory suggested that this antiferromagnetism arises from a Fermi surface instability, possibly manifesting as a spin density wave state [6, 37, 39, 40]. This peculiar magnetic order, combined with the crystal symmetry where opposite-spin sublattices are related by rotational symmetry rather than simple translation or inversion, led to the classification of RuO_2 as a potential altermagnet. Altermagnetism predicts a non-relativistic, momentum-dependent spin splitting in the electronic band structure, often described as d-wave like [22, 38, 39].

The proposed altermagnetic state and associated spin splitting in RuO_2 have generated significant interest in its potential spintronic applications [33, 36, 39, 41, 42]. A key property is the efficient charge-to-spin conversion via the anisotropic spin splitting effect, predicted to generate spin currents with polarization primarily aligned along the Néel vector [36, 37, 41, 43]. This is distinct from the conventional SHE, which relies on relativistic spin-orbit coupling [44]. Experimental studies, often utilizing RuO_2 /ferromagnet heterostructures (such as RuO_2 /Permalloy), have reported the observation of spin-orbit torques (SOTs) attributed to the altermagnetic spin-splitting effect (ASSE), including spin-splitter torque (SST) [35, 36, 45–47]. These torques enable deterministic, fully field-free magnetization switching in adjacent magnetic layers [41]. Furthermore, an AHE exceeding $1000 \Omega^{-1} \text{cm}^{-1}$ has been reported in epitaxial RuO_2 films, a phenomenon unexpected in conventional compensated antiferromagnets but consistent with the symmetry of altermagnets [8, 47]. Inverse effects,

such as spin-to-charge conversion, have also been investigated using techniques like spin pumping, revealing anisotropic and temperature-dependent behavior [36, 48]. The characteristic spin splitting and time-reversal symmetry breaking in momentum space associated with altermagnetism are believed to underpin these emergent phenomena. Time-domain terahertz spectroscopy has also been employed to study charge dynamics in RuO₂ [42].

Despite the compelling evidence from early studies and transport measurements, the magnetic ground state and altermagnetic character of RuO₂ have recently become a subject of significant controversy [7, 49–53]. Several recent experimental investigations utilizing highly sensitive techniques such as muon spin rotation (μ SR), refined neutron diffraction, polarized neutron diffraction, angle-resolved photoemission spectroscopy (ARPES), and resonant elastic x-ray scattering report the absence of detectable long-range magnetic order in both bulk single crystals and epitaxial thin films of RuO₂ [7, 49–54]. These studies place strict upper limits on the magnitude of potential Ru magnetic moments, estimating them to be orders of magnitude smaller (e.g., $\approx 10^{-4}$ μ B/Ru) than the previously reported values of 0.05 μ B [49, 50, 52, 54]. Furthermore, ARPES and optical conductivity measurements suggest that the electronic structure of RuO₂ is consistent with nonmagnetic calculations, lacking the predicted momentum-dependent spin splitting characteristic of altermagnetism [7, 53]. Critiques of earlier diffraction results suggest that phenomena like multiple scattering in single crystals may have been misinterpreted as magnetic signals [49, 54]. Alternative explanations for the observed transport effects that were initially attributed to altermagnetism are being explored, and theoretical studies suggest that factors such as Ru vacancies or strain could potentially induce magnetism, offering a path to reconcile the conflicting experimental results [13, 40, 52, 55].

The current state of research on RuO₂ is characterized by a fundamental debate regarding its magnetic classification. Key open questions revolve around definitively establishing the presence or absence of magnetic order in stoichiometric, high-quality samples and understanding the microscopic origin of previously reported spin-related phenomena, such as the AHE and SOTs, if bulk magnetism is indeed absent or significantly weaker than initially thought [49–51]. Future research directions are critical to resolving this controversy and fully understanding the properties of this intriguing material. This includes meticulous sample preparation and characterization, particularly regarding stoichiometry and strain, as suggested by theoretical work [40]. Advanced experimental techniques with high sensitivity to local magnetic moments and spin textures are necessary to provide conclusive evidence [49, 50, 52].

Theoretical efforts must continue to refine models of electronic correlations and investigate the effects of imperfections and external tuning parameters like strain and doping [6, 39, 40, 52]. Ultimately, resolving the debate surrounding RuO_2 's magnetic nature is crucial for its potential integration into future energy-efficient spin-based memory and logic devices, and for advancing the fundamental understanding of unconventional magnetic phases and their associated phenomena [22, 41].

2.6.2 Yttrium iron garnet

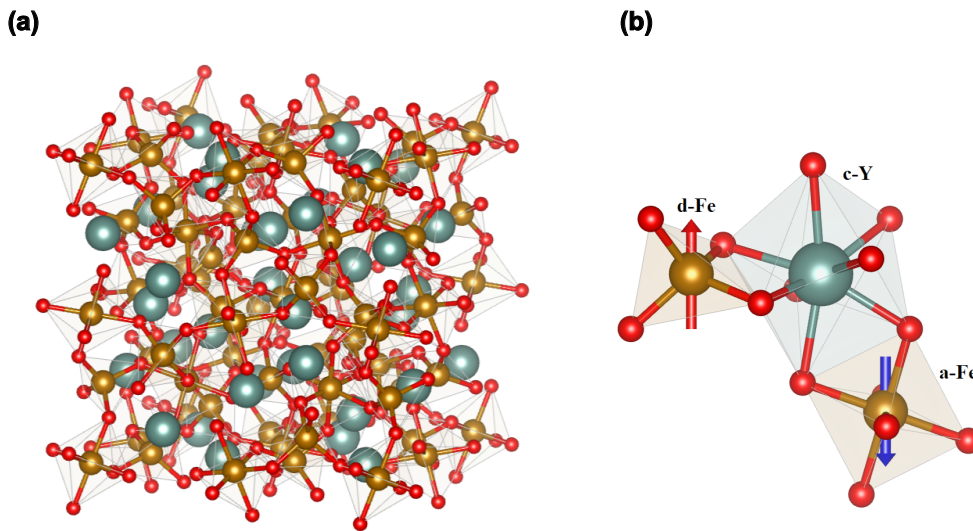


Figure 2.3 (a) Unit cell of Yttrium Iron Garnet (YIG), showing oxygen atoms in red, iron atoms in golden brown, and yttrium ions in turquoise. (b) Reduced crystal structure highlighting the relative alignment and magnetic moments of the iron ions at octahedral (d-Fe) and tetrahedral (a-Fe) coordination sites, as well as the yttrium ions at the dodecahedral (c-Y) sites.

Yttrium Iron Garnet ($\text{Y}_3\text{Fe}_5\text{O}_{12}$, YIG) is a ferrimagnetic insulator [56] with a complex cubic crystal structure. Its structure includes magnetic Fe^{3+} ions on octahedral and tetrahedral sites, with the net magnetic moment arising from the antiferromagnetic superexchange interaction between these sites via oxygen ions [57, 58]. Since its synthesis by Geller and Gilileo in 1957 [57], YIG has served as a classical object for experimental studies in magnetism and is essential for investigating new phenomena [59, 60]. YIG is notable for its exceptional physical characteristics, possessing the lowest known magnetic damping [61, 58]. This ultra-low damping facilitates long

magnon propagation distances (up to centimeters) at room temperature. Furthermore, its high Curie temperature of approximately 560 K allows for room temperature experiments. These properties make YIG a prototype material for fundamental studies of various magnonic phenomena [59].

YIG's significance extends to spintronics, particularly as a magnetic insulator which allows clear separation between spin and charge currents [58, 61, 62]. This has led to extensive research on spin-related phenomena at interfaces between YIG and normal metals like Platinum. Key effects include spin pumping, the SSE, and SMR [62–64]. YIG plays a central role in magnonics, a field focused on using spin waves (magnons) as information carriers [60, 59]. YIG's extremely low damping and long magnon propagation length are critical for the efficient transport and processing of information using these waves. Concepts learned from early experiments on bulk YIG are now being transferred to smaller structures for sub-micrometer devices, and YIG is utilized in magnonic crystals [60].

Spin information transport in magnetic insulators such as YIG occurs through the diffusion of non-equilibrium magnons, rather than conduction electrons. Experimental studies have successfully demonstrated this spin transport over significant distances [64, 65]. This diffusive transport is characterized by the magnon spin diffusion length, which defines the length scale over which the magnon spin current decays. Experiments have measured magnon spin diffusion lengths of several micrometers in thin YIG films at room temperature. Nonlocal transport is a key experimental geometry for studying this spin transport [64, 66]. In YIG/normal metal heterostructures, an electrical current in the injector metal strip generates a transverse spin current via the SHE, which excites non-equilibrium magnons in the YIG. These magnons diffuse through the YIG to a detector strip, where the reciprocal inverse spin Hall effect (iSHE) converts the magnon interaction into a detectable charge voltage. This nonlocal measurement directly probes and demonstrates efficient spin transport in YIG via magnons without charge currents [62, 64].

Chapter 3

Methods

This work focuses on the study of thin films, including their characterization and measurements. This chapter discusses the methods used for fabricating the films, characterizing them, and conducting electrical transport measurements. The thin film samples are grown using pulsed laser deposition (PLD) and DC sputtering. They are evaluated *in situ* using reflection high-energy electron diffraction (RHEED) and *ex situ* using X-ray diffraction (XRD). Electrical transport measurements are performed in various cryostats.

3.1 Pulsed laser deposition

Pulsed laser deposition is one among several physical vapor deposition (PVD) techniques for the deposition of a thin film. In PVD, material converts from a condensed phase to a vapor phase and back to a thin film condensed phase. In PLD, a pulsed laser beam is an agent to transition from the condensed phase to the vapor phase. PLD is a popular method for growing epitaxial complex oxides as it is relatively easy to maintain the stoichiometry while transferring material to a thin film.

In PLD, a high-intensity pulsed laser is applied to the surface of the target material to deposit the desired thin film. The photon energy from laser pulses is transferred to the target's surface, breaking atomic bonds. As a result, the atoms, some of which are ionized, move perpendicularly from the target's surface to the substrate's surface. The growth is usually done with high substrate temperature and partial background pressure of oxygen. The substrate temperature provides incoming atomic species with kinetic energy to mobilize, which assists epitaxial growth. The background oxygen pressure helps to maintain oxide composition and limits the high-energy ion flux on the substrate.

The vacuum chamber for PLD use two-stage pumping systems to reach a base pressure of $\sim 10^{-8}$ mbar and a loadlock chamber with a similar base pressure as the main chamber to minimize contamination. Mass flow controllers and gate valve to the pumping system control the gas pressure inside the chamber. Five targets can be installed at a time and rotated to a laser spot. Most of the significant features of the chamber are shown in figure 3.1. A second continuous wave infrared laser pointed at the underside of the substrate holder heats the substrate. A feedback loop regulates the laser output to maintain the substrate temperature after a calibrated pyrometer measures the temperature of the substrate holder. The UV light source is a KrF excimer laser with 248 nm wavelength. The Coherent COMPex Pro 205 laser produces 20 ns

long laser pulses with a frequency range of 1 to 50 Hz. When emitted by the laser, one pulse typically has an energy of roughly 200 mJ. This emitted laser pulse is guided with an optical system of lens and mirrors to contain the maximum energy of the pulse and to hit the target at an angle of 45 degrees. The energy density of 860 mJ/cm^2 of energy is present at the target, evenly dispersed throughout the laser spot's area. The target is rotated and moved back and forth during the deposition to prevent shooting a hole in the material.

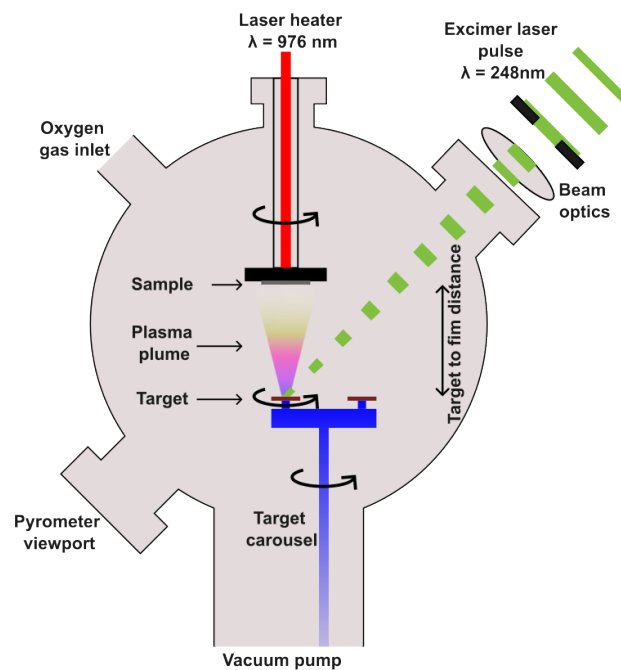


Figure 3.1 Sketch of the PLD chamber and the deposition process in the PLD system.

A disadvantage of PLD is its limitation on film thickness, which is only up to a few hundred nanometers. Low deposition rates in PLD and long deposition time causes significant damage to ablating target, and deposition conditions start to drift as the gas of the excimer laser fatigues over time.

3.2 DC Magnetron Sputtering

Among PVD techniques, DC magnetron sputtering stands out as a versatile technique, especially suitable for simple compound thin films. In contrast to PLD, where a high-energy photon pulse ablates material, sputtering ejects atoms by direct momentum transfer: inert-gas ions (*e.g.* Ar^+) are accelerated towards a negatively biased target, ejecting atoms that travel the gas phase and condense on the substrate. The stoichiometry of multi-element targets can deviate because different species of atoms are sputtered differently depending on their mass, yet a pre-sputtering process can reduce this effect. Furthermore, PLD can sometimes eject undesirable micro-sized globules or particles from the target, which is not a typical issue with sputtering.

A defining feature of magnetron sputtering is its magnetron arrangement. Permanent magnets behind the target generate a tangential magnetic field that forces secondary electrons into cycloidal paths, increasing their residence time. The resulting dense plasma sustains high ionisation at lower pressures, affords deposition rates higher than normal sputtering [67–69].

Films of Pt, Permalloy ($\text{Ni}_{80}\text{Fe}_{20}$), Au, and Al were deposited by magnetron sputtering for spin-Hall studies, electrical contacts, and capping layers. Unless stated otherwise, the standard recipe employs an Ar pressure of 0.02 mbar, a plasma current of 25 mA, and depositions are made at room temperature.

3.3 X-ray characterization

X-rays are electromagnetic waves with a wavelength of 0.1 to 100 Angstrom. The choice of x-ray of wavelength smaller than typical inter planer spacing of the atomic lattice is a suitable probing wave. X-rays can be used to characterize the thin film's crystalline characteristics and thickness irrespective of its crystalline order. Various x-ray scans are discussed in the coming sub-section, providing information about the thin film samples.

The Bruker D8 Discover 4-circle diffractometer with a Cu anode is the equipment used to conduct the x-ray measurements for this study. Measurements are made in the Bragg-Brentano geometry in this system. After leaving the Cu anode in line-focus, the x-rays are reflected by a Göbel mirror, collimating and partially monochromatizing the x-ray beam. The beam is then contained by being guided through a slit. A 2-bounce Ge channel-cut monochromator is utilized to decrease contributions to the x-ray spectrum other than the $\text{Cu K}_{\alpha 1}$ emission. The beam then strikes the sample, which can be

rotated across three perpendicular axes to reach every lattice plane. After passing through a set of motorized slits, the reflected beam is subsequently absorbed by the proportional counter acting as point detector.

3.3.1 X-ray reflectivity

In x-ray reflectivity (XRR), the interference between the externally reflected part of grazing incident x-rays gives information about the thin film. X-rays are reflected from the interfaces due to the difference in electron density across the interface. The critical angle of external reflection depends on the electron density of the top layer of our thin film stack and increases with the density of the top layer. Above this critical angle the partially transmitted part of incoming beam will be reflected from the following interface in the stack. This reflected part from the top surface and following interface will do constructive or destructive interference as a function of the grazing incident x-rays. For a given wavelength of x-ray, oscillation of the intensity in $2\theta/\omega$ scan, so-called Kiessig fringes, is linked to the thickness of the thin film. Here ω is the angle between the surface of the substrate to the incident x-ray beam, and 2θ is the angle between the incident and reflected x-ray beam. Other quantitative parameters of the thin-film stack, like surface roughness, interface roughness, and density of thin film can be deduced by analyzing the $2\theta/\omega$ scan. The advantage of XRR in finding the thickness and roughness of thin film is that it works independently of the crystalline quality of the thin film.

3.3.2 X-ray diffraction

The scattering of the x-rays from the atomic lattice planes leads to constructive or destructive interference as a function of the angle between the incident and scattered beam. In reciprocal space, the condition for x-ray diffraction is a consequence of momentum conservation. This condition is given in the form of Laue equation: the difference ($\Delta\mathbf{k}$) between the incident wave vector ($\mathbf{k}_{\text{in}}$) of the x-ray and scattered wave vector ($\mathbf{k}_{\text{out}}$) is equal to the reciprocal lattice vector ($\mathbf{G}$) in the plane of $\mathbf{k}_{\text{in}}$ and $\mathbf{k}_{\text{out}}$.

$$\Delta\mathbf{k} = \mathbf{k}_{\text{out}} - \mathbf{k}_{\text{in}} = \mathbf{G} \quad (3.1)$$

The diffraction condition in real space is Bragg's law, which can be deduced from the Laue equation. Bragg's law relates the path difference between two waves reflected

from the lattice planes to the condition of constructive interference between two waves.

$$n\lambda = 2d \sin \theta \quad (3.2)$$

Where n , λ , d , θ is the diffraction order, wavelength of incident x-ray, lattice plane spacing, and incident angle of x-ray with respect to the lattice plane.

We can do various scans with x-rays on the sample to extract its different crystalline characteristics. The most relevant scans used in the study are discussed here and related axes are shown in figure 3.2:

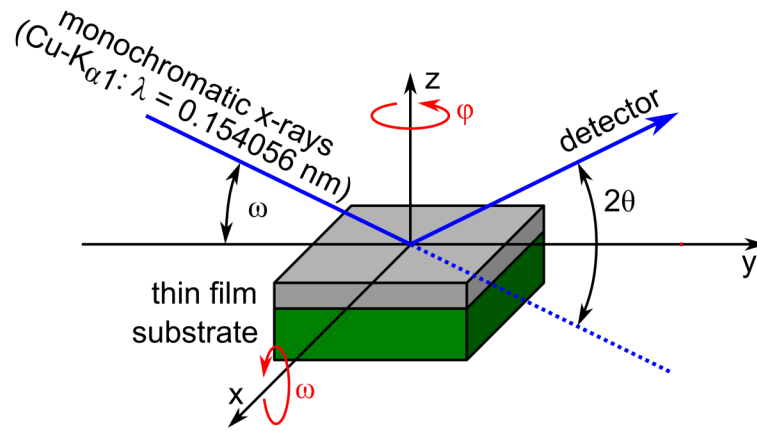


Figure 3.2 Monochromatic x-rays hit the substrate's surface at an angle ω with respect to the xy plane and diffracted intensity is collected with a point detector at an angle 2θ with respect to the incident beam. Showing ω axis and Φ axis [70].

$2\theta/\theta$ scans

In this scan, the incident angle (θ) of x-rays on the sample is increased and simultaneously diffracted intensity at the 2θ angle is recorded. This scan is done while keeping the reciprocal lattice vector in the plane of the incident and diffracted x-ray. The $2\theta/\theta$ scan is equivalent to increasing the magnitude of the scattering vector in the direction of the fixed reciprocal space vector. Hence, the scan matches the diffraction condition whenever the magnitude of the scattering wave vector is equal to the reciprocal space vector.

The presence of several diffraction reflexes from several different planes signifies a polycrystalline sample. The broadening of the diffraction peak on the 2θ could be due to the instrument, imperfect monochromatic beam of the x-ray source, relaxation

in the thin film strain, lattice imperfection, dislocation of the lattices, etc. Another reason is the finite size effect of our thin film sample in the direction of the reciprocal space vector. The Scherrer formula can estimate this finite size by broadening of reflex at 2θ . This estimate of size will establish an upper limit on the grain size of our samples. Another consequence of finite size is the presence of secondary oscillating peaks around primary diffracted reflection in case of low mosaicity of the thin film samples. These secondary oscillating peaks, also called Laue oscillations and period of oscillation, is related to the scattering volume of this crystallite in the direction of the scattering vector. These finite-size effects of epitaxial thin films can be simulated and compared to experiments.

Rocking or ω scans

In the rocking scan, the sample is rotated around ω while keeping 2θ fixed for a particular diffraction reflex. The broadening of the rocking curve is a measure of the order of mosaicity of different crystallites in the thin film. The full width quantifies the broadening of the peak at half maximum of the rocking curve.

Φ scans

In Φ scans, the sample is rotated in the plane of the sample after tilting the inclined lattice plane with respect to the sample surface such that the corresponding reciprocal lattice vector is in the plane of $\mathbf{k}_{in}$ and $\mathbf{k}_{out}$. Φ scans show the rotation symmetry of reciprocal space vectors of selected inclined lattice planes. Comparing this periodicity to the symmetry of crystal helps to detect the presence of differently ordered domains. Phi-scans became helpful in confirming in-plane orientation and establishing the epitaxial relationship between the substrate and thin film in the case of an epitaxial thin film.

3.4 Reflection high energy electron diffraction

Reflection high energy electron diffraction (RHEED) is forward scattered diffraction of grazing incident electrons from the sample surface. The working principle of RHEED and XRD is similar, given by the Laue equation in the reciprocal space. The reciprocal space here under consideration is the sample's surface, as electrons can not penetrate inside the sample except for a few lattice planes. In other words, the overlap of the

Ewald sphere with the surface's reciprocal lattice is the diffraction spots' position. In the case of a 2D surface, these reciprocal lattice points become continuous in the direction perpendicular to the surface, i.e., rods. Hence, the diffraction spots are the cross-section of these rods with Ewald's sphere (see fig. 3.3). The advantage of using a high-energy collimated electron beam is that the angular spread of diffraction spots is limited due to smaller wavelength than in x-ray diffraction. This diffraction pattern can be imaged by a few centimeter large fluorescent screen. In contrast to photons, electrons have far greater interactions with atoms due to their charge. Because of this, the Born approximation is inappropriate for electron scattering, and it is difficult to calculate diffraction intensities. However, momentum conservation in reciprocal space holds for both electron and photon scattering and therefore peak positions are given by Bragg's law.

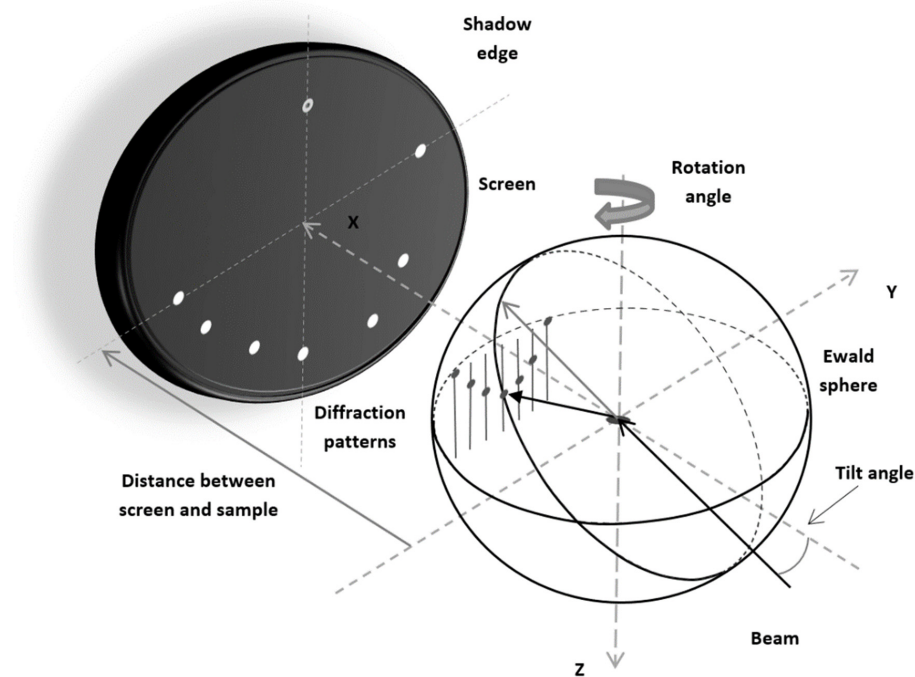


Figure 3.3 The Ewald construction for RHEED [71].

A disadvantage of using RHEED in PLD is that electrons that scatter with the different particles in the thin film deposition environment lose the information in contrast to molecular beam epitaxy. The decreased mean free path of electrons during the deposition in the PLD due to the high-pressure environment leads to this

information loss. The typical high pressure to deposit oxides in the PLD is in order of 10^{-2} mbar. Therefore, the RHEED system is equipped with a double differential pumping section and a RHEED screen protruding inside the vacuum chamber to minimize the electron interaction with the background environment and to retain maximum diffraction information from the sample's surface. The typical RHEED apparatus is shown without double differential pumping section and a RHEED screen protruding inside the vacuum chamber in figure 3.4.

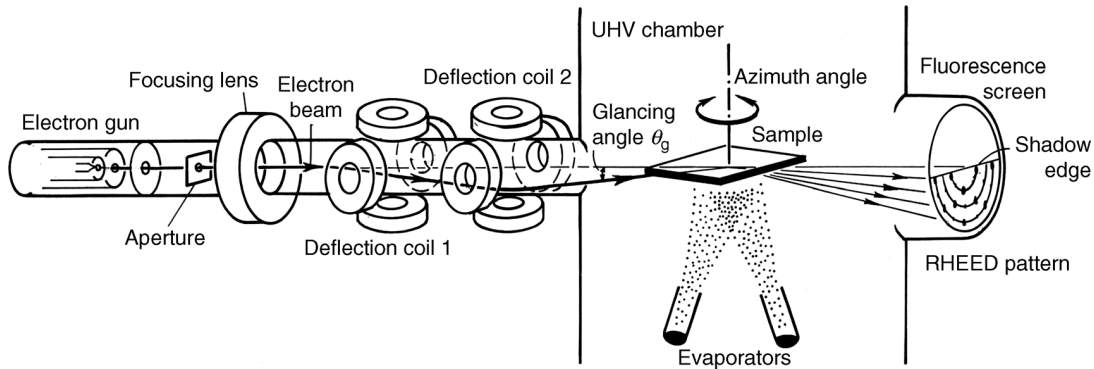


Figure 3.4 Schematic diagram of RHEED apparatus [72].

The accelerating voltage (V_a) for electrons can go up to 40 kV (Kilovolt) in the RHEED system, and all the diffraction images in this work are captured at 30 kV. The associated de Broglie wavelength of electrons is given by the ratio of Planck's constant to the electron's momentum (p). It can be related to the accelerating voltage as in equation 3.3. For $V_a = 30$ kV, the de Broglie wavelength of electrons can be calculated to 7.06×10^{-12} m.

$$\lambda_{\text{deBroglie}} = \frac{h}{p_e} = \frac{h}{m_e \cdot v_e} = \frac{h}{m_e \cdot \sqrt{2 \cdot \frac{e}{m_e} \cdot V_a}} = \frac{h}{\sqrt{2 \cdot m_e \cdot e \cdot V_a}} \quad (3.3)$$

The actual sample is not always with a single crystalline and atomically flat surface. The reciprocal lattice rods undergo changes that alter their cross-section with Ewald's sphere depending on the surface quality. Figure 3.5 shows the RHEED pattern from some possible actual sample surfaces. The study of diffraction patterns can provide information about the flatness of the surface, and monitoring the evolution of the pattern while growing can provide information about growth style, epitaxial relation between the substrate and thin film, and the number of atomic layers grown. The intensity of spots or streaks in the RHEED pattern changes their intensity as a function of the surface roughness. Hence, this intensity oscillates in the formation of each

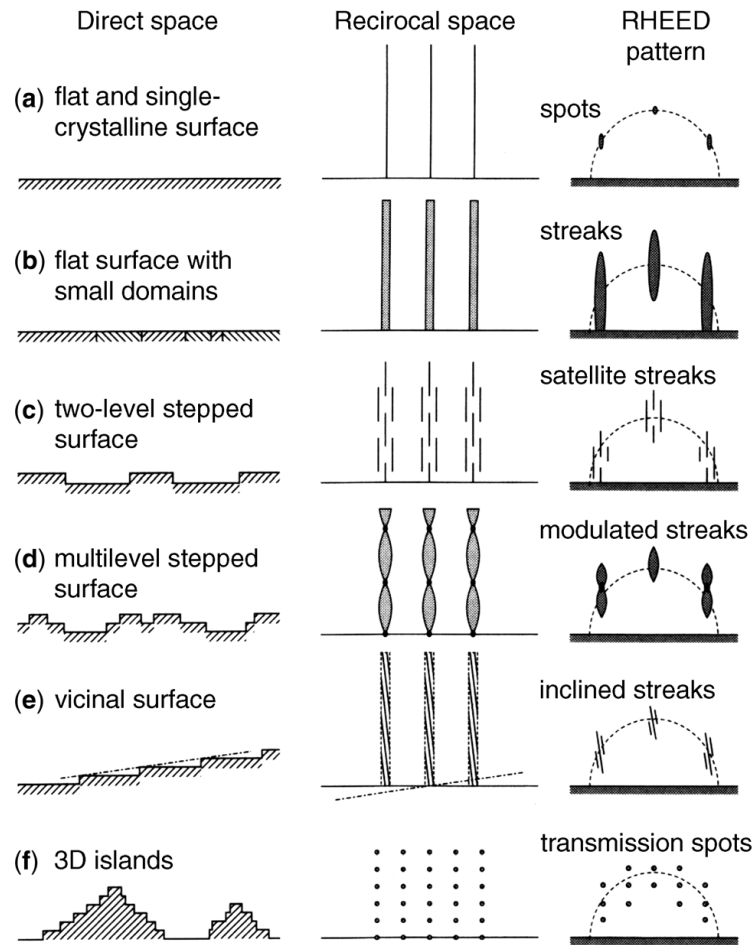


Figure 3.5 Schematics of various kinds of realistic surfaces, in real-space morphology, in reciprocal space, and their RHEED patterns [72].

thin film layer in case of thin film's layer-by-layer growth. So, RHEED can work complementary to the XRD to analyze the structural properties of our sample and its surface.

3.5 Lithography

Lithography is a fundamental process for transferring a design onto a thin layer of radiation-sensitive material, known as a resist, serving as a crucial step in micro- and nanofabrication [73, 74]. This technique enables the creation of complex patterns down to the nanometer scale, driving the development of integrated microelectronic circuitry. The general process involves depositing a resist layer on a substrate, selectively exposing it to a patterned energy source (such as light or electrons), and then developing

the resist to remove either the exposed or unexposed areas. The resulting resist pattern then acts as a mask for subsequent processing steps, such as etching or deposition, to transfer the pattern into the underlying material. The continuous push for smaller, faster, and cheaper components necessitates lithographic techniques capable of ever-higher resolution and precision.

Optical lithography, or photolithography, is the workhorse of the microelectronics industry due to its high throughput and efficiency. This technique employs light, typically ultraviolet (UV) light, as the exposure source. In conventional optical lithography, a physical mask, usually a glass plate with a chromium pattern, is used to define the areas of the resist exposed to light. The exposure causes a chemical reaction in the resist, altering its solubility in a developer solution. The resolution limit in optical lithography is primarily determined by the wavelength of the light used, as described by Rayleigh's equation. To overcome this limitation and achieve smaller feature sizes, shorter UV wavelengths, and resolution enhancement techniques, like phase-shifting masks, are employed [74, 75]. While traditional optical lithography relies on masks, direct-write optical lithography offers an alternative, particularly useful in research and development where mask designs change frequently. This approach projects the pattern directly onto the resist using computer-controlled optics, eliminating the need for a physical mask. In our context, a MicroWriter ML3 tool was utilized for optical lithography [76].

3.6 Electrical Transport Measurements

For Electrical transport measurements, samples were fabricated into suitable geometries for electrical transport characterization, including Hall bar configurations. These structures were defined using optical lithography, followed by the deposition or etching of conductive layers using magnetron sputtering. To facilitate electrical connections, Au contacts were deposited by sputtering, utilizing a thin adhesion layer of chromium (Cr) due to the poor adhesion characteristics of Au on substrate surfaces. The metallic layers were patterned using a lift-off technique, ensuring clean and well-defined structures.

Following fabrication, samples were mounted onto printed circuit boards (PCBs) tailored specifically for various measurement orientations, including both in-plane and out-of-plane configurations. The PCBs were integrated into setups capable of precise angular control. A piezoelectric rotator (Attocube ANRv51RES) provided

fine angular positioning, allowing the sample orientation relative to an externally applied magnetic field to be varied continuously within an angular range of up to 300° . These measurements were performed inside a helium-cooled cryostat system capable of maintaining stable temperatures between 4 K and 300 K. Superconducting magnets with field strengths of up to 12 T or 15 T were utilized to explore the magnetic field dependence of transport phenomena. Additionally, a vector magnet capable of generating magnetic fields up to 1 T in arbitrary directions and up to 5 T along a single axis facilitated angular-dependent measurements without necessitating re-mounting. The experimental setup allowed for characterization of longitudinal and transverse resistivities (ρ_L and ρ_T , respectively) across varying temperatures, magnetic fields, and sample orientations.

Instrumentation for electrical transport measurements primarily involved a Keithley 6221 current source paired with a Keithley 2182 nanovoltmeter, configured in Delta Mode operation. In this mode, the applied current is reversed at 50 Hz frequency, generating a square-wave current pattern. Correspondingly, voltages were measured alternately for positive and negative current directions, significantly reducing noise levels compared to conventional DC measurements.

Local MR measurements employed Hall bar structures, where a central conductive bar transmitted a direct current, and voltages were probed at contact pads distributed longitudinally and transversely. The configurations allowed for detailed examination of intrinsic resistive properties and Hall effects. To minimize temperature fluctuations, the samples were first allowed to reach thermal equilibrium and a stable temperature before the measurements began. This ensured the reliable detection of even small spin-related signals, especially in temperature-sensitive materials such as platinum (Pt). The polarity and amplitude of the measured voltages in these configurations depended critically on the spin Hall angles of the electrodes and their relative signs, offering detailed insights into spin transport phenomena.

3.7 Muon Spin Rotation/Relaxation (μ SR)

Muon Spin Spectroscopy is an advanced experimental technique used to probe the local magnetic and electronic properties of condensed matter systems. The acronym μ SR stands for Muon Spin Rotation, Relaxation, or Resonance. A key advantage of μ SR is that it utilizes an extrinsic probe (μ^+) and does not rely on specific host nuclei, making it universally applicable. The μ SR technique employs the positive muon (μ^+),

which is a charged lepton, a subatomic particle with a charge of $+e$, a spin of $s = 1/2$, and a large magnetic moment. The muon's magnetic moment is approximately 3.18 times that of a proton, enabling it to act as an exquisitely sensitive magnetic probe. The average lifetime of the muon is $\tau_\mu \approx 2.2 \mu\text{s}$ [77, 78]. High-flux beams of muons are produced artificially at accelerator facilities, such as the Paul Scherrer Institute (PSI) [77]. Positive pions (π^+), created by colliding high-energy protons with a low atomic number target (e.g., beryllium or carbon), decay almost instantly ($\tau_\pi \approx 26 \text{ ns}$) into a positive muon and a muon neutrino. The resultant positive muons are typically extracted as highly spin-polarized surface muons ($\sim 100\%$ polarization), where the muon spin is aligned antiparallel to its momentum [77, 78].

The physical basis of μ SR is the anisotropic emission of positrons (e^+) resulting from the muon decay, governed by the weak interaction. The decay process is given by:

$$\mu^+ \longrightarrow e^+ + \nu_e + \bar{\nu}_\mu$$

The positron is preferentially emitted along the instantaneous direction of the muon spin. Figure 3.6 shows the experimental setup for the μ SR measurements. By detecting the time and position of the decay positrons using detectors symmetrically arranged around the sample, the time evolution of the ensemble-averaged muon spin polarization $P(t)$ can be determined. The signal measured, the time-dependent asymmetry $A(t)$, is proportional to the muon polarization $P(t)$ [77, 78]:

$$N_i(t) = N_0 e^{-t/\tau_\mu} [1 + AP(t) \cos \theta_i] + N_{\text{bkg}}$$

where $N_i(t)$ is the count rate in detector i , N_0 is the initial count rate, τ_μ is the muon lifetime, A is the asymmetry parameter (determined by detector geometry and positron energy spectrum), and θ_i is the angle between the muon spin and the detector direction. In practice, the normalized asymmetry function $A(t)$ is typically calculated from the counts in opposing detector sets (N_F and N_B) using:

$$A(t) = \frac{N_F(t) - \alpha N_B(t)}{N_F(t) + \alpha N_B(t)}$$

where α is a constant reflecting the relative efficiency of the detectors, chosen such that $A(t) = A_0 P(t)$. The initial asymmetry A_0 contains information about the total fraction of muons contributing to the measured signal. The time evolution of the polarization $P(t)$ contains the specific physics information about the interaction between the muon's magnetic moment and the local fields in the material.

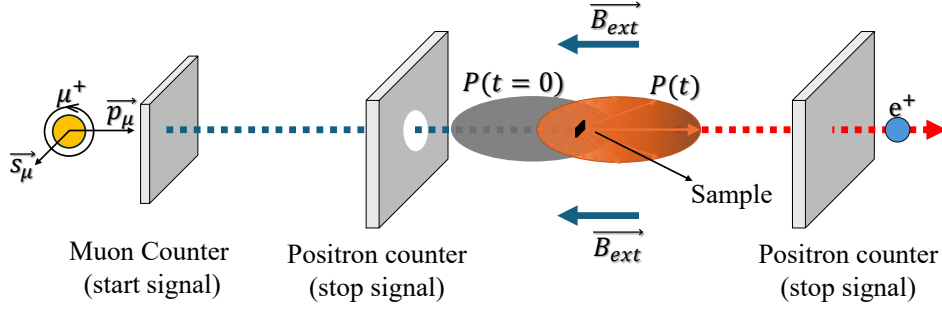


Figure 3.6 Schematic illustration of the muon spin rotation experiment. Spin-polarized muons (μ^+) with initial polarization $P(t=0)$ are implanted into the sample in an external magnetic field B_{ext} . The muon spin s_μ precesses around the local magnetic field ($\mathbf{B}$) at the muon stopping site. The decay positrons (e^+) are detected by positron counters placed opposite to each other, while the muon counter provides the start signal for timing measurements.

Low-Energy Muon Spin Spectroscopy (LE- μ SR)

Conventional surface muons (~ 4.1 MeV) have a stopping range in solids spanning hundreds of micrometers to millimeters. This high energy limits standard μ SR applications to studies of bulk material properties. To extend μ SR to reduced-dimensionality systems, such as thin films (30 nm thick, as studied in the work of this thesis) and interfaces, Low-Energy Muons (LEM) are required. These muons have tunable kinetic energies in the keV range, enabling depth-resolved investigations on the nanometer scale. The experiments were conducted using the LEM spectrometer located on the μ E4 beamline at the Swiss Muon Source (S_μ S) at the PSI, Switzerland. PSI is currently the only facility offering a continuous beam of low-energy muons suitable for regular μ SR user programs at the keV scale [79].

The LEM production process utilizes the muon moderation technique. Surface muons are directed onto a cryogenic moderator (e.g., a thin solid layer of a van der Waals gas like nitrogen or argon, held at ~ 10 K). The muons lose energy until they reach epithermal energies (~ 10 eV), efficiently escaping the moderator layer with nearly 100% polarization preserved. These epithermal muons are then electrostatically accelerated to the desired kinetic energy, ranging from 0.5 keV to 30 keV [79, 77]. The primary advantage of the LEM technique is the ability to precisely control the implantation depth of the μ^+ into the sample. The final implantation energy, enabling depth tuning, is achieved by applying an acceleration or deceleration high voltage to the electrically insulated sample mounted on the cryostat.

The stopping position of the muons is not a single point, but a distribution resulting from the random nature of energy loss and scattering processes during thermalization. The mean implantation depth and the shape of this distribution must be accurately determined for depth-resolved analysis. These depth profiles were estimated using the Monte Carlo TRIM.SP simulations [80, 81].

Data Acquisition and Analysis

In the LEM spectrometer at PSI, the incoming muons are tagged upon crossing an ultra-thin carbon foil, providing the start time (t_0) for the μ SR clock [82]. The decay positrons are then detected by scintillator detectors placed symmetrically around the sample [77]. To ensure high statistical accuracy for the depth-resolved magnetic measurements, approximately 3×10^6 counts were recorded per data point (per implantation energy and temperature step).

The experimental geometry used was the Transverse Field (TF – μ SR) method. In this configuration, an external magnetic field ($\mathbf{B}_{\text{ext}}$) is applied perpendicular (transverse) to the initial muon spin polarization ($\mathbf{P}(0)$). In TF – μ SR, the μ^+ spin precesses coherently around the total local magnetic field ($\mathbf{B}$) experienced at the stopping site. The time evolution of the asymmetry $A(t)$ displays an oscillation at the muon Larmor precession frequency (ω_μ), damped due to inhomogeneities in the local field distribution.

The raw μ SR time spectra were analyzed using the standard μ SR fitting program Musrfit [77, 81, 83]. For the depth-resolved TF – μ SR measurements, the time evolution of the asymmetry $A(t)$ arising from muons stopping in the magnetically ordered layers was fitted using a damped cosine function modulated by a Gaussian envelope [77, 84], $A(t) = A_0 \cos(\gamma_\mu B t + \varphi) e^{-\frac{1}{2}(\sigma t)^2}$. Here, γ_μ is the muon gyromagnetic ratio and φ is the initial phase offset. The parameters extracted from the fit are: A_0 : The initial asymmetry, which is proportional to the fraction of muons stopping in the specific region of the sample contributing to the oscillating signal. In LE – μ SR, the amplitude A_0 is critically important as its energy dependence is used to map the depth-resolved signal contribution (i.e., the depth-dependent magnetic behavior). B : The total local magnetic field magnitude sensed by the muon ensemble, determined by the Larmor precession frequency $\omega_\mu = \gamma_\mu B$. σ : The Gaussian muon spin damping rate (or relaxation rate), which quantifies the spread of the local magnetic field distribution at the muon stopping site (static disorder) or dynamic fluctuations of the field.

Chapter 4

RuO₂ growth modes and its RHEED characterization

4.1 Introduction

This chapter establishes the experimental basis for the rutile RuO₂ thin films used through this thesis, with a specific focus on *in-situ* RHEED as a real-time diagnostic during PLD. While the general PLD methodology and the underlying diffraction concepts are introduced in earlier chapters of the thesis, the emphasis here is on (i) implementing RHEED in the existing deposition environment and (ii) demonstrating how RHEED signatures can be used to distinguish growth modes and assess crystalline surface quality during RuO₂ growth. The ability to monitor surface evolution during deposition is particularly important for RuO₂, where subtle changes in deposition conditions can significantly affect morphology and crystallinity, and consequently, the suitability of the films for subsequent device fabrication and measurements.

4.2 RHEED installation

The aim was to install the RHEED into the PLD chamber to have an in-situ tool to characterize the growth of the thin film. The first challenge was moving the whole PLD chamber and excimer laser as we had limited space to install the electron gun for RHEED on one side of the chamber. The figures 4.1a and 4.1b are the positions of the PLD chamber with respect to the lab before and after. The idea was not to move a lot but still enough to get enough working space around the chamber. There was a risk of losing the already optimized growth conditions for the different materials by moving the chamber. The risk stemmed from the modified laser beam path after the reorientation. The appendix discusses the installation of the RHEED system and the construction of various tools to support the system. After moving the chamber, the growth conditions have not changed for all materials except the RuO₂. So, the RuO₂ growth has been optimized again and characterized with in-situ RHEED.

4.3 RuO₂ thin film growth-mode case study

This section will describe the growth mechanisms of RuO₂ thin film on the rutile-TiO₂ substrate. TiO₂ in the rutile phase has the same crystal structure as RuO₂ as shown in figure 4.2 with lattice parameters $a = b = 4.593 \text{ \AA}$ and $c = 3.959 \text{ \AA}$ [85]. Whereas the unit cell parameters are $a = b = 4.492 \text{ \AA}$ and $c = 3.106 \text{ \AA}$ [86]. TiO₂ with a lattice

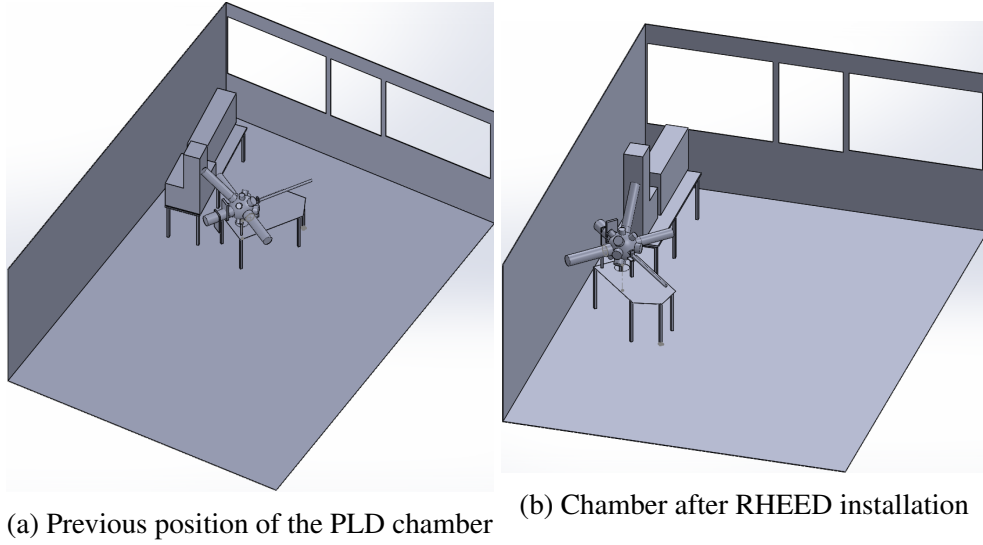
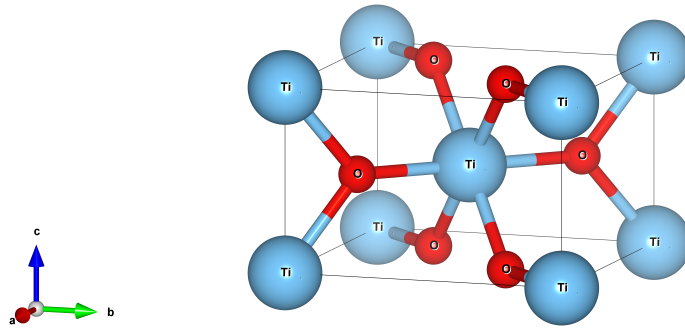


Figure 4.1 Reorientation of the PLD chamber.

mismatch of less than 5%, being diamagnetic and insulating, is a suitable choice for growing RuO₂ epitaxially.

Figure 4.2 Rutile-TiO₂ unit cell [30].

The RuO₂ growth optimization process after the change in the position of the PLD chamber involves polishing the target's surface, variation of growth temperature, and pulse energy of the laser. The new or flat surface of the target proved essential for the growth of the RuO₂ thin film, as well as the combination of growth temperature and laser pulse energy. The typical growth parameters are tabulated in table 4.1:

Table 4.1 Typical growth parameter for RuO₂

Growth variable	
Substrate	TiO ₂ [110]
Base pressure	10 ⁻⁸ mbar
Growth pressure	2 × 10 ⁻² mbar
Laser pulse energy	130 - 150 mJ
Pulse frequency	10 Hz
Background gas	Oxygen
Growth temperature	400 - 450 °C

The growth characterization of RuO₂ thin film on TiO₂ has been characterized by RHEED and complemented with XRD analysis. RHEED is done *in situ* while growing the thin film samples, and XRD is done after the deposition. The time evolution of the RHEED pattern after the deposition of the thin film provides information on the surface development in terms of smoothness and crystalline quality. The results from each technique will help to understand the growth mechanism of thin films.

The capillarity theory for heterogeneous nucleation of a solid film on a planar substrate can explain different growth mechanisms [87]. This theory calculates the change in total Gibb's free energy for thin film nucleation (ΔG). ΔG includes the change in volume free energy ΔG_v and surface free energy (γ) of three entities involved in the process (equation 4.1), i.e., vapor, a nucleus of thin film, and substrate (see fig. 4.3). The contribution in equation 4.1 are proportional to the curved surface area ($a_1 r^2$), the projected circular area on the substrate ($a_2 r^2$), and volume ($a_3 r^3$) of the spherical cap-shaped solid nucleus (see fig. 4.3). Thermodynamic stability occurred at a critical nucleus size and corresponding critical change in total free energy for the nucleation (ΔG^*) given by equation 4.2. By relating the contribution of γ_{nv} , γ_n , γ_{sv} , a_1 , a_2 , and a_3 geometrically to the contact angle (θ) as shown in figure 4.3, we can rewrite equation 4.2 to equation 4.3. The optimal contact angle, for layer-by-layer and island growth is close to zero and more significant than zero, respectively. Equations 4.2 and 4.3 in two limiting cases of the contact angle implies conditions for the different growth mechanisms. The island growth (Volmer Weber) is favorable when the vapor-nucleus and substrate-nucleus surface energy dominate substrate-vapor surface energy, and layer-by-layer (Frank van der Merwe) is favorable when substrate-vapor surface energy dominated over the other two [87]. Between these two extreme cases, an intermediary

state of the hybrid island and layer growth is known as mixed growth. Signatures of all these three types of growth in characterized sub-sections:

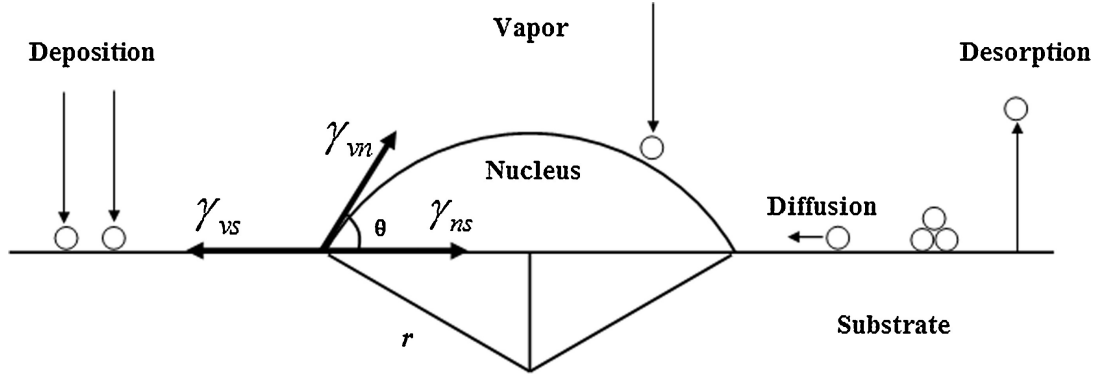


Figure 4.3 Schematic of heterogeneous nucleation on substrate surface during vapor deposition [88].

$$\Delta G = a_3 r^3 \Delta G_V + a_1 r^2 \gamma_{nv} + a_2 r^2 \gamma_{ns} - a_2 r^2 \gamma_{sv} \quad (4.1)$$

$$\Delta G^* = \frac{4(a_1 \gamma_{nv} + a_2 \gamma_{ns} - a_2 \gamma_{sv})^3}{27 a_3^2 \Delta G_V^2} \quad (4.2)$$

$$\Delta G^* = \frac{16\pi (\gamma_{nv})^3}{3 (\Delta G_V)^2} \left\{ \frac{2 - 3 \cos \theta + \cos^3 \theta}{4} \right\} \quad (4.3)$$

RHEED on TiO₂ (110)

The single crystalline and atomically flat substrate are two of the essential requirement for the growth of crystalline thin films. The figures 4.4a and 4.4b shows the RHEED pattern and the RHEED surface plot of the TiO₂ (110) oriented substrate with incoming electron beam along [001] direction of the substrate. The RHEED patterns are not fully symmetric because alignment is done by hand and incoming electron beam is not exactly parallel to the [001] direction of the substrate. For better visualization of the RHEED pattern, it is shown along with the surface plot of the same image throughout the thesis. The presence of RHEED spots in figure 4.4a and 4.4b is the characteristic signature of atomically flat single crystalline substrate [72]. The very bright spot on top is from a part of the electron beam not hitting the substrate. The different intensity of different diffraction spots is because of finite electron penetration beneath the surface. Then the intensity is modulated along the reciprocal rods, resulting in some diffraction

spots being stronger and others being weaker. Therefore, interference between waves scattered from various atomic layers causes the varying intensities of different places [72]. Hence, this substrate is a good candidate for the possible epitaxial growth of the thin film.

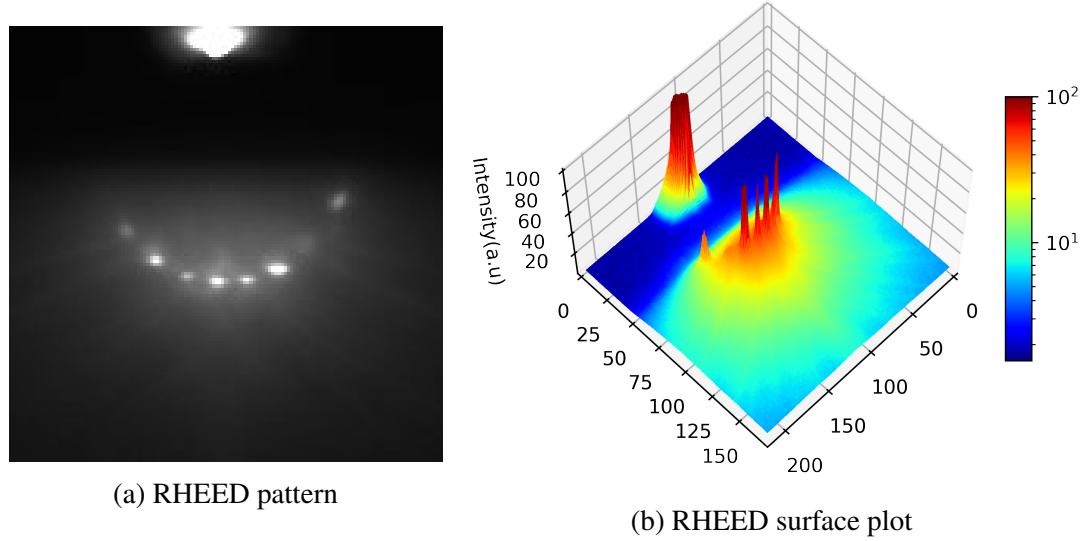


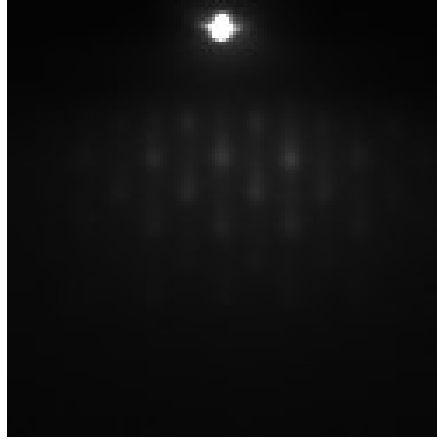
Figure 4.4 RHEED pattern TiO₂ substrate.

4.3.1 Volmer Weber or island growth

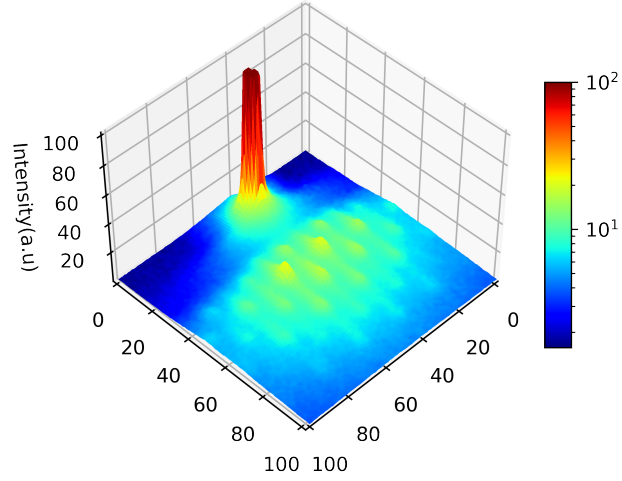
The sample discussed in this sub-section is grown at the growth temperature of 450 °C with the pulse energy of 150 mJ, and other growth parameters are the same as in table 4.1. While growing the thin film, the RHEED diffraction pattern was monitored and the sample was characterized with XRD after the deposition, as shown in figure 4.6b. Figure 4.5 shows the RHEED pattern after 40 seconds of growth and one obtained after the growth which took 250 seconds.

The RHEED pattern changes from RHEED spots along a circular arc to transmission diffraction spots distributed along a circular arc after 40 seconds, as shown in figure 4.5a and 4.5b. The RHEED pattern corresponds to the transmission diffraction pattern of the electron (see fig. 3.5) from the 3D crystals on the substrate surface. The diffraction spot intensity is relatively small compared to the RHEED spots from the bare substrate (fig. 4.4). This reduced intensity of diffraction spots is possible due to the increased roughness of the surface. The transmission diffraction spots are even more clearly present after the deposition of the thin film. There was a total of 2500 laser pulses with an expected thickness of 17 nm from the previous experience. These

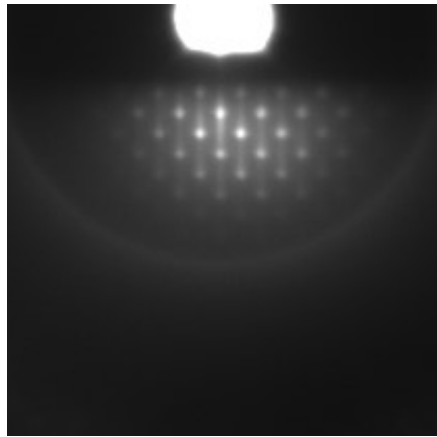
diffraction patterns signify that the island growth mechanism leads to rougher thin films, as we may expect from the thin film grown in a layer-by-layer fashion.



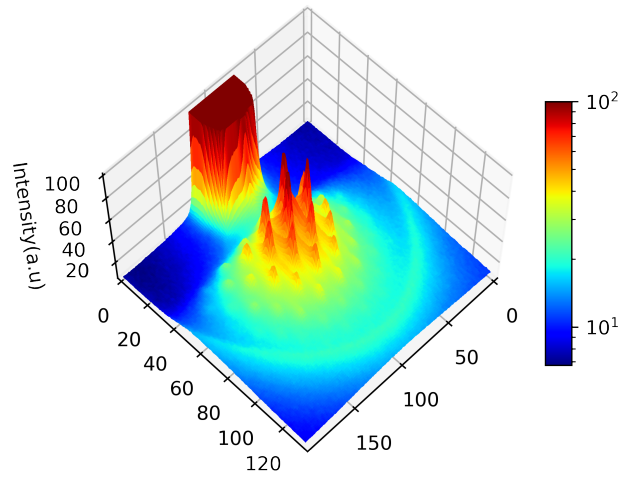
(a) RHEED after 40 sec of deposition



(b) RHEED surface plot after 40 sec of deposition



(c) RHEED after deposition completed



(d) RHEED surface plot after deposition completed

Figure 4.5 RHEED pattern for Volmer Weber growth mechanism.

X-ray characterization

After the thin film deposition, it is checked for the quality of the thin film with XRR and the $2\theta/\theta$ scan from the XRD. Figure 4.6a shows the XRR scan for the sample, which shows no oscillating intensity. Figure 4.6b shows the $2\theta/\theta$ scan from the sample, with the substrate [110] in the diffraction plane. Along with a sharp substrate peak, there is a broad and weak peak from the thin film to the right of the the substrate

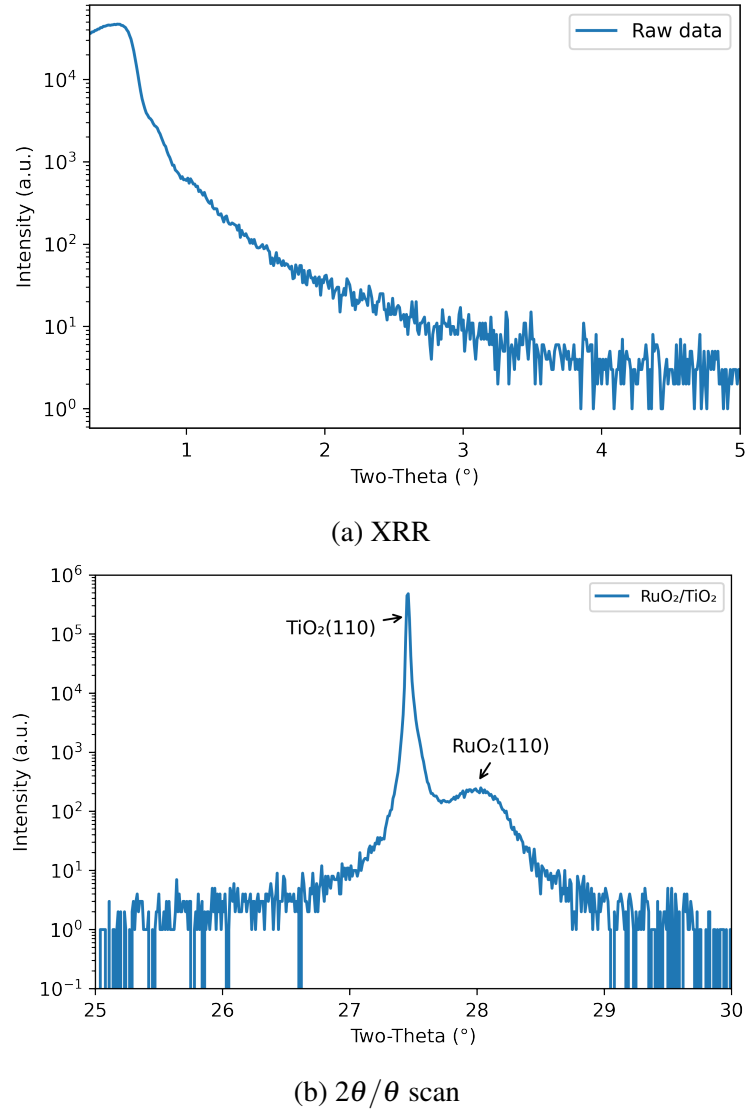


Figure 4.6 X-Ray characterization of thin film with island growth.

peak . This reflection is the only other reflection observed in the $2\theta/\theta$ scan from 20° to 90° . The absence of XRR oscillation could be either due to no deposition of thin film or film thickness of hundreds of nanometer or a rough, thin film. The thin film reflection can eliminate the possibility of no deposition in the $2\theta/\theta$ scan, and high film thickness is also not possible due to the expected deposition rate of thin film from the previous experience of RuO₂ growth. Hence, the thin film is expected to be rough, consistent with the observation of the RHEED pattern. The presence of only one reflection in $2\theta/\theta$ scan signifies the single out-of-plane orientation of the thin film and in accordance with RHEED transmission spots from 3D crystals.

The high roughness of thin film is attributed to the growth temperature due to the high mobility of the deposited atoms [89]. The growth at lower substrate temperature is expected to improve the thin film's surface quality. Still, we need a relatively high temperature to assist epitaxial growth of the thin film to have better thin film crystalline quality [90].

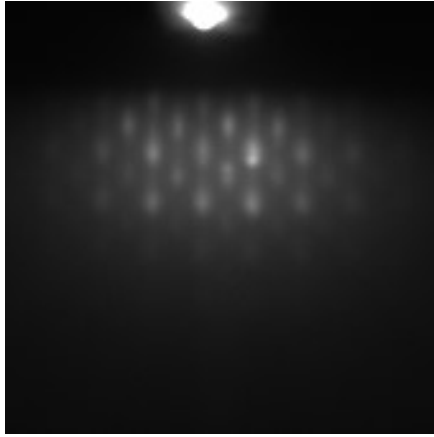
4.3.2 Mixed growth

The sample discussed in this sub-section is grown at the growth temperature of 400 °C with the pulse energy of 150 mJ, and other growth parameters are the same as in table 4.1. The RHEED diffraction pattern was monitored while the thin film was developing, and it was later characterized by XRD, as shown in figure 4.8. Figure 4.7 shows the RHEED pattern after 20 seconds of growth (thickness $\approx$ 1.4 nm) and the after completion of the growth (thickness $\approx$ 17 nm).

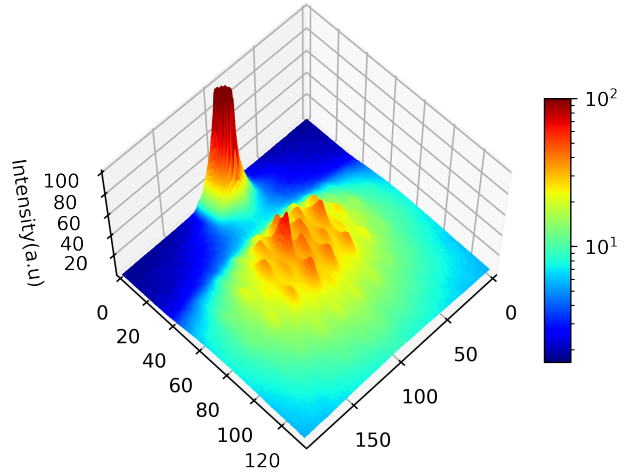
After 20 seconds of deposition, the RHEED pattern evolves from RHEED spots along a circular arc to transmission diffraction spots distributed along a circular arc, as seen in figures 4.7a and 4.7b. The transmission diffraction pattern of the electrons from the island stacking of the atoms on the substrate surface conforms to the RHEED pattern from figure 3.5. The transmission diffraction spots further evolved to the RHEED streaks after the deposition of the thin film, as seen in figures 4.7c and 4.7d. Such diffraction streaks are characteristic of the multi-grain surface with finite but low roughness (see fig. 3.5). There have been 2500 laser shots in total to deposit this sample. This evolution of diffraction patterns signifies that growth started in island fashion and evolved to the layer-by-layer fashion at the end of the deposition. This sample is expected to have a better surface than the sample grown in the Volmer Weber mechanism.

X-ray characterization

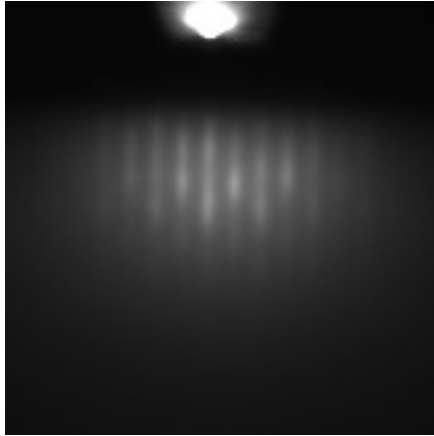
After the thin film is deposited, its quality is examined using the XRR and the $2\theta/\theta$ scan from the XRD. Figure 4.8a shows the XRR scan for the sample, and fitted data is simulated with the LEPTOS software package [91]. Figure 4.8b shows $2\theta/\theta$ scan from the sample, with the substrate [110] in the diffraction plane. There is a shoulder reflection to the sharp substrate peak from the thin film. Asymmetric Laue oscillations accompany the thin film peak in the scan only on the right side. The simulated XRR oscillation gives film thickness and surface roughness of 171.2 ± 3.5 Å and 11.3 ± 0.8 Å. The presence of Kiessig fringes in the XRR scan and fitted surface



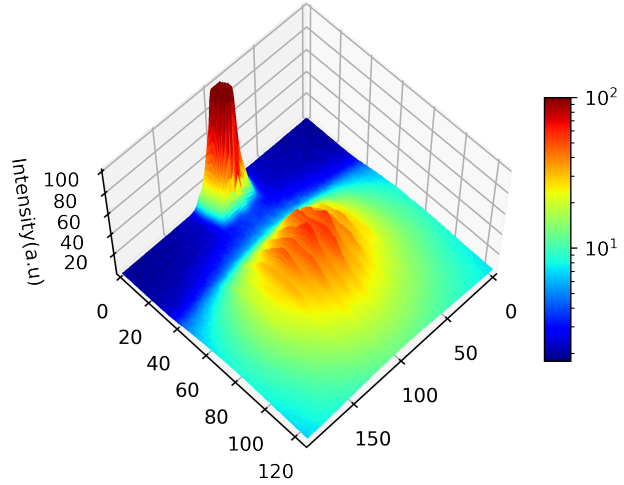
(a) RHEED after 20 sec of deposition



(b) RHEED surface plot after 20 sec of deposition



(c) RHEED after deposition completed

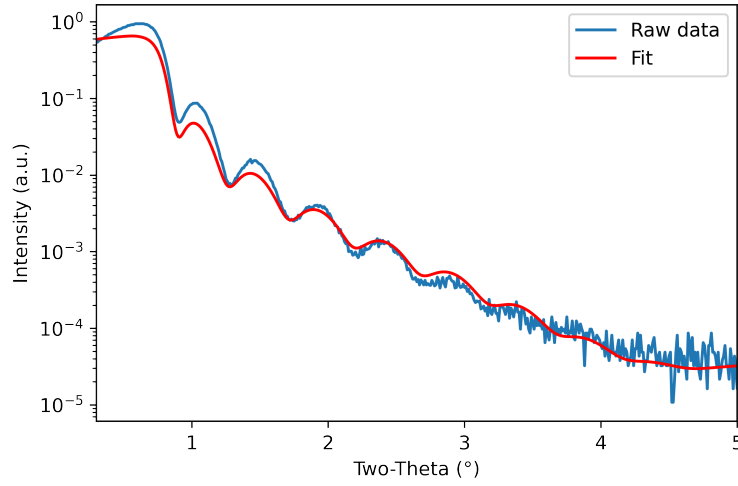


(d) RHEED surface plot after deposition completed

Figure 4.7 RHEED pattern for mixed growth mechanism after (thickness ≈ 1.4 nm) and (thickness ≈ 17 nm).

roughness of around $11 \text{ \AA}$ is consistent with the observation of the RHEED pattern with streaks. The only one reflection in $2\theta/\theta$ with asymmetric satellite peaks scan signifies the single crystalline thin film with asymmetric strain relaxation [92]. The satellite peaks are simulated with the Laue function [93] in figure 4.8b. Forty-nine lattice planes of inter-planar spacing of $3.205 \text{ \AA}$ contribute to the simulated Laue function. These planes give crystallite thickness perpendicular to the surface, approximately 0.92 times the whole thin film thickness from the XRR. This estimate of thickness from simulated Laue function is reasonable as Laue function estimates the crystallite

size and should always be lower than the thickness from the XRR as from XRR gives the total thickness of the thin film irrespective of crystallite size.



(a) XRR

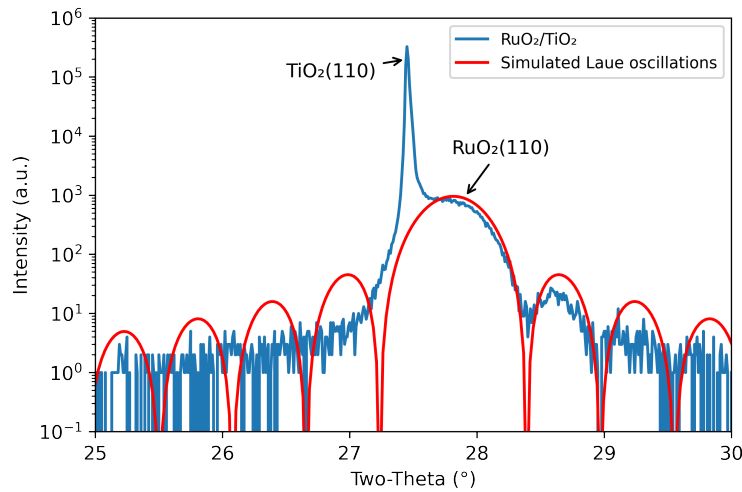
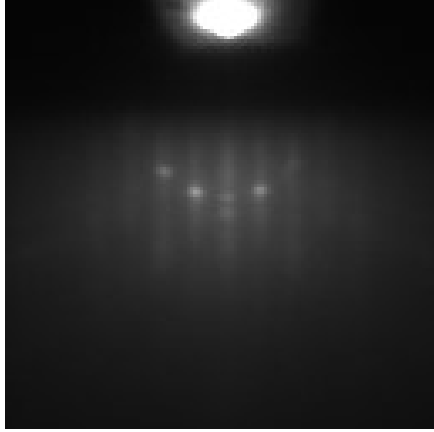
(b) $2\theta/\theta$ scan

Figure 4.8 X-Ray characterization of thin film with mixed growth.

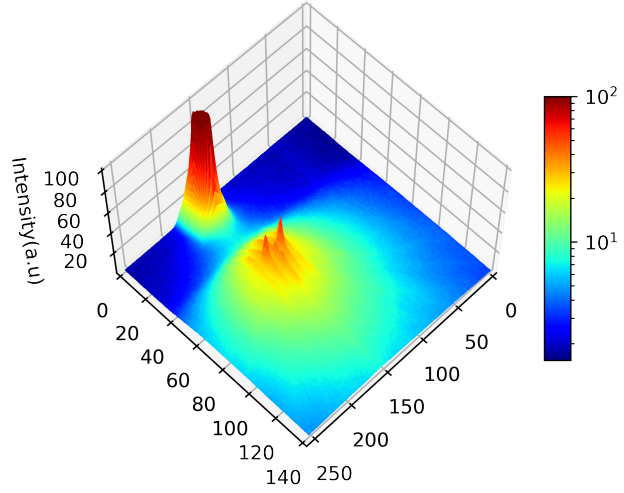
4.3.3 Frank van der Merwe or layer-by-layer growth

The sample mentioned in this subsection is grown using the same growth conditions as those listed in table 4.1, besides a growth temperature of 400 °C and pulse energy of 130 mJ total of 2500 shots. The RHEED diffraction pattern was monitored while the thin film was developing, and the sample was later characterized by XRD, as

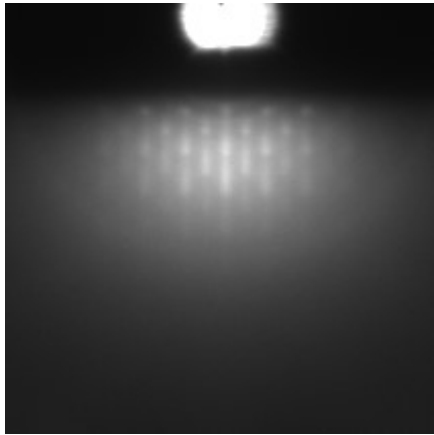
shown in figure 4.11. Figure 4.9 shows the RHEED pattern after 20 seconds of growth (thickness ≈ 1.4 nm) and after the completion of the growth (thickness ≈ 17 nm).



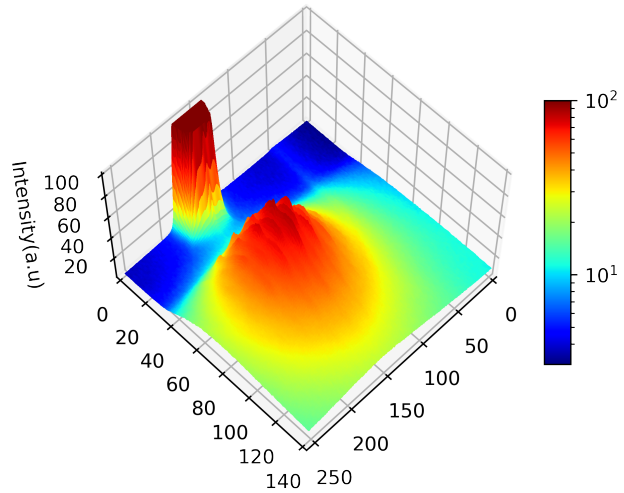
(a) RHEED after 20 sec of deposition



(b) RHEED surface plot after 20 sec of deposition



(c) RHEED after deposition completed



(d) RHEED surface plot after deposition completed

Figure 4.9 RHEED pattern for Frank van der Merwe growth mechanism after (thickness ≈ 1.4 nm) and (thickness ≈ 17 nm).

After 20 seconds of deposition, the RHEED pattern evolves from RHEED spots along a circular arc to RHEED streaks with oscillating intensity, as seen in figures 4.9a and 4.9b. The area integrated intensity of three regions shown in figure 4.10a is plotted in figure 4.10b as a function of time. The decrease in intensity immediately after starting the deposition is attributed to the increased surface roughness. The RHEED diffraction streaks further evolved to the RHEED streaks with modulated intensity

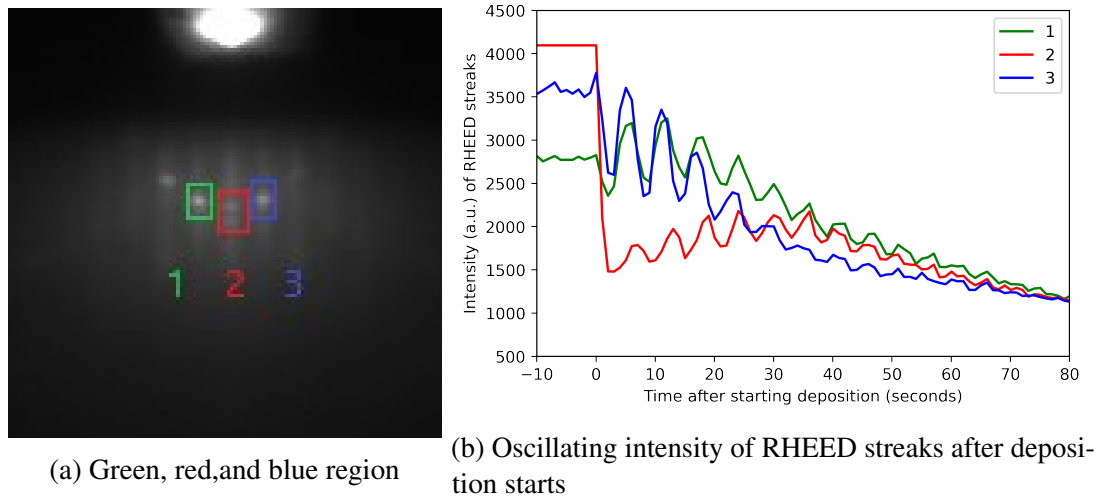
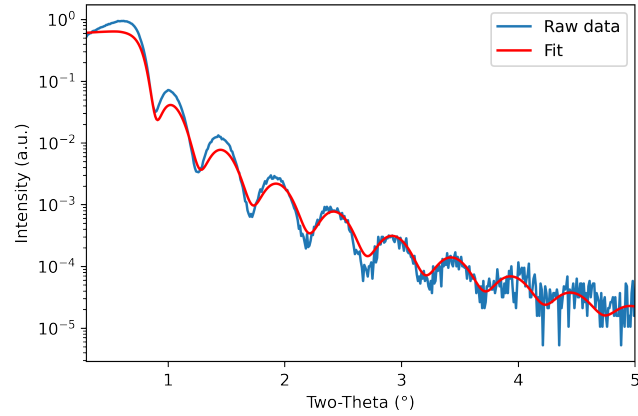


Figure 4.10 Oscillating integrated intensity of RHEED streaks.

in space after the deposition of the thin film, as seen in figures 4.9c and 4.9d. The oscillating intensity at the beginning of deposition in time and modulated diffraction streaks after the deposition are characteristic for layer-by-layer growth fashion and single crystalline multi-stepped surface [72]. The fast-Fourier transform of the first six oscillations gives a time period of 6.4 seconds of mono-layer formation time. This mono-layer formation time estimates the total deposited thickness of 125.2 Å. This sample is expected to have even better surface quality than the sample grown in the mixed mechanism.

X-ray characterization

Using the XRR and the $2\theta/\theta$ scan from the XRD, the thin film's quality is examined after it has been deposited. The fitted data is simulated using the LEPTOS software package, and the figure 4.11a depicts the XRR scan for the sample. Figure 4.11b shows the substrate [110] is shown in the diffraction plane taken from a $2\theta/\theta$ scan of the sample. There is a shoulder reflection to the sharp substrate peak from the thin film. The thin film peak in the scan is only accompanied by symmetric Laue oscillations. The simulated XRR oscillation gives film thickness and surface roughness of 169.8 ± 2.3 Å and 6 ± 1.3 Å. The presence of Kiessig fringes in the XRR scan and fitted surface roughness of around 6 Å is consistent with the observation of multi-stepped surface due to modulated RHEED streaks. The only one reflection in $2\theta/\theta$ with symmetric satellite peaks scan signifies the single crystalline thin film with no relaxation [92]. The symmetric satellite peaks are simulated with the Laue function [93] in figure



(a) XRR

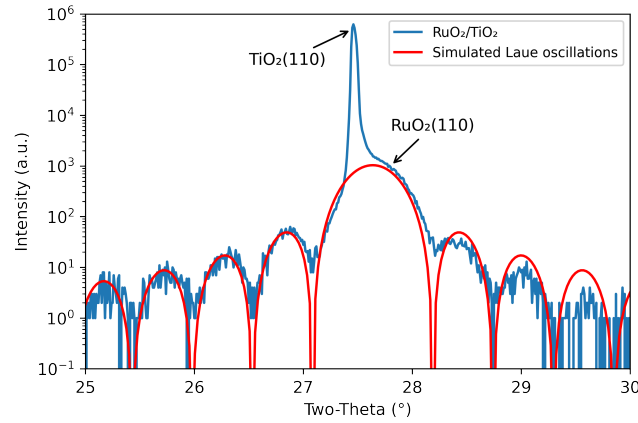
(b) $2\theta/\theta$ scan along (110) plane

Figure 4.11 X-ray characterization of thin film with layer growth.

4.11b. Fifty-one lattice planes of inter-planer spacing of 3.225 Å contribute to the simulated Laue function. From the peak in 2θ , we get a lattice constant of $a = b = 4.561$ Å, which is different from the bulk value. Therefore, we conclude that the film is coherently strained. These planes give crystallite thickness perpendicular to the surface, approximately 0.97 times the whole thin film thickness from the XRR. This estimated crystallite size is even better than mixed growth.

The layer-by-layer growth fashion with the multi-stepped surface, as characterized by the RHEED analysis, conforms with symmetric Laue oscillations and Kiessig fringes in the XRD analysis. The average multi-stepped surface is two-stepped as surface roughness from XRR is about two times the inter-planer spacing. The sample is a single crystalline with epitaxial growth as symmetric Laue oscillations illustrate uniform lattice constant across the thin film thickness. The estimated thickness from

the RHEED oscillations is smaller than the estimation from the Kiessig fringes or Laue oscillations, which match very well. Such difference has been reported before where RHEED oscillation frequency does not match the monolayer formation time [94]. However, these results are not well understood and attributed to defect formation. This conspiracy could be explained by a difference in the sticking coefficient of the first tens of layers and the remaining layers. This justification is reasonable that monolayer formation time from RHEED is calculated from only the first few monolayers. The validity of this explanation will be tested by growing thinner films in the continuation of this thesis work.

4.4 Collaborative Works

This subsection summarizes three collaborative studies to which I contributed through the fabrication and optimization of specialized samples. My primary role was the growth of high-quality epitaxial RuO_2 thin films on TiO_2 substrates using PLD. In these preparations, careful control of film thickness, crystallinity, and strain was required to support the advanced spectroscopic and optical measurements carried out by our collaborators. Although these samples also enabled additional investigations that are not discussed here, the three articles highlighted below are central to the thesis narrative because they collectively report altermagnetic features in RuO_2 .

In the first study, Fedchenko *et al.*[38] used soft X-ray angle-resolved photoemission spectroscopy (ARPES) to probe the electronic structure of the epitaxial films. The work reports signatures consistent with time-reversal symmetry breaking in the band structure without a net magnetization, discussed there as a hallmark of the altermagnetic phase in RuO_2 .

Building on this, Lytvynenko *et al.*[95] investigated the element-specific magnetic response using magnetic circular dichroism in X-ray photoelectron spectroscopy and X-ray magnetic circular dichroism (XMCD). The study links the epitaxial strain of the films to crystal-field distortions and discusses these distortions as relevant for stabilizing altermagnetic order, enabling dichroic signals at the Ru core levels.

Finally, Weber *et al.*[96] leveraged the optical quality of the films to demonstrate all-optical excitation of spin polarization. By varying the linear polarization of the optical pump, the reported response follows an angular dependence that the authors interpret as consistent with the characteristic d -wave symmetry discussed for altermagnetic band structures.

4.5 Conclusion and outlook

In this chapter, RHEED was established as an *in-situ* probe for PLD growth and applied to re-optimize and systematically characterize RuO₂ thin films on rutile TiO₂(110). Implementing RHEED required reorienting the PLD chamber and laser path, which altered previously optimized conditions and necessitated renewed growth optimization. This outcome highlights the sensitivity of RuO₂ growth to target condition, substrate temperature, and laser pulse energy. With real-time feedback during deposition, RHEED could be directly correlated with *ex-situ* XRR and XRD, enabling a consistent assessment of surface morphology and crystalline quality.

Overall, combining *in-situ* RHEED with *ex-situ* X-ray analysis provided an internally consistent workflow to recover RuO₂ growth after the chamber modification and to identify a growth window yielding improved epitaxial quality and surface morphology. Establishing this level of growth control is essential for the subsequent thesis chapters, which use RuO₂ thin films as a platform where interface quality and crystallographic definition are critical for interpreting transport, device, and spectroscopy results.

The collaborative studies summarized in Sec. 4.4 further motivate this direction by reporting altermagnetic features in RuO₂ using experiments that rely on well-defined epitaxial films. Building on this foundation, the next two chapters will present my experiments on RuO₂ thin films, with a focus on how interfaces and surface states influence the magnetic signatures discussed for this material system.

Chapter 5

Interface Spin Current in RuO₂/Py Heterostructures

5.1 Introduction

This chapter presents one of the experimental results of the thesis by testing the consequences of spin currents associated with altermagnetism in RuO₂ via angle-dependent magnetoresistance (ADMR) in epitaxial RuO₂/Py heterostructures (Py $\equiv$ Ni₈₀Fe₂₀, Permalloy). The RuO₂/Py platform is used precisely to test whether the magnetoresistive symmetry is governed by bulk spin-current generation in RuO₂ or by spin-orbit scattering localized at the RuO₂/Py boundary. The experimental strategy in this chapter follows the same logic: emphasize symmetry, sign, and systematic trends as the most direct fingerprints of the underlying conversion mechanism. It translates the broad questions raised in Chapter 1 (Background and Motivation) into specific, testable results and links them to the methodological principles outlined in Chapter 2 (Methodology). By emphasizing the data and presenting a concise, results-first narrative, this chapter lays the groundwork for the dissertation.

The analyses reported here address two linked objectives: (i) to determine whether the origin of the ADMR in RuO₂/Py is consistent with an interface-generated spin-current (IGSC) mechanism rather than a purely bulk spin Hall magnetoresistance (SMR) response or altermagnetic spin-split magnetoresistance; (ii) to map systematic trends versus film thickness, device crystallographic-orientation, temperature, and magnetic field. Together, these objectives test the central hypothesis that symmetry breaking and spin-orbit coupling at the oxide/ferromagnet (FM) boundary can set the overall sign and angular symmetry of the magnetoresistive response.

To maintain focus on the findings, the theory is presented briefly, and further details can be found in the article about this work, which outlines how interfacial boundary conditions and bulk diffusion pathways shape ADMR symmetries [97]. Where appropriate, the discussion connects the present results to the broader literature on spin-orbit conversion, providing context without duplicating the comprehensive survey given in Chapter 1 (Background and Motivation).

5.2 Results

This section assembles the experimental evidence supporting an IGSC spin-charge conversion origin for the ADMR observed in RuO₂/Py heterostructures. We proceed from structure to transport: first establishing epitaxy, surface quality, and interface morphology; then defining measurement geometries and presenting baseline magneto-transport; and finally mapping systematic trends versus crystallographic orientation,

temperature, magnetic field, and nominal thickness. Growth and device fabrication details are provided in Chapter 2 and referenced where appropriate to keep the narrative focused on the data and their interpretation.

The opening subsections document structural validation by XRD, RHEED, and atomic force microscopy, followed by out-of-plane R_{xx} and R_{xy} sweeps that set the ferromagnetic and transport baselines. We then introduce the three ADMR rotation planes (α in-plane, γ in xz , and β in yz) and present the central observations, emphasizing the robust negative ADMR emerging in the β -plane. The latter subsections examine how these signatures evolve with bar orientation ($J_C \parallel c$ vs. $J_C \perp c$), with temperature (20 K to 300 K), and with applied field (0.95 T to 5 T), and assess reproducibility across devices and thicknesses. The control measurements on bare Py and targeted artifact checks ruled out alternative explanations.

5.2.1 Structural and Surface Characterization

The structural figure for this section brings together three complementary diagnostics. The XRD panel highlights the single-orientation out-of-plane texture: as thickness increases, the RuO_2 film peak grows relative to the TiO_2 substrate peaks, evidencing coherent growth along the film normal and excluding secondary orientations. The RHEED pattern exhibits a transmission-type pattern, indicating crystallinity, and also suggests island growth. Finally, the atomic force microscopy quantifies the roughness contrast between stacks with and without RuO_2 and documents sparse tall crystallites that are geometrically prominent yet electrically irrelevant due to their lack of lateral connectivity. Together, these data establish the structural basis for interpreting interfacial transport: the films are epitaxial and crystalline, with moderate roughness.

X-Ray Diffraction (XRD): $\text{RuO}_2(100)$ Epitaxy on TiO_2

We first verified the crystalline quality and interface integrity of the $\text{TiO}_2(100)/\text{RuO}_2(100)/\text{Py}/\text{Al}$ heterostructures. The heterostructures are covered with 2 nm of Al, which oxidizes and protects the sample from the environment, while not contributing to transport. These measurements are presented in Fig. 5.1. High-resolution X-ray diffraction (XRD) with the scattering vector normal to the substrate shows clear evidence of epitaxial $\text{RuO}_2(100)$ growth on rutile $\text{TiO}_2(100)$: the $\text{RuO}_2(100)$ film peak is discernible already at low thickness and strengthens

systematically as the RuO₂ layer is thickened, consistent with improved diffracting volume and coherent out-of-plane texture. The substrate reflections dominate the scan at the lowest thickness, but the RuO₂ reflections become progressively more pronounced, establishing single out-of-plane orientation without detectable secondary RuO₂ polymorphs within our resolution. Extended scans and side-by-side comparisons for TiO₂/RuO₂/Py/Al and TiO₂/Py/Al samples are provided in Fig. 5.2. This confirms the presence of only one out-of-plane orientation for crystallites of RuO₂(100).

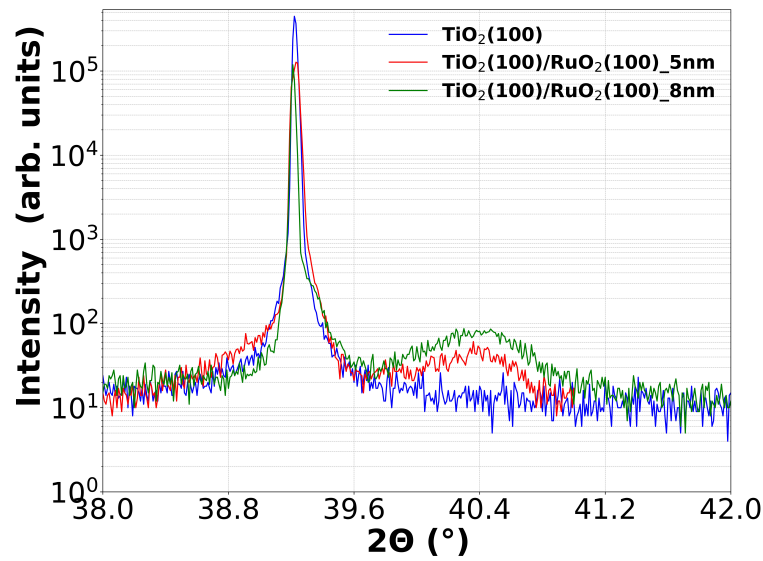


Figure 5.1 XRD patterns of TiO₂(100)//RuO₂(100)/Py films measured with the scattering vector normal to the (100)-oriented rutile substrate.

RHEED Patterns: In-Plane Ordering and Surface Quality

Figure 5.3 presents the RHEED images of these measurements. Figure 5.3(a): RHEED after RuO₂ deposition for the electron beam aligned along TiO₂[001]. The pattern remains well-defined, confirming in-plane ordering, yet evolves toward a transmission-like signature with broadened streaks/spots, signaling increased effective roughness relative to the substrate. This evolution is typical for conductive oxides grown at moderate temperatures by PLD. The key takeaway for the transport sections is that the interface is structurally abrupt and crystalline but not atomically flat. Figure 5.3(b): substrate-only RHEED (reference panel) exhibits sharp features characteristic of a smooth TiO₂(100) surface.

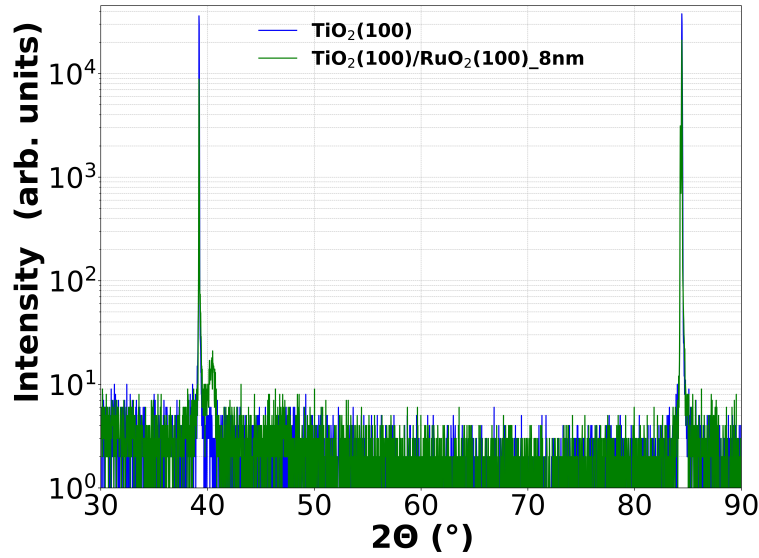


Figure 5.2 XRD on $\text{TiO}_2/\text{RuO}_2/\text{Py}/\text{Al}$ and $\text{TiO}_2/\text{Py}/\text{Al}$ samples.

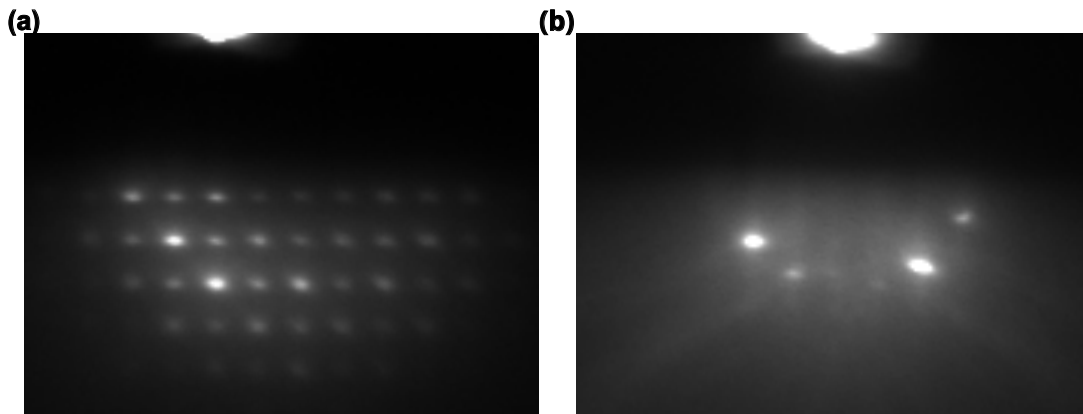


Figure 5.3 RHEED images measured on (a) $\text{TiO}_2/\text{RuO}_2$ and (b) TiO_2 .

Atomic Force Microscopy: Roughness and Morphology

Figures 5.4(a) and 5.4(b) show the measured height maps for $\text{TiO}_2/\text{RuO}_2[8\text{ nm}]/\text{Py}[4\text{ nm}]/\text{Al}$ and $\text{TiO}_2/\text{Py}[4\text{ nm}]/\text{Al}$, acquired over identical microscopy scan parameters. The quantitative comparison emphasizes two points. First, the root-mean-square (rms) roughness increases from $\sim 0.4\text{ nm}$ (without RuO_2) to $\sim 2.1\text{ nm}$ (with RuO_2), capturing the net morphological impact of the oxide layer. Second, the RuO_2 stack exhibits isolated nanoscale crystallites that reach up to $\sim 70\text{ nm}$ in height. Excluding features above 20 nm from the rms calculation prevents these visually striking protrusions from biasing the roughness estimates. This separation between geometric outliers and the continuous background is crucial for

transport, as conduction predominantly follows the laterally connected film rather than isolated spikes. These microscopy results help contextualize the RHEED indication of non-atomically flat surfaces.

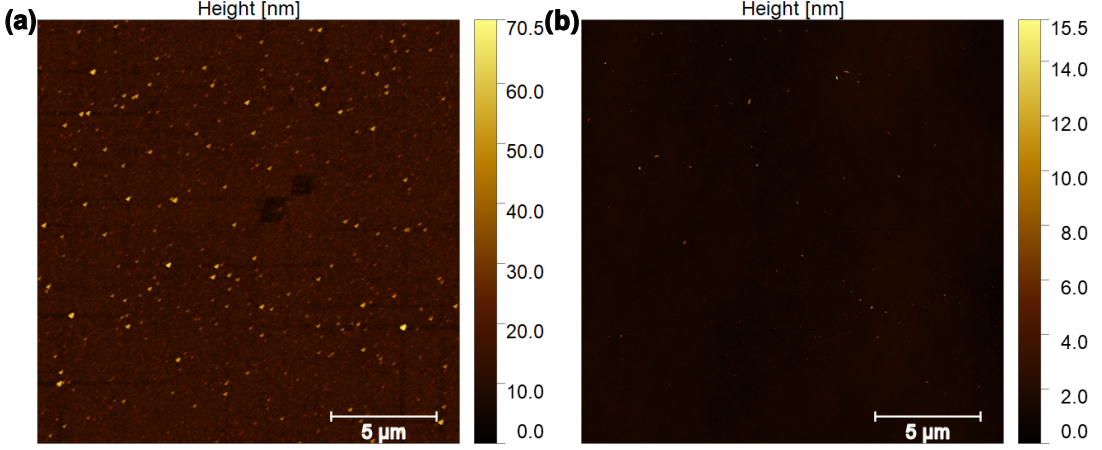


Figure 5.4 Atomic force microscopy images measured on (a) TiO₂/RuO₂/Py/Al and (b) TiO₂/Py/Al.

5.2.2 Baseline Transport: Out-of-Plane R_{xx} and R_{xy} Sweeps

Before turning to ADMR, we establish the magnetic and transport baselines from out-of-plane field sweeps at 20 K over ± 5 T in Hall bar devices (geometry and delta-mode protocols as described in Chapter 2). These baseline transport measurements are presented in Fig. 5.5. The longitudinal channel, $R_{xx}(H_z)$, exhibits the monotonic evolution characteristic of AMR in the FM, validating the expected rotation of the magnetization toward the film normal under increasing field. The transverse channel, $R_{xy}(H_z)$, exhibits a sizable AHE whose sign indicates a dominant electron-like carrier, consistent with Py. Because the AHE and planar Hall contributions dominate R_{xy} in this bilayer, we do not analyze SMR-like signatures in the Hall response and instead use R_{xy} primarily as a ferromagnetic proxy for magnetization orientation in subsequent ADMR sections. These baselines fix the magnetization calibration used below when interpreting the ADMR angular dependencies. An obvious small asymmetry in longitudinal in Fig. 5.5 for opposite field direction is present and traceable to finite transverse resistivity pickup from minor lead misalignment.

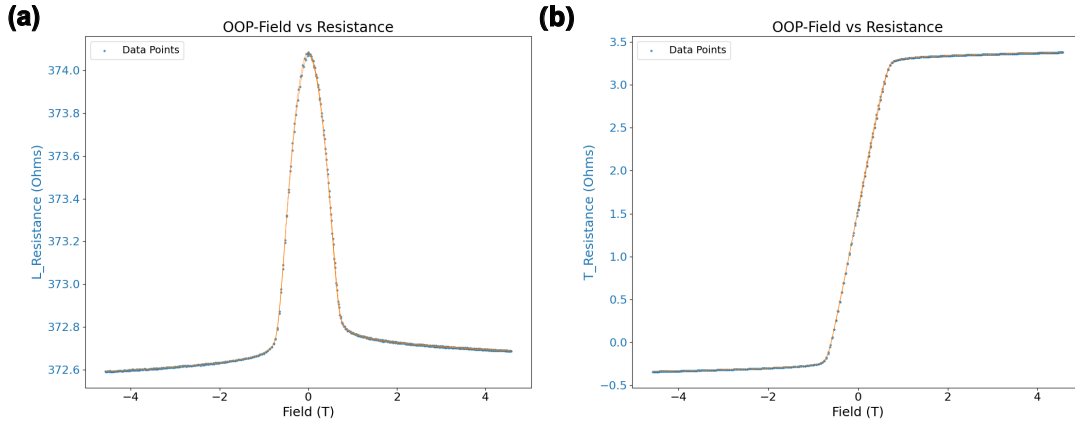


Figure 5.5 Out-of-plane field sweep for the $\text{RuO}_2(100)[5 \text{ nm}]/\text{Py}$ bilayer showing (a) longitudinal resistance R_{xx} and (b) transverse resistance R_{xy} .

5.2.3 ADMR Measurement Geometries (α , β , γ Planes)

ADMR was recorded by rotating a fixed magnetic field within three orthogonal planes while monitoring the longitudinal resistance of six-terminal Hall bars (measurement wiring, delta-mode protocols, and rotation definitions as described in Chapter 2). Figure 5.6 represents the measurement geometry. The α -plane corresponds to in-plane rotation (xy), the γ -plane to rotation in the xz-plane, and the β -plane to rotation in the yz-plane. Figure 5.7(a,b) shows the angular sweeps for RuO_2/Py alongside a bare Py control measured under identical conditions, highlighting the distinct symmetry and sign in the β -plane for the bilayer. At $T = 20 \text{ K}$ and $\mu_0 H = 0.95 \text{ T}$, the $\text{RuO}_2(100)[5 \text{ nm}]/\text{Py}$ heterostructure exhibits relative resistance changes of order -3×10^{-3} in the α -plane, -4×10^{-3} in the γ -plane, and -1×10^{-3} in the β -plane (all referenced to the $\alpha = 0^\circ/\gamma = 0^\circ/\beta = 0^\circ$ states). In stark contrast to α - and γ -planes, the β -plane response in bare Py is negligible under the same conditions, while the bilayer shows a pronounced negative ADMR.

ADMR in the α -Plane (In-Plane Rotation)

In-plane rotation (α -plane) for RuO_2/Py : the α -dependence follows the familiar twofold pattern also observed in bare Py on TiO_2 . The resistance is larger when the magnetization is more parallel to the current and smaller when it is transverse, consistent with conventional AMR in the FM. Figure 5.7(a,b) shows that the angular line shape and phase are essentially identical between the bilayer and the Py control, establishing that interfacial contributions do not qualitatively alter the α -plane symme-

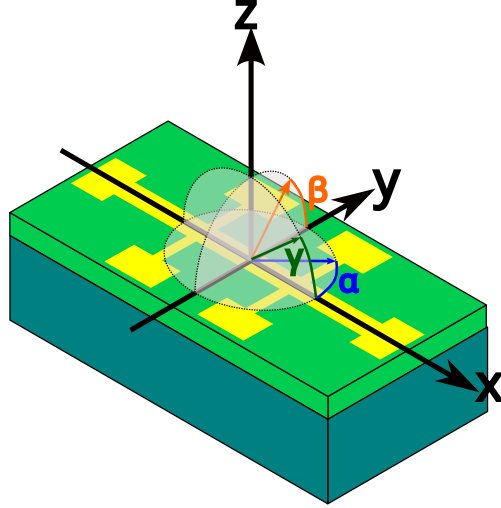


Figure 5.6 Longitudinal resistance measurement scheme for the α -, β -, and γ -planes with magnetic-field sweeps.

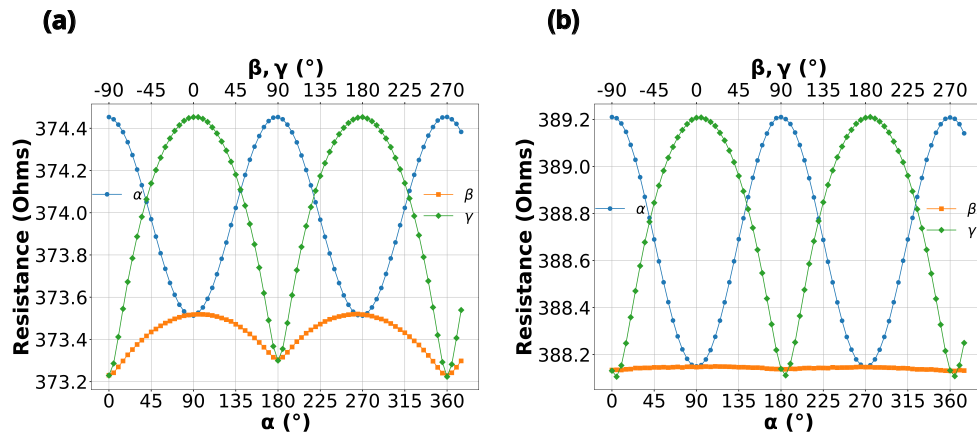


Figure 5.7 Longitudinal resistance variations for magnetic-field direction in the α -, β -, and γ -planes: (a) RuO₂(100) [5 nm]/Py heterostructure and (b) Py thin film measured at 20 K, for J_C being parallel to the c -axis under a 0.95 T magnetic field.

try. This baseline will be used below to isolate the interfacial signature that appears most clearly in the β -plane.

ADMR in the γ -Plane (XZ Rotation)

For rotation in the xz-plane (γ -plane), the RuO₂/Py device again shows a large twofold modulation comparable to the α -plane amplitude at 20 K, with a relative change of $\sim -4 \times 10^{-3}$. Figure 5.7(a,b) shows the γ -sweep next to the α -sweep to emphasize that both predominantly reflect ferromagnetic AMR symmetry in our geometry, providing a high-contrast reference against which the out-of-plane response in the β -plane can be contrasted. The agreement between bilayer and bare Py in the γ -plane further supports the interpretation that any unconventional symmetry should be sought in the β -plane response rather than in in-plane α -plane rotations. Having established the α and γ responses as AMR-like references, we now turn to the out-of-plane β -plane where the sign inversion emerges.

ADMR in the β -Plane (YZ Rotation): Negative ADMR

The key observation of this chapter is a robust *negative* ADMR state for β -plane rotations: the longitudinal resistance is *lower* when the magnetization is driven out of plane than when it is in plane, as in Fig. 5.7(a,b). This sign is opposite to expectations from conventional SMR/Rashba–Edelstein magnetoresistance (REMR)/anomalous Hall magnetoresistance (AHMR) scenarios that typically yield a higher-resistance out-of-plane state. The line shape exhibits a 90° phase offset relative to the SMR baseline, resulting in a negative ADMR. At 20 K the peak amplitude is $\sim -1 \times 10^{-3}$ and is strongly temperature dependent (see Fig. 5.9), while its dependence on field strength is weak for $\mu_0 H$ between 0.95 T and 5 T (see Fig. 5.12).

Crystallographic Orientation Dependence (Hall Bars $J_C \parallel c$ vs. $J_C \perp c$)

To test for crystallographic spin-split altarmagnetic magnetoresistance fingerprints, we patterned orthogonal Hall bars with current along RuO₂[001] ($J_C \parallel c$) and along RuO₂[010] ($J_C \perp c$) on the same films, as shown in Fig. 5.8. Figs. 5.9 and 5.10 show the two orientation datasets side by side for the thicknesses RuO₂[5 nm]/Py and RuO₂[8 nm]/Py, respectively, emphasizing the near independence of bar orientation.

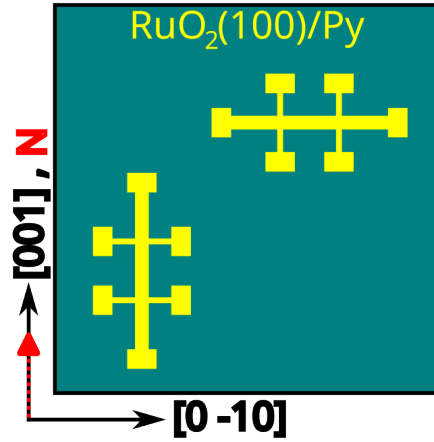


Figure 5.8 Patterned orthogonal Hall bars with current along RuO₂[001] ($J_C \parallel c$) and along RuO₂[010] ($J_C \perp c$).

The similar ADMR upon rotating J_C relative to c argues against a dominant bulk mechanism, such as antiferromagnetic-SMR or altermagnetic spin-split MR, tied to a particular transport axis. A small asymmetry around $\beta = \pm 90^\circ$ is present and traceable to finite transverse resistivity pickup (e.g., AHE) from minor lead misalignment; the corresponding tilt of longitudinal equipotentials is estimated at $\sim 3^\circ$ and does not affect the extracted sign.

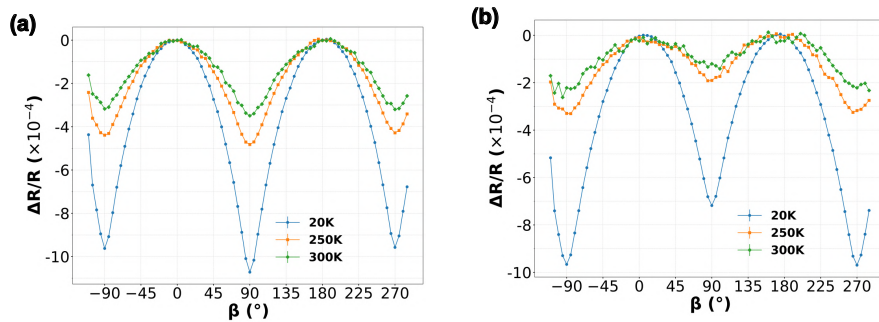


Figure 5.9 Longitudinal resistance variations in the β -plane at different temperatures for (a) $J_C \perp c$ and (b) $J_C \parallel c$ configurations for RuO₂[5 nm]/Py under a 0.95 T magnetic field.

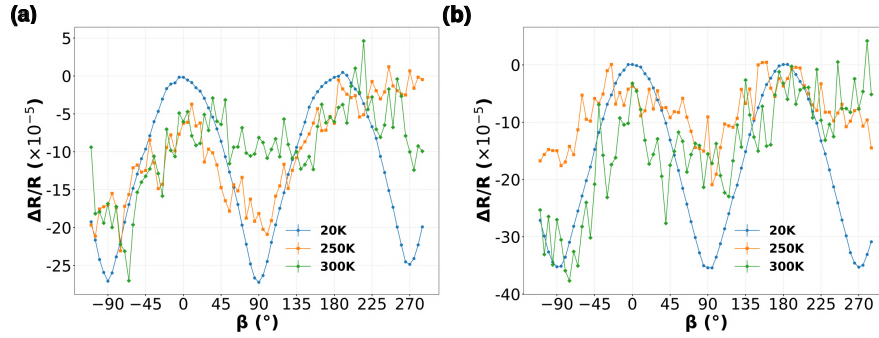


Figure 5.10 Longitudinal resistance variations in the β -plane at different temperatures for (a) $J_C \perp c$ and (b) $J_C \parallel c$ configurations for $\text{RuO}_2[8 \text{ nm}]/\text{Py}$ under a 0.95 T magnetic field.

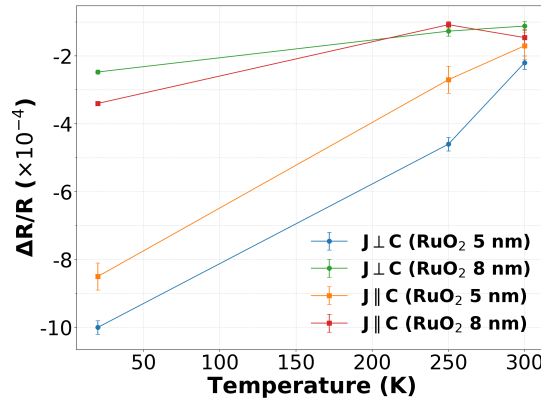


Figure 5.11 Temperature dependence of the peak-to-peak magnetoresistance amplitude in the β -plane.

Temperature Dependence (20 K to 300 K)

Figure 5.11 summarizes peak-to-peak ADMR amplitude versus T from Figs. 5.9 and 5.10 for both Hall bars and different thicknesses for β -plane sweeps. The negative β -plane state persists in both orientations with similar amplitude, shows lower ADMR amplitude at higher RuO_2 thickness, and its temperature evolution is comparable between the two bar directions. In α and γ , the amplitudes at 300 K are roughly half their values at 20 K, tracking the temperature dependence expected for ferromagnetic AMR. By contrast, the β -plane signal is much more sensitive to temperature, dropping to about one-fifth of its low- T magnitude compared to 300 K magnitude. This strong suppression, together with the preserved sign, suggests a different origin than ferromagnetic AMR.

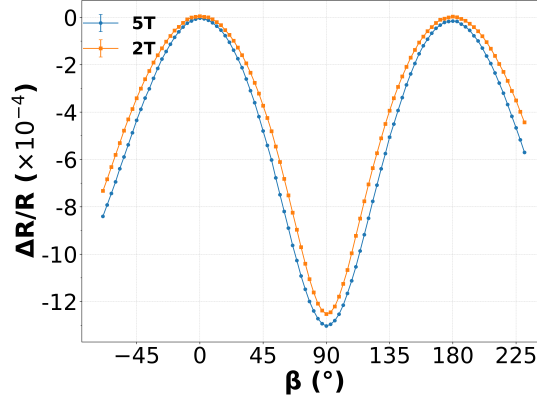


Figure 5.12 β -plane ADMR measured at 2 T and 5 T for RuO₂[5 nm]/Py for $J_C \parallel c$ at 20 K.

Magnetic-Field Dependence (0.95 T to 5 T)

We evaluated the field dependence of the β -plane ADMR by repeating full angular sweeps at $\mu_0 H = 2$ T and 5 T (reference state at 0.95 T) and plotted the results in Fig. 5.12. Within experimental uncertainty, the amplitude and phase of the negative state are unchanged, indicating that the effect is not driven by field-induced reconfiguration of bulk order (e.g., domain realignment) in RuO₂ or Py over this range.

Summary of Experimental Findings

Across geometries, temperatures, and fields, the data converge on a coherent picture: RuO₂/Py exhibits a reproducible negative ADMR state for β -plane rotations that (i) is absent in bare Py controls, (ii) is largely independent of crystallographic current direction ($J_C \parallel c$ vs. $J_C \perp c$), and (iii) is weakly dependent on magnetic field between 0.95 T and 5 T.

5.3 Discussion

Our measurements reveal a clear dependence of the resistance on the magnetization orientation in both the α - and γ -planes (Fig. 5.7(a)). Such behavior is consistent with AMR in FMs, which arises from spin–orbit coupling and the resulting band splitting [28]. In AMR, electron scattering is usually enhanced when the magnetization is parallel to the charge current compared to when it is perpendicular. This identical pattern is also observed in the bare Py thin film (Fig. 5.7(b)), aligning precisely with

the theoretical predictions and establishing a baseline for the comparison of more complex heterostructures.

A significant difference is observed in the β -plane measurements of RuO₂/Py heterostructures compared to bare Py. ADMR for magnetic-field direction in the β -plane has been extensively documented across various heterostructures, including FM/HM [10, 62, 98–103], FM/light metal (LM) [104], (AFM)/HM [105], and bare FM systems [101, 102, 106]. The underlying mechanisms attributed to this ADMR include SMR [10, 62, 103], REMR [98], orbital Rashba–Edelstein magnetoresistance (OREMR) [99], IGSC [100, 104], AHMR [102], and the geometric size effect (GSE) [100, 104], depending on the system. In our measurements, the bare Py film exhibits negligible β -plane ADMR, whereas the RuO₂/Py bilayer shows a pronounced angular dependence in this plane (Fig. 5.7(a) and (b)). Typically, in FM/HM, FM/LM, or bare FM systems, a high-resistance (“positive”) ADMR state is expected for out-of-plane magnetization when mechanisms such as SMR, REMR, OREMR, or AHMR are dominant. However, our results indicate a lower-resistance (“negative”) ADMR state when the magnetization is oriented out of plane compared to the in-plane configuration. This discrepancy suggests that conventional SMR, REMR, OREMR, and AHMR cannot account for the observed ADMR. Negative ADMR for magnetic-field direction in the β -plane is more commonly associated with AFM/HM systems due to SMR, FM/HM and FM/LM interfaces due to IGSC and GSE, and in bare FM films primarily due to GSE. To determine the most plausible explanation for our findings, we will further examine the roles of altermagnetic SMR, antiferromagnetic SMR, GSE, and IGSC in the RuO₂/Py system.

5.3.1 Excluding the conventional spin Hall magnetoresistance and certain alternative mechanisms

Recent theoretical calculations suggest that RuO₂ can act as an altermagnet, providing a crystal-axis-dependent spin-polarized current when the transport direction is along [010] but not along [001] [22]. We expect the magnetic easy axis of the RuO₂ to be along the c -axis, which should not be significantly influenced by the comparably weak external magnetic field of 0.95 T used in our experiments [8]. If altermagnetism were the main mechanism for the ADMR signal, we would expect a pronounced crystallographic dependence (i.e., “positive” ADMR along particular axes). However, the observed ADMR in our β -plane measurements is largely independent of the crystallographic orientation (compare Fig. 5.9(a) and (b)) and shows a consistent

“negative” sign. This does not necessarily rule out altermagnetism but indicates that we do not observe the effects of spin-polarized transport induced by spin-polarized current from RuO₂.

Negative ADMR in AFM/HM systems typically results from the interaction of a spin current driven by the SHE with the Néel order in the AFM. However, we do not observe any dependence on the *c*-axis (Néel order) by changing the Hall bar parallel or perpendicular to the *c*-axis [107, 108] or a significant correlation with magnetic-field magnitude that would suggest domain realignment [109], both of which would be anticipated in an AFM scenario. Nonetheless, a pronounced realignment of domains is not expected for RuO₂ at a magnetic field of 5 T [110]. Furthermore, the temperature dependence of the magnetoresistance remains similar for two orthogonal crystallographic orientations, which further rules out the AFM-based explanation as we expect it to behave differently along Néel order and orthogonal to it as we change temperature [105, 109].

The geometric size of a thin film can result in different scattering conditions based on whether the magnetization is in plane or out of plane, sometimes leading to negative ADMR for magnetic-field direction in the β -plane [111]. The relatively large magnitude of the negative ADMR observed in the RuO₂/Py sample, compared to a single Py film produced under similar conditions, exceeds the predictions of simple GSE models [100, 104]. The texture and thickness of a thin film have been demonstrated to influence the GSE. For instance, a textured film has been shown to enhance ADMR, while a thicker film may diminish it in the β -plane [112]. In our study, both Py thin films were grown on substrates with very similar lattice constants, suggesting they possess comparable textures, which should result in similar GSE effects. Furthermore, the effective thickness of the conducting channel is increased for the bilayer compared to bare Py, resulting in a higher negative ADMR. This outcome contradicts the anticipated decrease in negative ADMR with an increase in thickness due to GSE. Nevertheless, we observe significantly different ADMR behaviors than GSE in the bare Py and heterostructure, surpassing simple GSE explanations. Previous research has attempted to isolate GSE contributions by varying the thickness of the Py layer, demonstrating that GSE alone cannot account for the observed strong negative ADMR [100, 104].

5.3.2 Interfacial Origin of Negative ADMR: IGSC + iSHE Mechanism

Based on our prior discussion, it is evident that altermagnetic SMR, antiferromagnetic SMR, and GSE do not provide a satisfactory explanation for the observed ADMR. Therefore, it is necessary to consider an alternative mechanism that may account for this signal. The observations suggest IGSCs [9] at the RuO₂/Py interface could be the dominant source of the negative ADMR in the β -plane [100, 104]. The theoretical framework and understanding of the negative ADMR in this work were developed by our theory collaborators and reported in detail in the main article [97]. I gratefully acknowledge the theoretical development by EWIESE MOHAMMAD ABABNEH and PROF. VIVEK AMIN. Any inaccuracies in the condensed summary below are mine. IGSCs are spin currents generated from spin–orbit scattering within one mean free path of the interface. For example, an in-plane charge current J_C induces a perpendicularly flowing spin current Q_{zy} with flow direction z (i.e., out of plane) and spin direction y (parallel to the spin direction of the spin Hall current) due to spin–orbit scattering at the interface [113, 9, 114–116]. The momentum-dependent and spin-dependent scattering processes that cause IGSCs are inherently captured in multilayer Boltzmann simulations but can also be captured by drift–diffusion models by inserting a spin-current source term at the interface [113, 114].

The mechanism by which the interface causes a longitudinal magnetoresistance: At the RuO₂/Py interface, the in-plane charge current generates a spin current Q_{zy} . This particular orientation of IGSC is created via the spin–orbit filtering process [9]. This spin current is converted to a longitudinal charge current via the iSHE. Even if the source term for the IGSC in the drift–diffusion equations is magnetization independent, the boundary conditions at the interface, described by magnetoelectronic circuit theory, are magnetization dependent. As a result, the total spin current that forms in the bulk nonmagnetic layer depends on whether $M \parallel y$ or $M \perp y$. Thus, the contribution to the longitudinal charge current from the iSHE will be different for $M \parallel y$ or $M \perp y$, resulting in a contribution to the ADMR. The drift–diffusion calculations show that the total spin current in the nonmagnetic layer (and thus the total backflow of longitudinal charge current) is greater when $M \parallel y$ versus $M \parallel z$ for a large parameter space, indicating that “negative” ADMR can result from the combination of IGSCs and the iSHE.

The previous work by Kang *et al.* [100] studies the longitudinal magnetoresistance versus magnetic-field direction in the β -plane using a spin drift–diffusion model [11]

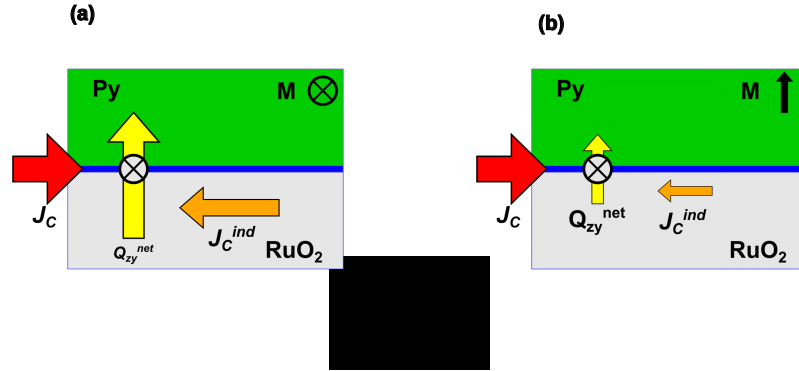


Figure 5.13 Schematic representation of IGSC-induced negative ADMR. When a charge current J_C (red arrow) is injected, a spin current Q_{zy} (yellow arrow) is generated perpendicular to the interface. The magnitude of the spin current Q_{zy} is maximized and minimized when the magnetization M (black arrow) is (a) parallel and (b) perpendicular to the spin index on the y -axis, respectively. Consequently, the inverse process of SHE converts the interfacial spin current back into an induced charge current J_C^{ind} (orange arrow), resulting in a resistance change.

that includes IGSCs [9, 113–115] and utilizes symmetry arguments based on Onsager reciprocity. In their model, the contribution from IGSCs and the iSHE effect is canceled by a reciprocal contribution. In the reciprocal contribution, the SHE first generates an out-of-plane flowing spin current and the interface converts it into an in-plane charge current via spin–orbit scattering. One could refer to this as interfacial spin-to-charge conversion or an *inverse* IGSC. There is a caution that the Onsager reciprocity of IGSCs has not been confirmed and is unlikely to hold due to the breaking of time-reversal symmetry at the interface, and these contributions do not cancel in realistic systems. The work introduces an entirely interfacial magnetoresistance due to both IGSCs and their inverse counterparts. Such contributions are allowed by symmetry but should be studied entirely in the context of the Boltzmann formalism, since the conversion processes take place entirely within one mean free path from the interface.

$$\frac{\Delta Q_{zy}}{Q_{zy}} = \frac{Q_{zy}(M \parallel z) - Q_{zy}(M \parallel y)}{Q_{zy}(M \parallel z) + Q_{zy}(M \parallel y)}, \quad (5.1)$$

Thus, in this work, the focus is on the interplay between IGSCs and the iSHE, as it is the simplest way to demonstrate how negative ADMR can result from interfacial

scattering. Figure 5.13(a) and (b) illustrate how the combination of IGSCs and the iSHE leads to a greater longitudinal resistance for $M \parallel y$ compared to $M \parallel z$. Here, J_C represents the applied charge current, while J_C^{ind} corresponds to the induced charge current generated via the spin-to-charge conversion processes. The net spin current Q_{zy}^{net} at the RuO_2/FM interface follows the relation $Q_{zy}^{\text{net}}(M = z) < Q_{zy}^{\text{net}}(M = y)$, since the absorption of spin current by the FM layer Q_{zy}^{abs} is larger when M is aligned along the z -axis compared to the y -axis. The ADMR sign is extracted using Eq. (5.1). Unlike contributions from the bulk SHE and AHE, for which spin currents are generated away from the interface and are partially reflected at the RuO_2/Py boundary, the IGSCs originate directly at the interface and are not subject to such reflections [100, 9]. This mechanism can account for the observed lower resistance when the magnetization is out of plane, resulting in the negative ADMR state. The temperature dependence of this effect further supports this interpretation, as elevated temperatures reduce coherent spin-dependent scattering and decrease the mean free path, thereby diminishing the contribution from IGSC and weakening the ADMR (Fig. 5.11). Additionally, the independence of ADMR from the applied magnetic-field strength suggests that the IGSCs are primarily governed by the magnetization direction rather than the magnitude of the external magnetic field, which explains the lack of significant influence of magnetic-field strength in our data (Fig. 5.12).

5.4 Conclusion

This chapter established a predominantly interfacial origin of the observed negative ADMR in RuO_2/Py bilayers, consistent with interface-generated spin current at the interface and supplemented by a bulk inverse spin Hall effect. The sign, phase, and geometry dependence across in-plane and out-of-plane rotation planes point to a boundary-governed mechanism rather than a conventional bulk SMR response. Systematic series in thickness, current orientation ($J_C \parallel c$ vs. $J_C \perp c$), temperature, and field strength corroborate this assignment, while controls and artifact checks bound thermoelectric and geometric size effects. A minimal drift-diffusion framework with an interfacial conversion term captures the qualitative trends without fine tuning, providing a compact physical picture for the negative ADMR.

The main limitations arise from uncertainties in interfacial parameters (such as effective spin-mixing conductance and spin-memory loss) and potential device-to-device variations in interface transparency and roughness. While the collective symmetry evi-

dence disfavors alternative explanations (conventional SMR, REMR/OREMR, AHMR, GSE), small admixtures cannot be fully excluded at the few-percent level. The immediate next steps include targeted interface spectroscopy, deliberate termination, and control of stoichiometry. These efforts are expected to tighten parameter bounds and further distinguish interfacial from bulk channels within a unified modeling framework.

The IGSC-dominated interpretation and the derived criterion for achieving negative ADMR rather than altermagnetic SMR from the heterostructure suggest investigating the magnetism of RuO₂. The following chapter uses low-energy muon spectroscopy to investigate the debated magnetism of RuO₂. It lays out practical levers, such as different substrates, growth techniques, and growth parameters, to determine what establishes magnetism in RuO₂. In doing so, it explains the interesting localization of magnetic order, regardless of the growth method or substrate.

Chapter 6

Surface Magnetism in RuO₂ Thin Films: A Muon Spectroscopy Study

6.1 Introduction

This chapter presents an experimental investigation of the magnetic ground state of rutile RuO₂ in thin-film form using depth-resolved low-energy muon spin rotation/relaxation (LE- μ SR). Motivated by the broader context of altermagnetism and the ongoing debate regarding the magnetic ground state of RuO₂ [17, 38, 40, 49]. The intrinsic magnetic ground state of RuO₂ remains highly debated, especially when comparing findings between thin films, which show evidence of altermagnetic features or unusual magnetic and transport properties [7, 8, 13–15, 35, 36, 38, 40, 43, 45–47, 96, 97, 117–130], and bulk single crystals, which are widely suggested to be non-magnetic [49, 50, 53, 54, 131–137]. This inconsistency underscores the critical need for experimental techniques that can probe magnetic properties with high spatial resolution in reduced-dimensional systems. This chapter details the experimental results of this thesis, utilizing depth-resolved LE- μ SR to definitively clarify the nature and, crucially, the spatial extent of the magnetic order in RuO₂ thin films grown under different conditions [138].

The motivation behind using LE- μ SR is its unique capability to perform depth-resolved measurements, enabling the differentiation between magnetic phenomena originating from the film's interior and those confined to the surface or interface regions [77, 138, 139]. The measurements focus on epitaxial RuO₂ films grown on TiO₂(110) and Al₂O₃(1 $\bar{1}$ 02) substrates using two distinct growth routes (pulsed-laser deposition and reactive sputtering), enabling a comparison across substrate, orientation, and deposition method. As detailed in Chapter 2 (Methodology), film synthesis, structural verification (XRD, RHEED), and LE- μ SR data acquisition and analysis protocols follow standardized, reproducible procedures [81, 83]. Within this framework, I analyze how the muon asymmetry and damping vary with energy and temperature to identify magnetic order confined to the film surface and to estimate the corresponding magnetic volume fraction. These results provide an internally consistent picture across materials systems and measurement conditions.

Within the overall thesis, this chapter operationalizes the scientific objectives introduced in Chapter 1 (Introduction): (i) to establish whether magnetic order in RuO₂ thin films is intrinsic [6, 33, 39] or bulk-like non-magnetic [40, 49] or instead localized to reduced-dimensional surface regions [13, 15, 17, 96], (ii) to quantify its spatial extent and temperature evolution using a local probe with depth sensitivity, and (iii) to test robustness against growth pathway and crystallographic orientation.

The core finding presented in this chapter is the direct evidence for a surface-confined, inhomogeneous static magnetic order in the RuO_2 thin films [140]. Analysis of the transverse magnetic field (TF) μSR data, collected across a temperature range from 4 K to 290 K, reveals that the magnetic signals predominantly originate from the near-surface region, specifically the top $\lesssim 10$ nm of the film [49, 140]. Furthermore, the affected magnetic volume fraction is small, quantified at approximately 8.5% of the near-surface volume. These depth-resolved results successfully reconcile discrepancies in existing literature by emphasizing the significant impact of reduced-dimensional contributions, including surface defects, strain, and dimensionality effects, on the emergence of weak magnetic signals in RuO_2 thin films. The following sections will present the specific energy- and temperature-dependent μSR parameters that support this conclusion, leading to a thorough discussion of the implications for the magnetic ground state of RuO_2 .

6.2 Experimental Results

6.2.1 Sample preparation and Structural Characterization

This study investigated the magnetic properties of ruthenium dioxide (RuO_2) thin films grown via two distinct deposition methods on two different substrates. The first set of samples consisted of 33 nm thick $\text{RuO}_2(110)$ films grown on $\text{TiO}_2(110)$ substrates using pulsed laser deposition (PLD) under oxygen pressures of 0.025 mbar (± 0.005) and substrate temperatures of 375 °C (± 25). The second set comprised 30 nm thick $\text{RuO}_2(101)$ films deposited on $\text{Al}_2\text{O}_3(\bar{1}102)$ substrates using reactive DC magnetron sputtering under a pressure of 0.004 mbar, with a mixture of $\text{O}_2/(\text{Ar} + \text{O}_2) = 0.167$ and substrate temperatures of 700 °C. I gratefully acknowledge DR. JAE-HYUN HA and SIYEON AN members from DR. DONG-SOO HAN's group for preparing the $\text{RuO}_2(101)/\text{Al}_2\text{O}_3(\bar{1}102)$ samples. Figure 6.1 shows the XRD patterns measured with the scattering vector normal to the (110)-oriented rutile TiO_2 and the $(\bar{1}102)$ -oriented Al_2O_3 substrate. Both $\text{RuO}_2(110)/\text{TiO}_2(110)$ and $\text{RuO}_2(101)/\text{Al}_2\text{O}_3(\bar{1}102)$ thin film samples show an XRD pattern corresponding to a single crystallographic orientation of the deposited layer. The $\text{RuO}_2(110)/\text{TiO}_2(110)$ samples specifically exhibited Laue oscillations in the XRD pattern, which is characteristic of a single-crystalline film with sharp interfaces [141]. Further structural verification for the PLD-grown films was provided by in-situ RHEED. Figure 6.2 presents the RHEED pattern for the $\text{TiO}_2(110)/\text{RuO}_2(110)$ sample, obtained with the electron beam aligned along

TiO₂[001], where the observation of modulated streaks corroborated single-crystalline growth and indicated a low surface roughness of only a few atomic layers. Each sample has a surface area of about 1 cm × 1 cm. Both sets of four samples on the same substrate were mounted on a nickel-coated aluminum plate to maximize the incident muon beam cross-section and improve counting statistics, resulting in a total area of 2 cm × 2 cm.

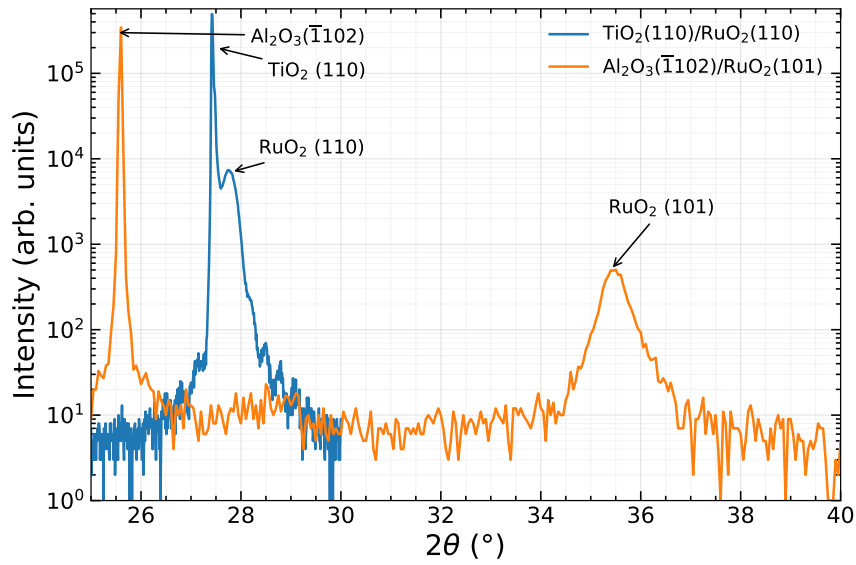


Figure 6.1 XRD patterns measured with the scattering vector oriented normal to the (110)-oriented rutile TiO₂ and the ($\bar{1}102$)-oriented Al₂O₃ substrate.

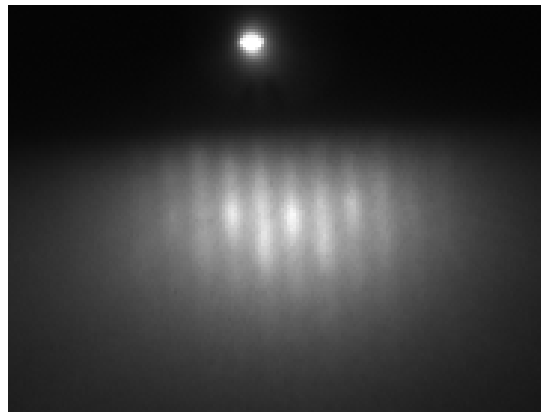


Figure 6.2 RHEED pattern of the TiO₂(110)/RuO₂(110) heterostructure, recorded with the electron beam aligned along the TiO₂ [001] crystallographic direction.

Low-Energy Muon Probe Principle and Depth Profiling

LE- μ SR experiments were conducted at the Swiss Muon Source (S μ S) at the Paul Scherrer Institute (PSI) using the low-energy muon (LEM) spectrometer [142]. The experimental setup and measurement procedures are described in detail in the Methods chapter. The central capability utilized in this research is the depth-resolved sensitivity supported by varying the implantation energy of the positive muons (μ^+) [77, 138, 139, 143]. Muons, initially slowed to tens of eV, were electrostatically accelerated to energies ranging from 1 keV and 12 keV, allowing for the magnetic environment to be selectively probed from the near-surface region into the film bulk and the substrate. The time spectra of the decaying muon spins were fitted using a damped cosine function with a Gaussian envelope: $A(t) = A_0 \cos(\gamma_\mu B t + \phi) e^{-\frac{1}{2}(\sigma t)^2}$. This fit gives key parameters: the initial asymmetry A_0 , the local magnetic field B , and the muon spin damping rate σ ; analysis was carried out with `musrfit` [83, 81]. I sincerely acknowledge DR. ZAHER SALMAN, beamline scientist at PSI, for helping with the measurements at the beamline and with the data analysis of spectroscopy. Measurements were primarily performed in a TF of 10 mT, with supplementary measurements conducted at 140 mT, applied perpendicular to the initial muon spin polarization and the sample surface. The temperature was varied from 4 K to 290 K. For the RuO₂(110)/TiO₂(110) films, the muon spin was oriented in-plane and normal to the c -axis. For the RuO₂(101)/Al₂O₃($\bar{1}102$) films, the muon spin was oriented in-plane and 55.3° to the c -axis.

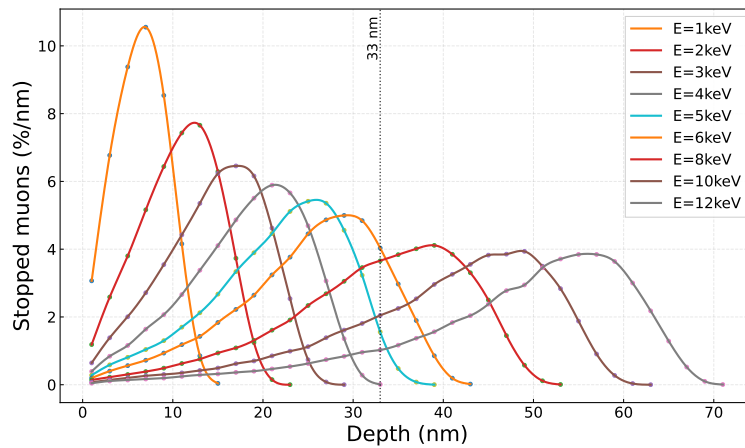


Figure 6.3 Simulated stopping profiles of muons in the RuO₂(110)/TiO₂(110) sample for different implantation energies, and the dashed line represents the depth of the thin film beyond which muons stop into the substrate.

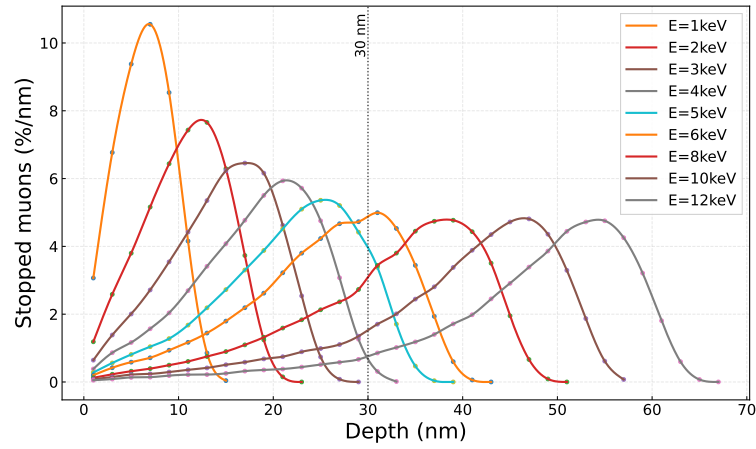


Figure 6.4 Simulated stopping profiles of muons in the RuO₂(101)/Al₂O₃($\bar{1}102$) sample for different implantation energies, and the dashed line represents the depth of the thin film beyond which muons stop into the substrate.

To map depth, Monte Carlo TRIM.SP simulations were used to compute energy-dependent stopping distributions for the exact film/substrate stacks studied in this work [80, 81]. For ~ 1 keV, the majority of muons stop within the top ~ 10 nm of the RuO₂ films, while for $E \sim 8$ keV to 12 keV a large fraction reaches the substrate. The simulated profiles for RuO₂(110)[33 nm]/TiO₂(110) and RuO₂(101)[30 nm]/Al₂O₃($\bar{1}102$) samples are shown in Figure 6.3 and Figure 6.4, respectively. These simulated profiles are incorporated when interpreting the energy and temperature dependences of σ and A_0 below.

6.2.2 Energy Dependence of Magnetic Response at Low Temperature (10 mT TF)

TF LE- μ SR measurements were conducted across the full range of implantation energies (1 keV to 12 keV) to map the depth-dependent magnetic behavior in both film systems at low temperatures, comparing the response to that at 290 K. For the RuO₂/TiO₂ films, low implantation energies, particularly 1 keV (where muons stop predominantly in the top ~ 10 nm of the film), showed a pronounced and gradual increase in the damping rate (σ) and a significant decrease in the initial asymmetry (A_0) at 25 K compared to 290 K as shown in Figures 6.5(a). Conversely, higher energies (12 keV), where muons stop mainly in the TiO₂ substrate, exhibited much weaker temperature dependence, although an anomaly was noted at 4 K due to the formation of the paramagnetic muonium complex in rutile TiO₂ [144, 145]. For the

$\text{RuO}_2/\text{Al}_2\text{O}_3$ films, low implantation energies (1 keV to 2 keV) also showed increases in σ and decreases in A_0 at 4 K relative to 290 K as shown in Figures 6.5(b). However, at higher implantation energies, where muons stopped primarily in the Al_2O_3 substrate, a more substantial increase in σ and a stronger decrease in A_0 were observed; this is attributed to the muon experiencing the nuclear dipolar field of Al moments and muonium formation within the Al_2O_3 substrate [146–149]. Overall, the data consistently demonstrated that the most significant changes in σ and A_0 occur at low implantation energies, hinting that the magnetic response originates from the near-surface regions of the RuO_2 thin films.

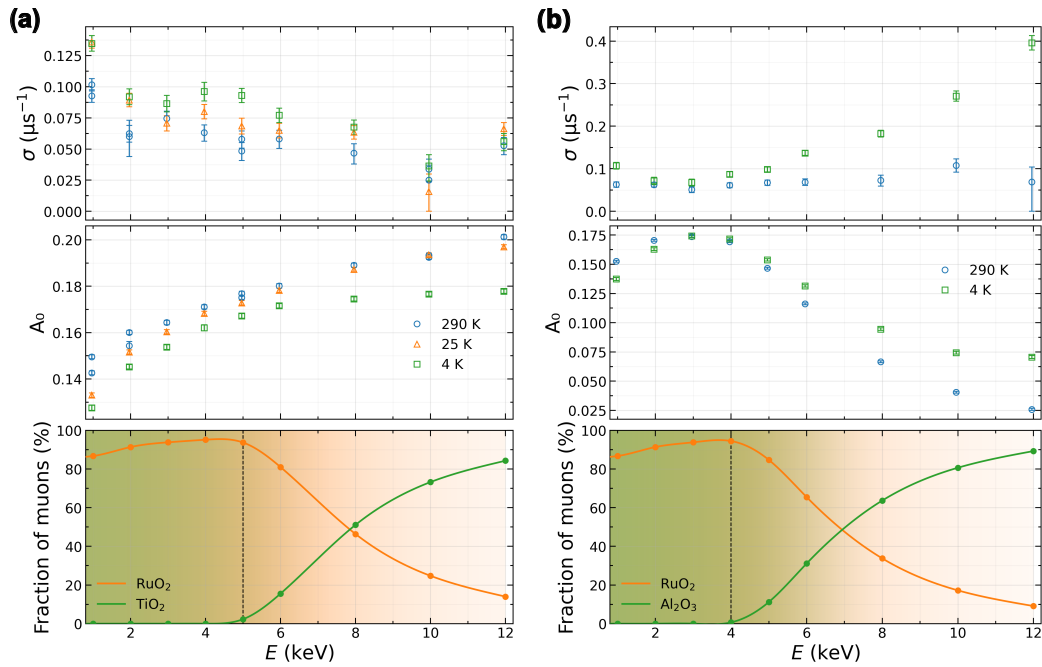


Figure 6.5 Dependence of muon spin damping rate σ , initial asymmetry A_0 , and fraction of muons stopping in the thin film and substrate on implantation energy at different temperatures for (a) $\text{RuO}_2(110)/\text{TiO}_2(110)$ films (b) $\text{RuO}_2(101)/\text{Al}_2\text{O}_3(1102)$ films at applied magnetic field of 10 mT and (c) The green and orange shaded areas represent the fraction of muons in the RuO_2 and substrate layers, respectively.

6.2.3 Detailed Temperature Evolution of Surface Magnetic Features (10 mT TF)

Detailed temperature-dependent measurements (4 K to 290 K) further confirmed the surface localization of the magnetic signal. Figures 6.6(a) and 6.6(b) illustrate the

evolution of σ and A_0^N (normalized A_0 with respect to weighted mean of $A_0(290\text{ K})$ values) with different muon implantation energies for RuO₂ films grown on TiO₂ and Al₂O₃ substrates, respectively. For both RuO₂ systems, a gradual increase in σ and a corresponding decrease in the normalized asymmetry $A_0^N \equiv A_0(T)/A_0(290\text{ K})$ were observed as the temperature was lowered. This variation was most substantial and systematic at the 1 keV implantation energy. In contrast, at higher implantation energies, which probe deeper into the film and substrate, only a weak variation of σ and A_0^N with temperature was detected. This dependence strongly suggests that the underlying magnetic response is confined to the near-surface region, of approximately the top 10 nm thick, of the RuO₂ thin films. It is noted that for the RuO₂/TiO₂ films, the 4 K data points were excluded from the linear fits of the temperature dependence due to the dominant paramagnetic background signal arising from the TiO₂ substrate below 10 K [144, 145]. The variations in the local magnetic field B were found to be within the systematic error limits of the apparatus, indicating no observable temperature or implantation energy dependence in B .

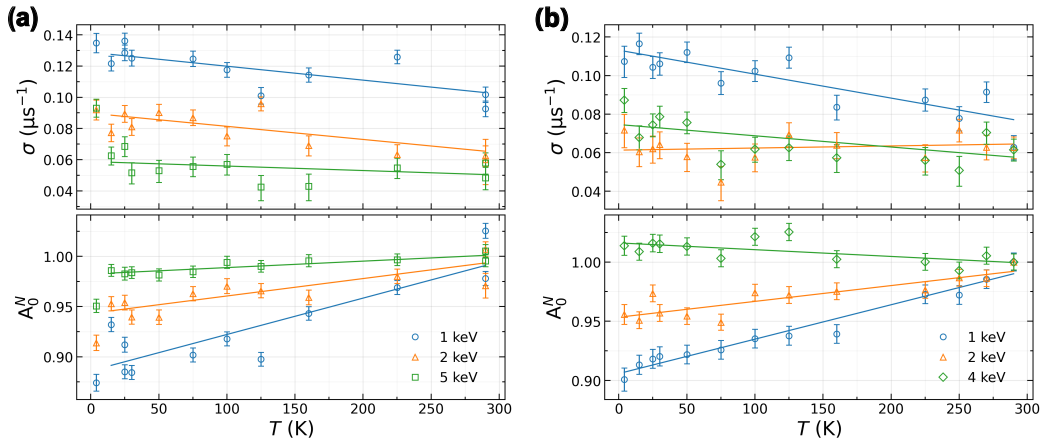


Figure 6.6 Temperature dependence of normalized initial asymmetry A_0^N and muon spin damping rate σ films at different implantation energies for (a) RuO₂(110)/TiO₂(110) films and (b) RuO₂(101)/Al₂O₃($\bar{1}102$) films at applied magnetic field of 10 mT.

Quantification of Surface Magnetic Volume Fraction

A quantitative analysis of the initial asymmetry, A_0 , measured at an implantation energy of 1 keV, was performed to estimate the extent of the magnetic order. The relative change in A_0 between 290 K and 4 K, defined as $(A_0(290\text{ K}) - A_0(4\text{ K}))/A_0(290\text{ K})$, indicates that $10.5\% \pm 3.6\%$ for TiO₂/RuO₂ and $8.4\% \pm 0.9\%$ for Al₂O₃/RuO₂ (95%

confidence interval) of the near-surface volume exhibits inhomogeneous magnetic order [150]. The combination of the small magnetic volume fraction, absence of a resolvable systematic shift in the mean field B , the gradual temperature dependence of σ and A_0^N , and the confinement to the surface layer suggests that the magnetic response is characterized by inhomogeneous static magnetic order confined to the surface of the thin film. Crucially, the LE- μ SR data provided no evidence for long-range magnetic order residing inside the thin film.

6.2.4 Verification of Trends at High Transverse Field (140 mT TF)

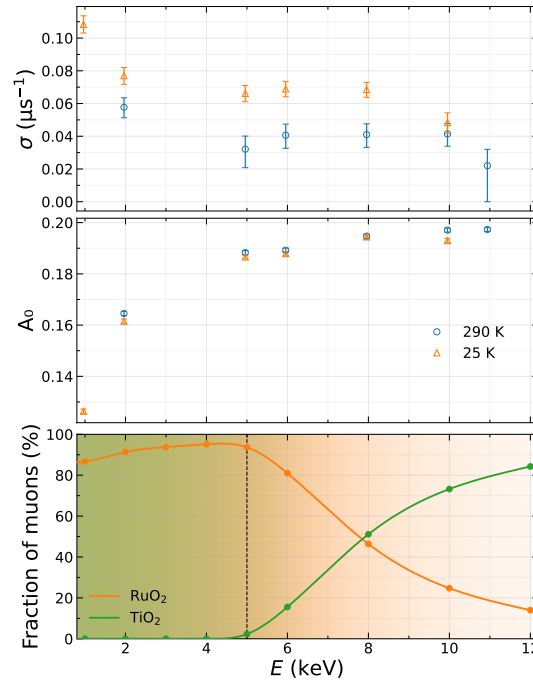


Figure 6.7 Dependence of initial asymmetry A_0 , muon spin relaxation rate σ , and fraction of muons in thin film and substrate on implantation energy at 25 K and 290 K for $\text{RuO}_2(110)/\text{TiO}_2(110)$ films with applied magnetic field of 140 mT.

To confirm the robustness of the observed surface magnetic features, additional LE- μ SR measurements were conducted at a higher transverse magnetic field of 140 mT for both RuO_2 samples. Figure 6.7 and Figure 6.8 shows the extracted parameters σ and A_0 temperature dependence TF muon spectroscopy with applied field of 140 mT for $\text{TiO}_2(110)/\text{RuO}_2(110)$ and $\text{Al}_2\text{O}_3(\bar{1}102)/\text{RuO}_2(101)$ thin film samples, respectively. These high-field results yielded trends that are qualitatively analogous to the primary 10 mT measurements. The consistency between the 10 mT and 140 mT data validates

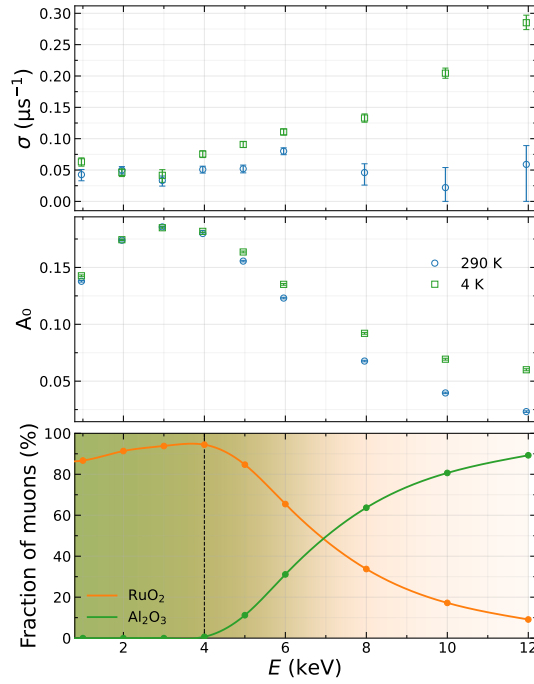


Figure 6.8 Dependence of initial asymmetry A_0 , muon spin relaxation rate σ , and fraction of muons in thin film and substrate on implantation energy at 4 K and 290 K for RuO₂(101)/Al₂O₃($\bar{1}102$) films with applied magnetic field of 140 mT.

that the observed surface-localized effects are not artifacts of the low magnetic field conditions.

6.3 Discussion

6.3.1 Signature for Surface-Confined Magnetism in RuO₂ Thin Films

The depth-resolved LE- μ SR data establish a consistent phenomenology: the largest low-temperature increase of the muon spin damping rate σ and reduction of the initial asymmetry A_0 occur at the lowest implantation energies ($E \sim 1$ keV to 2 keV), where the stopping distribution is confined to the top ~ 10 nm of the films. With increasing E , both effects weaken and eventually track substrate-dominated responses, demonstrating that the static field inhomogeneity is a near-surface property of RuO₂ rather than a bulk-like order extending throughout the ~ 30 nm to 33 nm films. A magnetic volume fraction derived from the low- T change of A_0 at 1 keV indicates that

the weighted mean of approximately $\sim 8.5\%$ of the near-surface volume exhibits such static inhomogeneous order. The qualitative trends persist at both 10 mT and 140 mT transverse fields, confirming that the inference is not an artifact. Taken together, these observations provide direct local-probe evidence for surface-confined static magnetism in epitaxial RuO₂ thin films. Importantly, the mean precession field B shows no systematic shift across temperature and energy within experimental uncertainties, arguing against a net FM or AFM component and favoring inhomogeneous static magnetic order [49, 50]. This surface-localized magnetism represents a critical finding that may explain why thin films of RuO₂ demonstrate anomalous or altermagnetic-like features [8, 14, 15, 38, 47, 97, 117, 118, 130], contrasting sharply with bulk single crystals, which are widely reported to be non-magnetic [50, 53, 54, 131–137]. A central question arising from this localization is whether the observed behavior represents intrinsic surface altermagnetism or is a manifestation of extrinsic effects such as strain, interface defects, or localized states.

6.3.2 Robustness and Dependence on Growth Conditions

The surface-localized response is robust across two distinct heteroepitaxial systems and growth routes: PLD-grown RuO₂(110) on TiO₂(110) and reactively sputtered RuO₂(101) on Al₂O₃(1 $\bar{1}$ 02). Despite differences in orientation, lattice mismatch, and oxygen chemical potential during growth, both sample families show the same depth hierarchy: strong near-surface changes in σ/A_0 at low T and weak-to-negligible evolution for deeper stopping profiles. This consistency suggests a common underlying mechanism is responsible for the surface order, which is not dependent on the specific substrate or deposition technique. This mechanism is likely related to surface defects or dimensionality effects [13, 14, 17, 40, 97, 130]. The lack of bulk long-range order in both film types underscores the fact that structural perfection deep within the film resembles the non-magnetic bulk state.

That said, subtle differences between substrates (e.g., the onset of muonium-related substrate signals in TiO₂ at the lowest temperatures and the nuclear-dipolar background of Al₂O₃) highlight the importance of carefully considering background channels. Within those constraints, cross-system agreement strengthens the case that the magnetism resides in the RuO₂ surface region. A systematic thickness series and controlled surface treatments (oxidation/passivation, annealing) are natural next steps to further test robustness (see Outlook).

Intrinsic vs. Defect-Induced Origin

Two broad classes of mechanisms can account for the observations. An *intrinsic* scenario posits that symmetry breaking at the surface stabilizes (or amplifies) magnetic correlations that are weak or absent in the bulk, potentially connected to the altermagnetic framework in which compensated order coexists with momentum-dependent spin splitting [17, 40]. Surface reconstructions, altered Ru–O bond angles, or modified orbital hybridization could enhance exchange pathways locally, yielding static inhomogeneous fields without producing a net moment detectable as a shift in B [14].

An alternative defect- or interface-driven scenario attributes the magnetism to oxygen-vacancy complexes, cation off-stoichiometry, step edges, or interfacial intermixing. Such defects are known to induce localized moments and glassy freezing in otherwise nonmagnetic oxides, particularly near surfaces and interfaces where formation energies are reduced [151–155]. Several empirical features support this view: (i) the small affected volume fraction; (ii) the inhomogeneous broadening without a coherent oscillatory component; and (iii) the smooth, non-critical temperature evolution of σ . Distinguishing these scenarios requires perturbations that specifically tune surface stoichiometry and bonding (e.g., oxygen chemical potential, capping layers) combined with probes sensitive to moment distribution and symmetry.

Relation to Prior Thin-Film and Bulk Studies (LE- μ SR, ARPES/MCD, TR-MOKE, SP-STM)

The current depth-resolved LE- μ SR results are highly effective in reconciling conflicting observations within the RuO₂ literature by establishing a spatial constraint on the magnetism. This surface confinement can explain why techniques with surface sensitivity (MCD-ARPES, TR-MOKE) observe magnetic fingerprints, while bulk probes may not. Prior μ SR studies: Our findings are consistent with an earlier μ SR study on 10 nm RuO₂ films, which reported stronger damping rates compared to the bulk [49]. However, due to the limited thickness of those samples, the earlier work lacked the depth resolution necessary to definitively attribute the signal to surface magnetism. By utilizing thicker films (30 nm and 33 nm) and the depth-profiling capability of LE- μ SR, we provide direct confirmation that the weak static magnetic fields are indeed confined to the top ~ 10 nm. Surface-sensitive techniques confirming altermagnetism: Several highly surface-sensitive techniques have provided microscopic evidence supporting the altermagnetic classification of RuO₂ films. This includes the direct observation of

strong time-reversal symmetry breaking in the electronic band structure via magnetic circular dichroism (MCD) in ARPES [38]. Furthermore, time-resolved magneto-optic Kerr effect (TR-MOKE) studies have confirmed the d-wave symmetry of the altermagnetic phase in ultrathin films through excitation-angle-dependent transient spin polarization [96]. These results suggest that the electronic structure is inherently altermagnetic. The interpretation of intrinsic surface magnetism is challenged by certain high-resolution surface probes. Notably, spin-polarized scanning tunneling microscopy (SP-STM) measurements conducted on atomically ordered ultrathin (110) surfaces found no evidence of magnetic contrast [156]. This suggests that the inhomogeneous static magnetic order detected by μ SR may either be below the SP-STM detection limit, or, crucially, that it originates from highly localized defects rather than a uniform, intrinsic surface electronic order.

Implications for Reduced Dimensionality and Spintronic Functionality

Surface-confined magnetism in RuO_2 thin films has two immediate implications. First, it points to reduced dimensionality as a key control knob: symmetry lowering, reconstruction, and altered coordination at the surface can stabilize magnetic textures absent in the bulk. Second, even a small magnetic volume fraction, if located at the interface to a spin-current source/detector, can exert a disproportionate influence on spin-dependent transport (spin filtering, interfacial exchange torques, spin-to-charge conversion) [35, 45, 46, 97]. If linked to altermagnetic correlations, such interfacial magnetism could enable momentum-selective spin polarization without macroscopic stray fields, attractive for device integration [22, 23]. Realizing this potential will require deterministic control of surface stoichiometry and reconstruction.

Methodological Limitations and Uncertainties of LE- μ SR Depth Profiling

While LE- μ SR provided unprecedented depth resolution, it is subject to certain methodological limitations. (i) *Averaging Nature*: The TF- μ SR technique inherently measures the average local magnetic field over the entire muon stopping distribution. It averages over different crystallographic orientations and momentum directions, which limits the ability to unambiguously distinguish between genuine intrinsic altermagnetic order (related to momentum space) and signals arising from spatially isolated, defect-related

anomalies. (ii) *Muon Stopping Profiles*: The exact muon implantation depth and distribution profiles are estimated using Monte Carlo TRIM.SP simulations. These simulations depend on assumptions about material density and thickness, introducing inherent overlapping tails of the distributions, which limit depth resolution to several nanometers. (iii) *Substrate Signals*: The substrate contributions had to be carefully isolated, particularly the paramagnetic muonium complex signal in TiO₂ below 10 K [144, 145], and the nuclear dipolar field and muonium formation in Al₂O₃ at higher energies [146–149]. These required specific corrections and exclusions (e.g., 4 K data for TiO₂) to ensure the magnetic response observed was solely attributed to the RuO₂ film.

Outlook

The findings in this chapter establish that any magnetic signature measured in RuO₂ thin films must be critically assessed for its spatial localization. Resolving the microscopic origin of the surface magnetism requires complementary approaches that offer different levels of resolution [157–161]. Building on these results, several targeted studies can decisively separate intrinsic from defect-mediated mechanisms. *Control experiments*: (a) oxygen stoichiometry tuning via post-growth anneals in controlled O₂/vacuum; (b) surface passivation with ultrathin AlO_x/TiO_x caps to suppress reconstructions. *Depth diagnostics*: polarized neutron reflectometry for depth-resolved magnetization [157]; NV-center magnetometry for nanoscale stray-field mapping [158, 159]; XMCD-PEEM for element- and depth-sensitive contrast [160]. *Band-structure tests*: MCD-ARPES with spin resolution to look for momentum-dependent spin splitting expected of altermagnets, ideally on the same samples measured by LE- μ SR [38, 158, 159].

6.4 Conclusion

In this chapter, depth-resolved LE- μ SR established that magnetic order in epitaxial RuO₂ thin films is confined to the near-surface region (top ~ 10 nm), affecting approximately $\sim 8.5\%$ of the local volume at low temperature, with no evidence for bulk-spanning long-range order. The effect is robust across growth routes, substrates, and applied fields (10 mT/140 mT). These findings reconcile thin-film reports of magnetic signatures with bulk null results by identifying a surface-localized origin and frame two viable mechanisms: intrinsic, symmetry-enabled surface correlations

and defect- or interface-driven magnetism. While the combined use of TF- μ SR and depth-dependent implantation provides high sensitivity to local magnetic environments, further investigations using complementary surface-sensitive techniques are required to unambiguously identify the microscopic origin of the observed inhomogeneous static magnetism and distinguish between intrinsic altermagnetism and surface defect-induced anomalies. This work establishes a crucial experimental constraint for future studies aiming to exploit RuO₂ in spintronic applications.

Chapter 7

Spin Hall Angle Dependence of Hall Effects in YIG/Ta and YIG/Pt

7.1 Introduction

This chapter examines the microscopic origins of Hall responses in magnetic insulator (MI)/heavy-metal (HM) systems, with a specific focus on the distinction between single- and double-spin Hall effect (SHE) conversion channels. The SHE and the inverse spin Hall effect (ISHE) enable charge–spin and spin–charge current interconversion, driven by strong spin–orbit coupling (SOC) in HM such as Ta and Pt [2, 25, 162–167]. The spin Hall angle θ_{SH} quantifies this conversion efficiency as the ratio of spin Hall conductivity σ_{SH} to longitudinal charge conductivity σ_{xx} [62, 168, 169]. It depends sensitively on material purity and microstructure (e.g., thin-film phase or crystallinity) [169–171]. When a charge current induces a transverse spin current (SHE), the resulting spin accumulation μ_s at the sample boundaries interacts with external parameters (magnetization or magnetic field), which modulates spin reflection/absorption. The remaining μ_s is converted back to charge current (ISHE), creating a measurable charge resistance change [10, 11, 62, 172].

In HM/MI bilayers, spin Hall magnetoresistance (SMR) originates from the spin-transfer torque at the interface, where SHE-induced spin current drives interfacial spin-flip scattering when perpendicular to the magnetization $\mathbf{M}$, or undergoes reflection when parallel [10, 62, 103, 172]. The effect is characterized by the bulk HM parameters θ_{SH} , spin diffusion length in HM λ_{HM} and the interfacial spin-mixing conductance $G^{\uparrow\downarrow} = G_r + iG_i$ [11, 62, 173–175]. The relative change in charge resistivity is predicted to scale quadratically with the spin Hall angle, $\propto \theta_{\text{SH}}^2$, reflecting the need for two sequential spin-to-charge conversion processes (first SHE, then ISHE). The SMR signal exhibits a characteristic $\cos^2 \theta$ angular dependence, for which θ is the angle between the magnetization $\mathbf{M}$ and the spin-accumulation axis [10, 11, 62, 174]. The Hall signal V_{xy} manifests as an AHE like signal when $\mathbf{M}$ has a component normal to the plane m_z [11, 62, 175, 176]. This component is primarily governed by the imaginary part of the spin-mixing conductance G_i , which represents the interfacial exchange field [11, 62, 173, 176]. The Hall variant of the SMR response V_{xy} , known as spin Hall anomalous Hall effect (SH-AHE), is also supposed to be proportional to θ_{SH}^2 , following the double spin-Hall mechanism [11, 12].

Hanle magnetoresistance (HMR), proposed by Dyakonov, is a magnetoresistance (MR) effect that exhibits nonquadratic magnetic field dependence [172, 177, 178]. Recently reported effects such as orbital HMR show that large MR signals (comparable to HMR/SMR) can originate from orbital angular momentum dynamics, especially in materials such as Mn or Ti with weak SOC, for which the intrinsic SHE is small

[99, 171, 179–181]. It arises when a perpendicular magnetic field induces spin precession (the Hanle effect) that suppresses the spin (or orbital) Hall effect-induced spin accumulation μ_s at film boundaries (surfaces/edges) [172, 177, 178]. This modulation results in a change in longitudinal charge resistivity through ISHE. The angular dependence of HMR mirrors that of SMR (a $\cos^2 \theta$ modulation), for which θ now represents the angle between the applied magnetic field $\mathbf{B}_0$ and the spin-accumulation axis. Both effects stem from the interaction of the SHE-induced μ_s with an external vector ($\mathbf{M}$ for SMR, $\mathbf{B}_0$ for HMR) [62, 177, 182]. Spin Hall Hanle effect (SH-HE), the Hall component of HMR shows nonlinear dependence on magnetic field strength [177, 178]. Theoretically, in thin films, the maximum magnitude of HMR is bound by θ_{SH} and scales as $\propto \theta_{\text{SH}}^2$. The corresponding Hall counterpart SH-HE, likewise scales as $\propto \theta_{\text{SH}}^2$, as predicted [177].

However, an alternative theoretical approach by S. S.-L. Zhang and G. Vignale suggests that the Hall signal can instead be generated through a single bulk SHE combined with interfacial spin-dependent scattering, resulting in a linear dependence on θ_{SH} , in fundamental contrast to the quadratic scaling of the double-SHE mechanism [12]. Determining whether the Hall effect scales linearly (θ_{SH}) or quadratically (θ_{SH}^2) distinguishes between two microscopic origins, interfacial spin-dependent scattering coupled with one bulk SHE step versus two sequential bulk SHE/ISHE processes (double SHE), which is critical for accurate device design and for interpreting spin-orbit phenomena [11, 12, 173, 183].

In this chapter, we test whether the OHE, SH-HE, and SH-AHE in bilayers of a MI with HMs that have opposite spin Hall angles adheres to the established double-SHE mechanism. We measure the Hall-like SMR/HMR response to determine whether it follows the double-SHE mechanism or whether a single-SHE plus interfacial spin-dependent scattering introduces a distinct linear scaling with θ_{SH} . To do so, we employ a systematic measurement scheme and selected bilayer systems that exhibit distinct dominant regimes for OHE, SH-HE, and SH-AHE, while employing HMs with opposite sign of θ_{SH} [174]. Yttrium iron garnet (YIG)/Ta and YIG/Pt are optimal bilayer stacks for studying this effect, offering different parameter windows and opposite spin Hall angles for Ta and Pt [169, 174, 184]. The primary objective is to quantitatively test the predicted linear scaling of the Hall-type response of SMR and HMR components with θ_{SH} . This comparison will assess whether the single-SHE mechanism is the dominant microscopic origin, as opposed to the double-SHE mechanism.

The measurements presented in this chapter show that the SH-HE exhibits consistent sign reversal between YIG/Ta and YIG/Pt systems and tracks θ_{SH} linearly, while the SH-AHE displays a more complex thickness-dependent evolution from 4 nm to 12 nm, suggesting competing spin conversion channels. Together, these results indicate that single-SHE processes predominantly govern spin-to-charge conversion in these canonical MI/HM systems, with implications for optimizing spintronic device architectures. The remainder of this chapter is organized as follows. Section 7.2 describes the fabrication of the YIG/Ta and YIG/Pt heterostructures and summarizes their structural characterization. Subsequent sections detail the magnetotransport measurement geometry and protocols, the separation of Hall-voltage components, and the theoretical modeling used to extract thickness-dependent transport parameters.

7.2 Heterostructure Growth and Structural Characterization

7.2.1 Fabrication of YIG/Ta and YIG/Pt Bilayers

The investigated heterostructures consist of MI/HM bilayers grown on non-magnetic insulating gadolinium gallium garnet (GGG)(001) substrates. Ta and Pt were selected as HMs due to their large and opposite spin Hall angle θ_{SH} [174]. The HM layer thicknesses d_{HM} were varied from 4 nm to 12 nm, and the thickness of MI YIG is 25 nm.

HM films were deposited using DC magnetron sputtering, and YIG was deposited using pulsed laser deposition. Critical parameters included an O_2 pressure in pulsed laser deposition of 2.7×10^{-2} mbar while depositing YIG, a deposition temperature of 480 °C, and an Ar pressure in the sputter chamber of 3.5×10^{-3} mbar while depositing the HM (Ta or Pt) at room temperature. Epitaxy of YIG films was confirmed by *in situ* reflection high-energy electron diffraction (RHEED), and the heterostructures were characterized by x-ray diffraction (XRD) for structural properties. Samples were patterned into Hall-bar structures using photolithography followed by argon ion etching. Typical dimensions include widths w of 20 μm , lengths of 100 μm for the hall bar, and a longitudinal separation l between voltage probes of 40 μm , as shown in Fig. 7.3(a). These dimensions were used to determine the charge resistivity. In the following subsection, I summarize the XRD and RHEED measurements used to verify the structural quality of the YIG and heavy-metal layers prior to transport experiments.

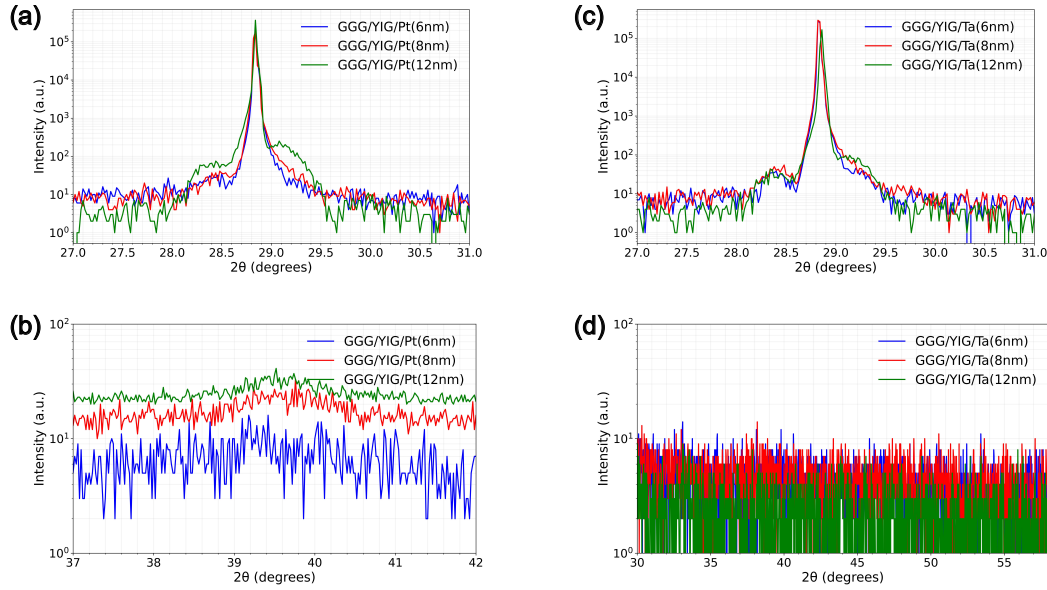


Figure 7.1 Structural characterization of YIG/HM heterostructures by XRD. (a) XRD patterns of YIG(001)/Pt bilayers with Pt thicknesses of 6–12 nm, showing the prominent YIG(004) Bragg reflection positioned near the GGG(004) substrate peak. (b) Angle-range showing the Pt(111) diffraction peak for YIG/Pt samples, confirming the textured polycrystalline nature of the Pt layer with (111) out-of-plane orientation. (c) XRD patterns of YIG(001)/Ta bilayers with Ta thicknesses of 6–12 nm, displaying similar YIG(004) features around the substrate reflection. (d) Corresponding angle-range XRD scans for YIG/Ta samples revealing no crystalline Ta peaks, consistent with amorphous Ta film structure.

7.2.2 Structural Analysis via XRD and RHEED

The XRD pattern of the YIG film shows a single-crystalline reflection near the substrate (001) reciprocal-lattice vector, and the streaky RHEED pattern (Fig. 7.2) further confirms the single crystallinity of the YIG films. Fig. 7.1 shows the typical X-ray diffraction (XRD) pattern of Pt or Ta on YIG/GGG. We observed the Pt(111) peak in YIG/Pt samples and no peak corresponding to Ta in XRD, indicating textured Pt and no observable texture in Ta thin films. Irrespective of a textured or amorphous heavy-metal layer, we expect the SHE and Hanle effect in Ta and Pt [182, 171, 170, 169].

Figure 7.2 presents the typical RHEED pattern for the GGG(001)/YIG(001) samples, obtained with the electron beam aligned along GGG[100]. The modulated streaks in the RHEED pattern corroborate the single-crystalline growth and surface with a few atomic-layer roughness. With the structural quality established, the next chapter

section introduces the magnetotransport measurement geometry and protocols used to decompose the Hall-voltage contributions.

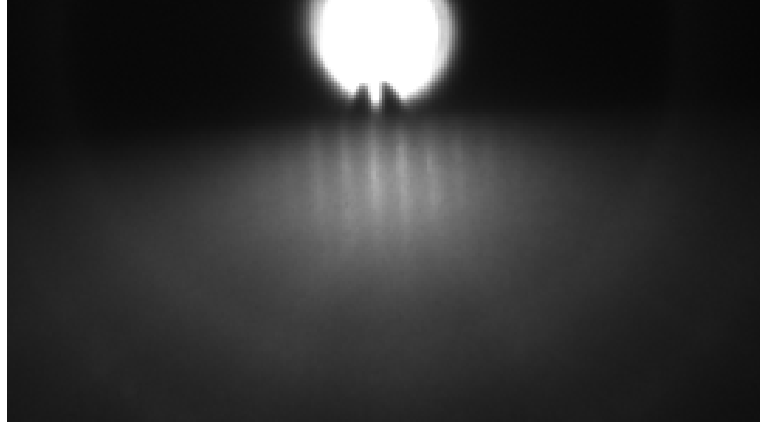


Figure 7.2 RHEED pattern of the GGG(001)/YIG(001) heterostructure, recorded with the electron beam aligned along the GGG[001] crystallographic direction.

7.3 Magnetotransport Characterization and Measurement Protocol

7.3.1 Experimental Configuration and Hall Bar Geometry

This section describes the measurement geometry and protocols used to acquire longitudinal and transverse magnetotransport signals from the YIG/HM Hall bars. Figure 7.3(a) summarizes the device geometry and measurement coordinate system used throughout this chapter. A right-handed coordinate system (x, y, z) is defined, in which the charge current J_c is along x , the Hall voltage is along y , and the film normal is along z . Fig. 7.3(a) illustrates the measurement configuration employed for Hall effect characterization of the bilayer samples. The Hall bar geometry features current injection through $\pm I$ probes and voltage detection via $\pm V$ probes arranged perpendicular to the current direction. Measurements were typically performed at room temperature. We denote the externally applied magnetic field as $\mathbf{B}_0 = \mu_0 \mathbf{H}$. This is the magnetic field set by the superconducting magnet at the sample position. High magnetic fields (up to 14 T) were used for magnetic saturation along the z direction and for disentangling OHE and Hanle precession effects [177, 178]. DC voltage measurements in delta mode were performed using a combination of a Keithley 6221

sourcemeter and a 2182A nanovoltmeter to measure longitudinal and Hall voltages, V_{xx} and V_{xy} , respectively. Thus, thermoelectric voltages and any time-varying offset voltages are eliminated. A constant direct current $I = 1$ mA was applied, corresponding to a maximum current density J of 1.25×10^{10} A m⁻². Hall voltage measurements for SH-AHE, SH-HE, and OHE were carried out by applying a magnetic field along the z direction (out of plane). In the next section, I describe how the field-sweep data are analyzed in distinct high- and low-field regimes to separate the contributions of OHE, SH-HE, and SH-AHE.

7.3.2 Longitudinal Resistivity and Phase Identification

Before analyzing the Hall responses, we first summarize the longitudinal resistivity and its thickness dependence, which are used to identify the heavy-metal phase. The electrical transport characteristics of the YIG/Pt and YIG/Ta heterostructures were characterized through four-probe resistance measurements in a Hall bar geometry with width $w = 20$ μ m, length $L = 40$ μ m, and applied current $I = 1$ mA. Fig. 7.4 shows the measured charge resistivity values for both Pt and Ta films with thicknesses ranging from 4 nm to 12 nm at room temperature (300 K).

The Pt thin films exhibit a pronounced thickness-dependent resistivity, decreasing from 40.8 $\mu\Omega$ cm at 4 nm to 26.2 $\mu\Omega$ cm at 12 nm, as shown in Fig. 7.4. As the film thickness increases, the resistivity approaches values closer to the bulk limit (10.6 $\mu\Omega$ cm for pure Pt). In contrast, the Ta films exhibit significantly higher resistivity, ranging from 307.1 $\mu\Omega$ cm to 221.2 $\mu\Omega$ cm across the measured thickness range (Fig. 7.4). The resistivity values for Ta are approximately 8 times larger than those of Pt at comparable thicknesses. The measured Ta resistivity values (221 $\mu\Omega$ cm to 307 $\mu\Omega$ cm) are consistent with β -Ta, particularly for ultrathin films with additional surface scattering effects. Ta (β -phase) is known to possess much higher resistivity (170 $\mu\Omega$ cm to 220 $\mu\Omega$ cm) compared to α -Ta (~ 13.5 $\mu\Omega$ cm). These longitudinal transport characteristics provide essential context for interpreting the magnetotransport decomposition introduced next.

7.3.3 Differentiation of Spin Hall Magnetoresistance and Anisotropic Magnetoresistance

To confirm that the observed magnetoresistance originates from SMR rather than AMR, angular-dependent scans were performed as summarized below. To distin-

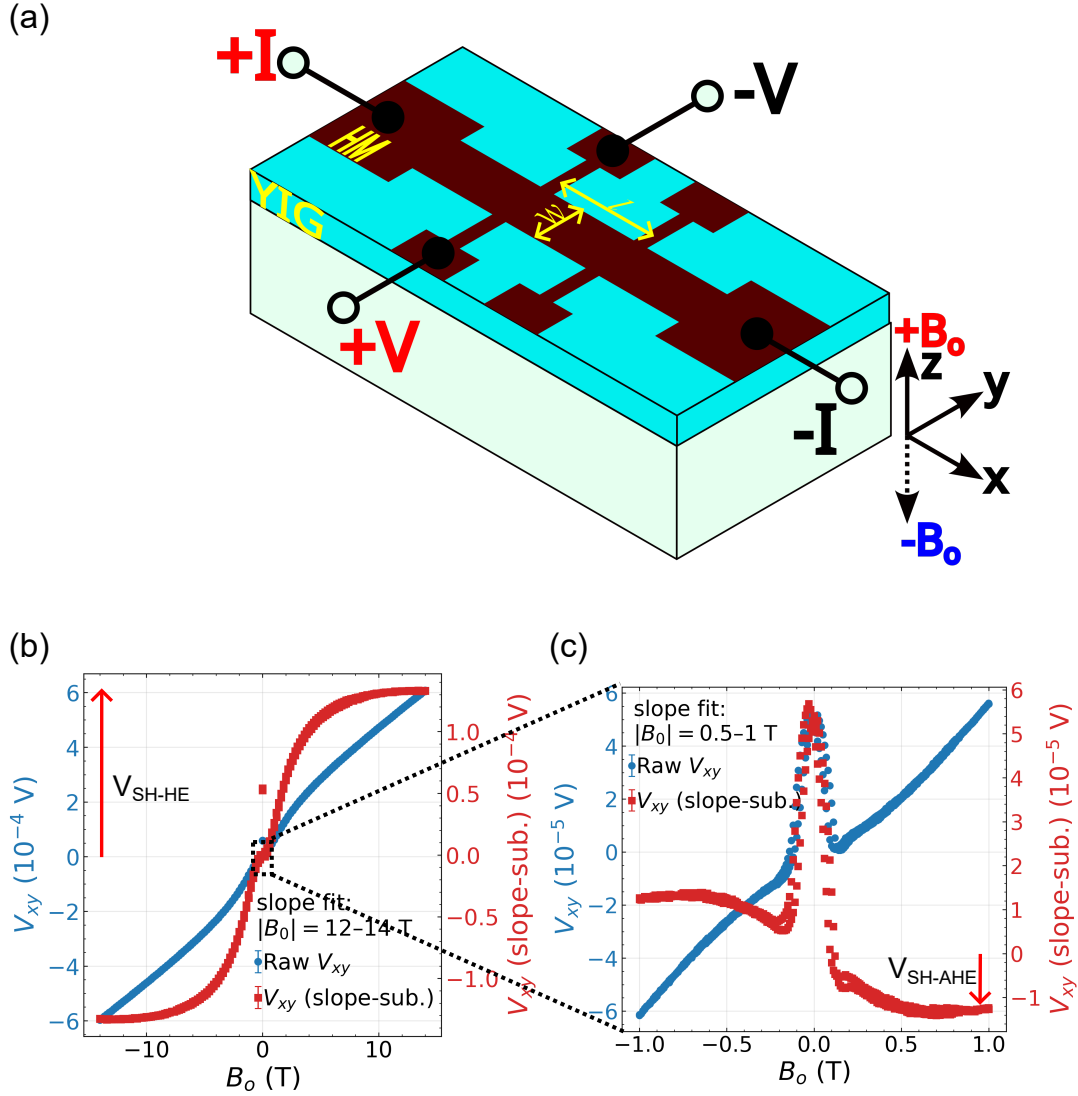


Figure 7.3 (a) Measurement schematic showing current and voltage probe configuration with perpendicular magnetic field. (b,c) Hall effect measurements in YIG/Ta(4 nm). (b) Hall voltage V_{xy} vs. magnetic field up to ± 14 T: raw data (blue) and nonlinear curvature after subtraction of the high-field linear slope (red), highlighting the SH-HE contribution. The high-field slope is determined by a linear regression of the data with $|B_0| > 12$ T. (c) Hall voltage V_{xy} vs. magnetic field up to ± 1 T: raw data (blue) and extracted SH-AHE component after subtraction of the linear background (red). The linear background is obtained from a straight-line fit to the data in the range $0.5 \text{ T} < B_0 < 1 \text{ T}$. The even-in- B_0 component at low fields ($|B_0| < 0.25 \text{ T}$) arises from transverse SMR, which depends on $M_x M_y$.

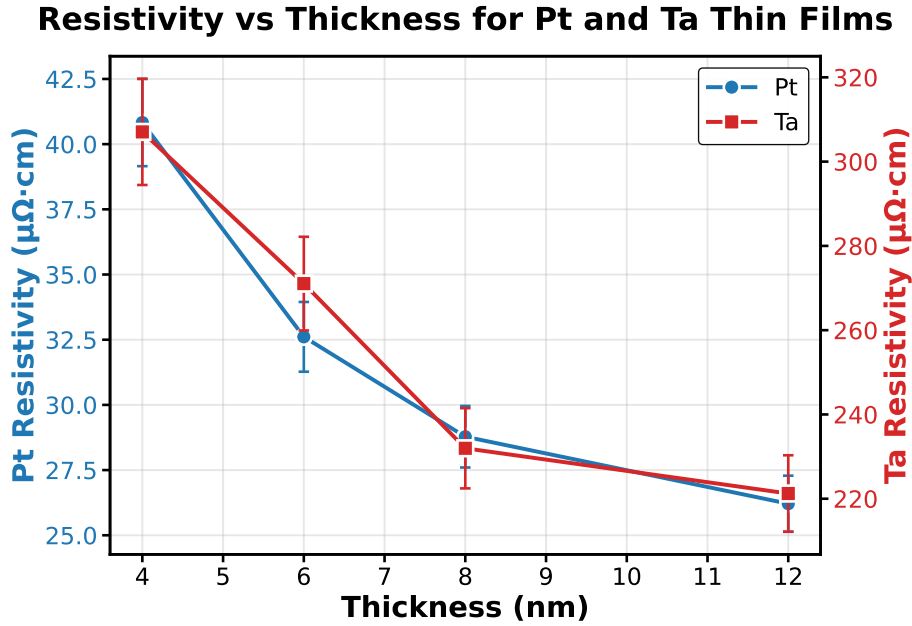


Figure 7.4 Resistivity versus thickness for Pt and Ta thin films at room temperature (300 K).

guish between SMR and anisotropic AMR contributions in our YIG/Pt and YIG/Ta heterostructures, we performed systematic angular-dependent magnetoresistance measurements at a fixed magnetic field of 0.95 T using a 3D vector-magnet cryostat system. Figure 7.5 presents the longitudinal resistance as a function of the in-plane rotation angle for both YIG/Pt(4 nm) and YIG/Ta(4 nm) bilayers, measured in three distinct planes (α , β , and γ) at temperatures of 300 K. The α -plane (xy-plane) corresponds to the case for which the external magnetic field is rotated in-plane, crossing both parallel ($\alpha = 0^\circ$) and perpendicular directions relative to the charge current. The γ -plane (xz-plane) involves rotating the external magnetic field from an in-plane orientation, for which $\gamma = 0^\circ$ (parallel to the charge current), to an out-of-plane configuration. In the β -plane (yz-plane), the field was rotated from an in-plane orientation, for which $\beta = 0^\circ$ (normal to the charge current), to an out-of-plane position.

For the YIG/Pt(4 nm) sample [Fig. 7.5(a)], we observe pronounced resistance modulations in both the α - and β -planes, exhibiting characteristic $\cos^2 \theta$ dependence with comparable amplitudes. Importantly, the γ -plane measurement shows negligible angular dependence, with any residual modulation attributable to slight sample misalignment. This angular dependence pattern is the hallmark signature of SMR, which arises from the interplay between spin currents generated by the SHE in the

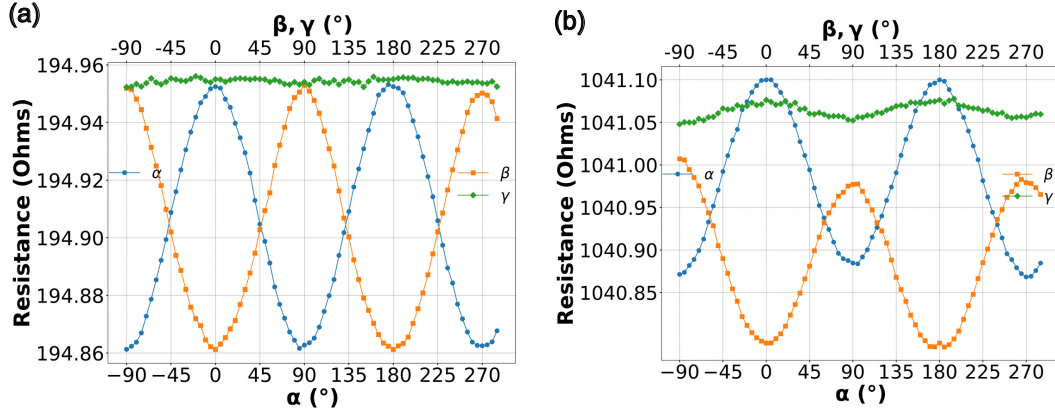


Figure 7.5 Angular-dependent magnetoresistance measurements for YIG/HM bilayers. (a) Longitudinal resistance as a function of magnetic field rotation angle for YIG/Pt(4 nm) measured at 0.95 T in the α , β , and γ rotation planes. (b) Corresponding angular scans for YIG/Ta(4 nm) in the same three rotation planes.

HM and the magnetization orientation of the adjacent MI. It further excludes AMR contributions (e.g., through a magnetic proximity effect). The YIG/Ta(4 nm) sample [Fig. 7.5(b)] displays analogous behavior with some temperature drift on top, with clear resistance modulations in the α - and β -planes and negligible signal in the γ -plane. The longitudinal charge resistance exhibits the characteristic angular dependence of SMR rather than AMR, thereby ruling out any dominant magnetic proximity effect at 300 K, which is known to play a larger role below 100 K [185–187], but can be excluded at room temperature as shown for different HM/MI combinations in literature [188–194]. Having established that SMR dominates the angular-dependent magnetoresistance at room temperature, I now turn to the regime-based separation of Hall-voltage contributions.

7.4 Isolation of Nonlinear Hall Features and Separation of Hall Contributions

The measured Hall voltage V_{xy} contains multiple contributions. In the following, I outline the protocol used to (i) identify the nonlinear curvature associated with the SH-HE from high-field (± 14 T) sweeps and (ii) extract the magnetization-dependent SH-AHE from separate densely sampled low-field (± 1 T) sweeps.

Figure 7.3(b) presents the field-dependent Hall voltage measured up to ± 14 T. The raw V_{xy} data (blue dots in Fig. 7.3(b)) exhibit a combination of OHE, SH-AHE and an additional nonlinear curvature due to SH-HE in YIG/Ta(4 nm). To highlight this nonlinear-curvature feature attributed to the SH-HE signal, the high-field slope is determined from the region $|B_0| > 12$ T, where the magnetization is saturated and additional nonlinearities are negligible, and is then subtracted from the full dataset. After subtracting this high-field slope, the extracted nonlinear component (red squares in Fig. 7.3(b)) reveals the SH-HE signature [176]. A purely ordinary Hall effect would produce only a linear slope, while a SH-AHE contribution would mainly appear as an offset that saturates once the magnetization saturates (here expected below $|B_0| \approx 0.25$ T). Since the remaining curvature evolves over much larger magnetic fields than the magnetization, it cannot originate from the SH-AHE. Moreover, it is unlikely to arise from a high-field recovery of the FI magnetization due to magnon suppression: such a recovery is expected to amount to only a few percent of the saturation magnetization, whereas the amplitude of the observed nonlinear curvature is about an order of magnitude larger than the SH-AHE signal.

Figure 7.3(c) shows the low-field behavior of the Hall voltage up to ± 1 T. Again, the raw V_{xy} data (blue dots in Fig. 7.3(c)) contains a linearly varying background together with anomalous contributions. Here, the background linearly varying signal (dominated by the field-dependent terms, i.e., OHE + SH-HE) is removed by fitting the response between 0.5 T and 1 T with a straight line and subtracting it from the data. This field window is chosen such that the magnetization-dependent anomalous contribution is expected to be saturated below $|B_0| \approx 0.25$ T, allowing the SH-AHE feature to be seen clearly. The resulting antisymmetric signal (red squares in Fig. 7.3(c)) after subtracting the linear background between 0.5 T and 1 T represents the SH-AHE. The symmetric signal for $|B_0| < 0.25$ T exhibits characteristic low-field spin Hall planar Hall effect behavior (transverse SMR) correlated with the in-plane YIG magnetization [175, 103].

The nonlinear curvature feature in Fig. 7.3(b) (red squares) cannot stem from the OHE (purely linear) or the SH-AHE (magnetization-dependent offset saturating at low fields), but is a characteristic feature of the SH-HE. In the next section, we use a more precise fitting-based approach, employing the relevant functional forms, to separate and extract magnetization-dependent Hall contributions (SH-AHE) and field-dependent responses (SH-HE and OHE) in magnetic heterostructures.

7.4.1 Multi-Component Fitting of Hall Voltage Signals

Building on the slope-subtraction procedure introduced above, we now adopt a more quantitative, multi-component fitting approach to disentangle the different Hall-voltage contributions. Figures 7.6(a,b) show the measured Hall voltage for the thickness series YIG/Ta(x nm) and YIG/Pt(x nm) in the magnetic field range of ± 14 T. This extended field range enables a reliable separation of the OHE: the SH-HE reaches its maximum below 12 T, while data above 12 T provide a window for robust statistical fitting and extraction of the OHE and SH-HE components. Figures 7.6(c,d) show Hall-voltage data for the same thickness series in the magnetic field range of ± 1 T. This low-field range emphasizes the SH-AHE, since the YIG magnetization is expected to saturate out-of-plane at approximately ± 0.25 T, allowing a clean extraction of the SH-AHE signal.

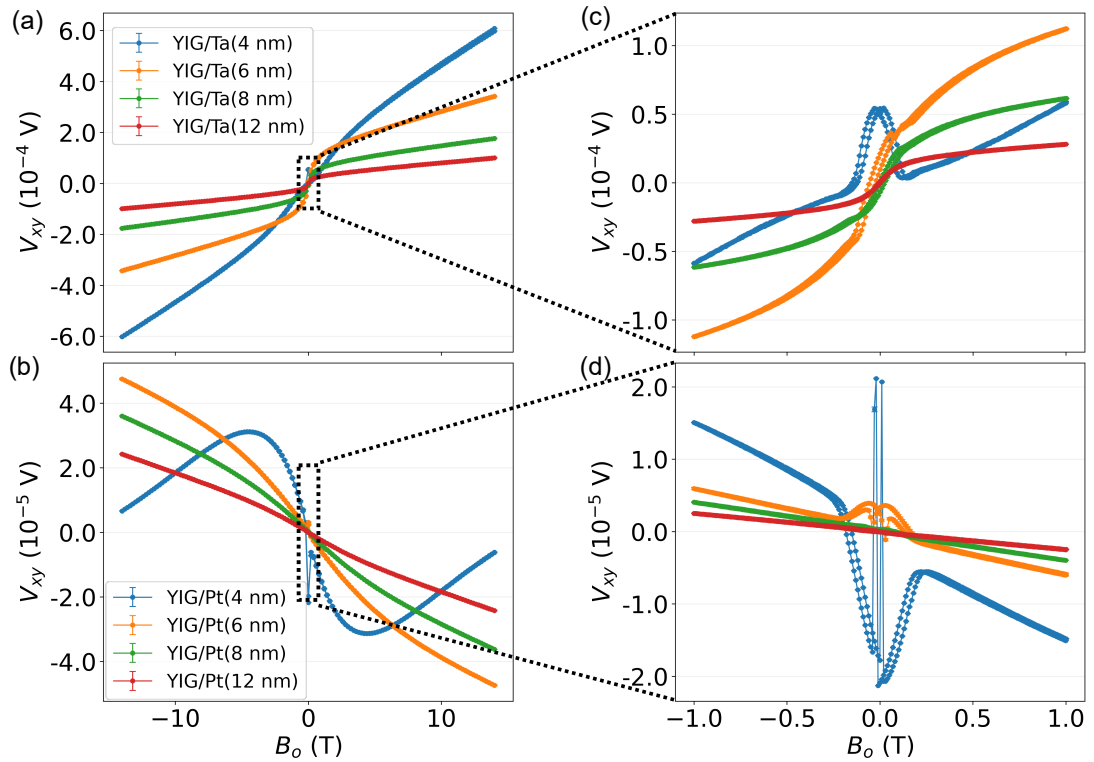


Figure 7.6 Magnetotransport characterization of (a,c) YIG/Ta and (b,d) YIG/Pt heterostructures. (a,b) Hall voltage V_{xy} as a function of out-of-plane magnetic field measured over the full magnetic field range of ± 14 T. (c,d) Close up of Hall voltage V_{xy} in the low-field regime ± 1 T.

In order to quantify the OHE, SH-HE, and SH-AHE contributions measured within the Hall voltage $V_{xy}(B_0)$ as a function of the out-of-plane magnetic field B_0 [177, 178],

we modeled the individual curves as

$$V_{xy}(B_0) = \frac{I}{d_{HM}} \left[\Delta\rho_2(B_0) n_z(B_0) + R_H B_0 \right] + V_{\text{SH-AHE}} n_z(B_0), \quad (7.1)$$

$$n_z(B_0) = \frac{B_0}{\sqrt{B_0^2 + \varepsilon^2}}, \quad (7.2)$$

$$\Delta\rho_2(B_0) = \kappa \Im \left[\frac{\Lambda(B_0)}{d_{HM}} \tanh \left(\frac{d_{HM}}{2\Lambda(B_0)} \right) \right], \quad (7.3)$$

$$\Lambda(B_0) = \left(\sqrt{\frac{1}{\lambda^2} + \frac{i}{\lambda_m(B_0)^2}} \right)^{-1}, \quad (7.4)$$

$$\lambda_m(B_0) = \sqrt{\frac{Dh}{2\pi g \mu_{B_0} |B_0|}}. \quad (7.5)$$

In Eq. (7.1), I is the applied direct current (DC), and d_{HM} is the thickness of the HM layer. The quantity R_H is the OHE coefficient; the corresponding OHE contribution $\propto R_H B_0$ is linear with the magnetic field. The $V_{\text{SH-AHE}} n_z(B_0)$ term approximates the purely odd (antisymmetric) SH-AHE term. A constant V_0 , an even (field-symmetric) baseline not of microscopic spin origin, has been subtracted to center the data around zero on the y-axis. The function $n_z(B_0)$ in Eq. (7.2) is a smooth approximation to the magnetization of YIG. It is introduced via a small regularization parameter ε to avoid numerical divergences at $B_0 = 0$ and is selected such that it yields 99.9 % out-of-plane saturation for an applied out-of-plane magnetic field of 0.25 T. The fits are performed only for magnetic field ranges larger than ± 0.25 T. In the range ± 0.25 T, the Hall voltage also depends on the in-plane magnetization of YIG in the form $M_x M_y$, which depends on magneto-crystalline anisotropy and precise measurement conditions and is therefore difficult to simulate exactly. Consequently, we fit only above ± 0.25 T and extract the SH-AHE amplitude. This separation methodology is now used for all datasets to distinguish between magnetization-dependent Hall contributions (SH-AHE) and field-dependent responses (SH-HE and OHE) in magnetic heterostructures.

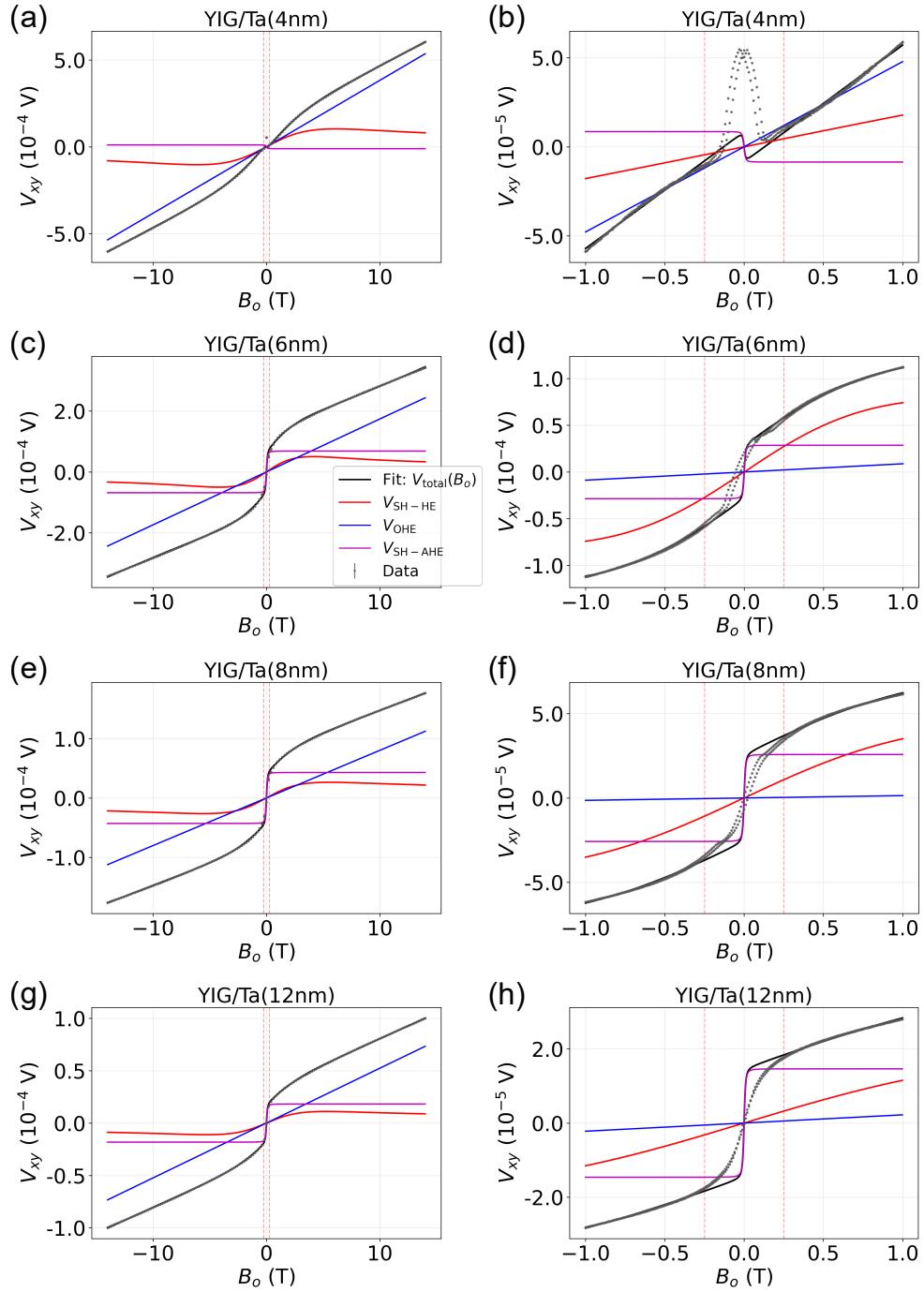


Figure 7.7 Component breakdown of magnetotransport signals for YIG/Ta bilayers with varying Ta thicknesses. All panels show measured Hall voltage V_{xy} (symbols) and fitted components including OHE, SH-HE, SH-AHE, and the total fit (solid line). (a,b) YIG/Ta(4 nm), (c,d) YIG/Ta(6 nm), (e,f) YIG/Ta(8 nm), (g,h) YIG/Ta(12 nm) measured over ± 14 T and ± 1 T ranges, respectively.

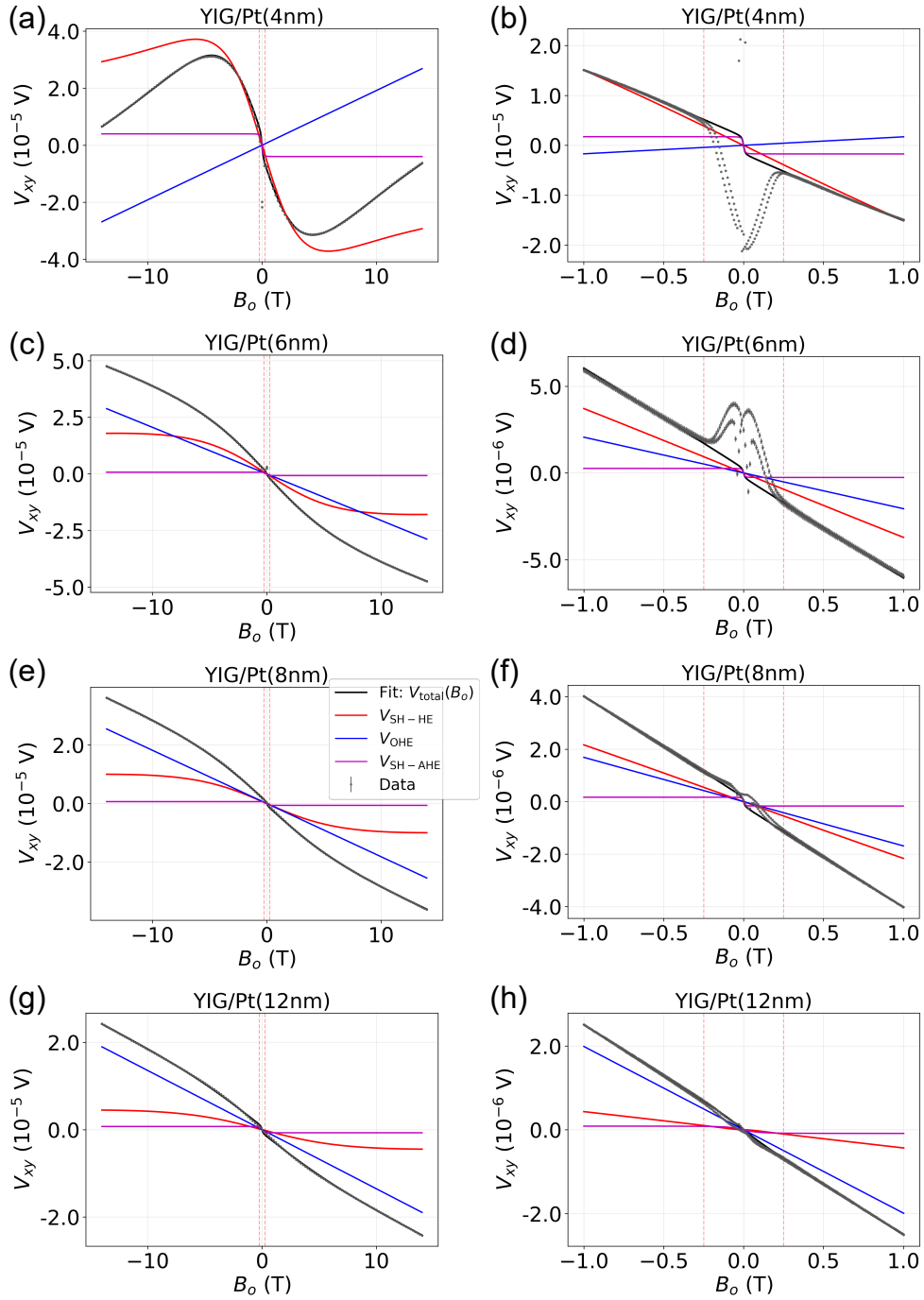


Figure 7.8 Component breakdown of magnetotransport signals for YIG/Pt bilayers with varying Pt thicknesses. All panels show measured Hall voltage V_{xy} (symbols) and fitted components including OHE, SH-HE, SH-AHE, and the total fit (solid line). (a,b) YIG/Pt(4 nm), (c,d) YIG/Pt(6 nm), (e,f) YIG/Pt(8 nm), (g,h) YIG/Pt(12 nm) measured over ± 14 T and ± 1 T ranges, respectively.

The term $\Delta\rho_2(B_0)$ in Eq. (7.3) is the odd Hanle contribution to the Hall signal. Its overall magnitude and sign are controlled by the prefactor κ (units of Ωm). For our fits, κ is allowed to be positive or negative to capture single and double SHE mechanisms from the data. The structure in square brackets in Eq. (7.3) contains two essential ingredients: (i) a complex spin-diffusion length $\Lambda(B_0)$ and (ii) a finite-thickness factor $\tanh(d_{HM}/[2\Lambda(B_0)])$ [177, 178]. The effective complex spin-diffusion length $\Lambda(B_0)$ is defined in Eq. (7.4). The real part (through $1/\lambda^2$) is set by the usual spin-diffusion length λ . The imaginary part is set by the field-dependent Hanle dephasing length $\lambda_m(B_0)$ defined in Eq. (7.5), which depends on the diffusion constant D , the g -factor $g \approx 2$, the Bohr magneton μ_{B_0} , and the reduced Planck constant $\hbar/(2\pi)$. Increasing $|B_0|$ decreases $\lambda_m(B_0)$ and strengthens the imaginary (precessional) contribution to $\Lambda(B_0)$. This causes $\Delta\rho_2(B_0)$ to develop a characteristic odd-in- B_0 peak at intermediate magnetic fields and then decay at high magnetic fields.

In this model, we treat the following quantities as fit parameters: κ , λ , D , R_H , and $V_{\text{SH-AHE}}$. The parameters I , d_{HM} , and $\varepsilon = 0.01$ are taken from the experiment and held fixed during the fit. We perform a nonlinear least-squares fit of Eqs. (7.1)–(7.5) to the measured Hall voltage $V_{xy}(B_0)$, typically restricting the fit to sufficiently large $|B_0|$ (for example, $|B_0| \gtrsim 0.25\text{ T}$ or higher) to neglect low-field spin Hall planar Hall effect contributions and ensure the magnetic layer is saturated out of plane.

Figure 7.7 and 7.8 present the multi-component fitting to the measured Hall voltage V_{xy} for YIG/Ta and YIG/Pt thickness series, respectively. It is decomposed into its constituent contributions to identify the underlying physical mechanisms governing the Hall response in these magnetic heterostructures. The raw experimental data exhibit excellent agreement with the total fit. The successful decomposition into these three components demonstrates that the measured Hall voltage in YIG/HM bilayers cannot be fully described by conventional Hall effects alone. The additional SH-HE and SH-AHE contributions arise from the interplay between spin currents generated via the SHE in the HM and the magnetic order in YIG, mediated by interfacial exchange coupling. This multi-component analysis is essential for accurately characterizing spin transport phenomena in ferromagnet/HM heterostructures and extracting material-specific parameters.

From this fit, we extract several physically important quantities: (i) R_H , the ordinary Hall coefficient; (ii) κ , which sets the overall scale and sign of the odd Hanle magnetoresistance and is linked to the strength of spin Hall spin accumulation and spin-charge conversion; and (iii) λ and D , which together characterize spin transport in the normal metal. Finally, (iv) $V_{\text{SH-AHE}}$ provides a quantitative estimate of the SH-AHE

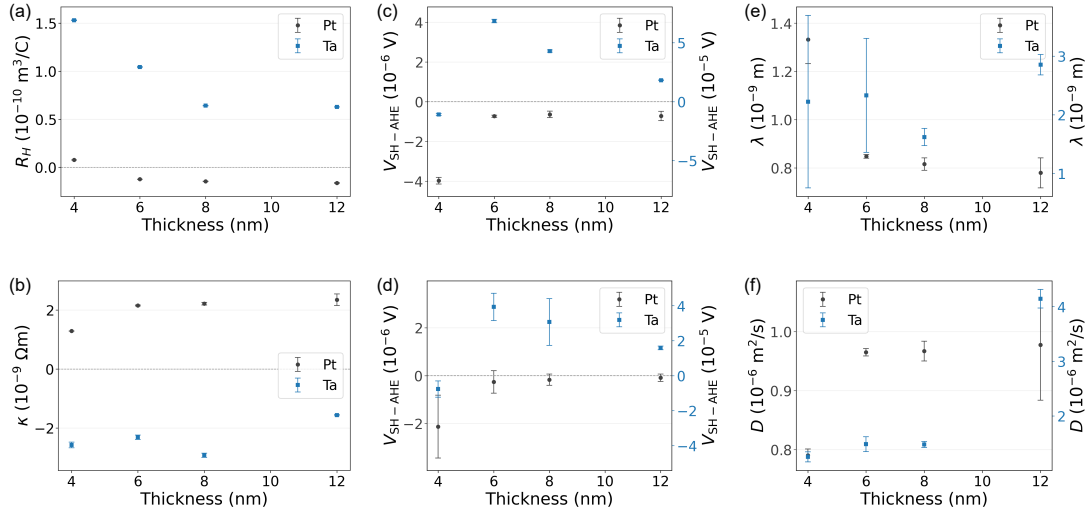


Figure 7.9 Thickness-dependent transport parameters extracted from the magneto-transport measurements of the bilayers. (a) Ordinary Hall coefficient R_H obtained from fitting the ± 14 T magnetic field scans. (b) Parameter κ that sets the SH-HE amplitude extracted from ± 14 T magnetic field scans. (c) Fitted SH-AHE amplitude extracted from ± 14 T magnetic field scans. (d) SH-AHE amplitude obtained from high-resolution ± 1 T field scans. (e) spin-diffusion length λ extracted from the ± 14 T magnetic field range. (f) Diffusion constant D extracted from the ± 14 T magnetic field range.

amplitude due to SMR, enabling separation of true SH-AHE and SH-HE spin-transport physics in the experiment. Finally, V_0 quantifies residual voltage backgrounds (for example, geometric offsets and wiring asymmetry), allowing us to distinguish true spin-transport signals from trivial voltage offsets.

7.4.2 Thickness-Dependent Evolution of Transport Parameters

We plot the fitted parameters for Ta and Pt as a function of thickness (see Fig. 7.9). In Fig. 7.9(a), R_H is positive for Ta, whereas for Pt, R_H is positive at a thickness of 4 nm and then switches sign. Figure 7.9(b) shows κ , which sets the SH-HE amplitude and is consistently of opposite sign for Ta and Pt across the thickness series. Figures 7.9(c,d) present the SH-AHE amplitude fitted from measurements at ± 14 T and ± 1 T, respectively, which constantly exhibit a negative sign for Pt. For Ta, the SH-AHE amplitude varies in sign across 4 nm to 6 nm and becomes constantly positive for thicker films. Figures 7.9(e,f) show D and λ for the same series, which determine the shape of the SH-HE curve.

7.5 Discussion

In this section, the thickness-dependent fit parameters extracted in the previous sections are interpreted in terms of the microscopic mechanisms underlying the Hall responses. I first discuss the ordinary Hall coefficient as a baseline, then the SH-HE, and finally the SH-AHE. Linear and nonlinear magnetic field dependencies are observed beyond the out-of-plane saturation of YIG. The OHE contribution is firstly discussed, followed by a comparison of single- and double SHE-dominated Hanle effects. Finally, we analyze the SH-AHE contributions and their origin in single and double SHE processes.

The ordinary Hall coefficient R_H and its sign agree with the literature. For Ta, the R_H is constantly positive, originating from dominant hole carriers at the Fermi surface [195–199]. Electronic transport in Pt is intrinsically multiband: the measured Hall response reflects the competing contributions of light, more dispersive (s-like) electron states and heavier (d-like) hole states [200]. The dominant carrier is determined by the balance of band-resolved mobilities and scattering. In the thin-film limit, for which interface and grain-boundary scattering are comparable to bulk phonon/impurity scattering, electron-like or hole-like Hall signatures may emerge depending on the microstructure and quality of the interfaces, even at similar thicknesses [176, 201–204]. This OHE coefficient provides a baseline to compare the signs of κ or $V_{\text{SH-HE}}$ and $V_{\text{SH-AHE}}$ and to decouple their odd (single SHE mechanism) or even dependence (double SHE mechanism) on the spin Hall angle θ_{SH} (Figs. 7.6–7.8).

Building on the high-field decomposition and the fitted parameter κ (Fig. 7.9(b)), the SH-HE contribution can be assessed independently of the SH-AHE. The observed nonlinear magnetic field dependencies, extending beyond the out-of-plane saturation of YIG, are consistent with the strong-precession condition that is required for the SH-HE (see Fig. 7.6 and the model in Eqs. (7.1)–(7.5)). Comparing Pt and Ta across thickness reveals that the sign of the SH-HE (through κ) tracks the sign of θ_{SH} (Fig. 7.9(b)), which points to a single SHE process rather than a double SHE mechanism in combination with the Hanle spin precession. This is consistent with a picture in which interfacial spin-dependent scattering combines with a single SHE conversion to yield a Hall response odd in θ_{SH} , as encoded by $\Delta\rho_2(B_0)$ in Eq. (7.3).

The estimated $\lambda(\text{Ta}) \approx 2 \text{ nm}$ is characteristic of the highly resistive β -Ta phase. This spin diffusion length is often consistent with the structural evolution observed in thin Ta films [169, 174, 205, 206]. Conversely, $\lambda(\text{Pt}) \approx 1.1 \text{ nm}$ falls within the range of short lengths observed when interface spin loss dominates or when high resistivity limits spin diffusion, confirming Pt's nature as a strong spin scatterer [170, 207, 208].

Longitudinal SMR in YIG/Pt(4 nm) and YIG/Ta(4 nm) shows quantitative agreement with the standard SMR model (Eq. (19) from Chen/Bauer) in angle-dependent magnetic field measurement [11] (see SI). For SH-AHE, the sign also aligns with θ_{SH} above 4 nm for both Pt and Ta (Figs. 7.9(c,d)). While the sign of SH-AHE for the 4 nm samples is even with θ_{SH} and, therefore, supports a double SHE interpretation. At larger thickness, the opposite SH-AHE sign for Pt versus Ta may reflect either a growing contribution of the single SHE mechanism or a change in the sign of the imaginary part of the spin-mixing conductance G_i . Although all films were prepared under identical nominal conditions, sequential growth may introduce interfacial variations that affect G_i . The systematic reduction of SH-AHE amplitude with thickness for 6 nm, 8 nm and 12 nm (Figs. 7.9(c,d)) is indicative of a controlled competition between single and double SHE mechanisms, rather than an arbitrary trend. Nevertheless, the abrupt sign change for Ta between 4 nm and 6 nm is indicative of a lack of controlled competition, with a potential role for G_i .

7.6 Conclusion and Outlook

The results presented in this chapter are inconsistent with a purely θ_{SH}^2 (double SHE) SH-AHE framework for YIG/Ta(Pt), motivating a model that includes single SHE contributions, as discussed in Ref. [12]. To avoid ambiguity, we propose the following nomenclature for Hall responses observed or discussed here:

- **Single spin Hall anomalous Hall effect (SSH-AHE):** preferred over "non-local AHE" (as proposed in Ref. [12]).
- **Double spin Hall anomalous Hall effect (DSH-AHE):** preferred over the generic "SH-AHE" reported experimentally in Ref. [176].
- **Single spin Hall Hanle effect (SSH-HE):** experimental observation reported here; a corresponding theory is, to our knowledge, not yet available.
- **Double spin Hall Hanle effect (DSH-HE):** preferred over the umbrella term "SH-HE" [182].

The spin mixing conductance G_i is not an intrinsic parameter of the SH-HE theory, but it can influence the SH-AHE via interfacial exchange effects. Such interfacial asymmetries could potentially influence the observed trends for SH-AHE. However, in the present study, we did not directly quantify interfacial parameters such as G_i ,

nor did we systematically vary the growth order to isolate interfacial asymmetries. To fully address these aspects future work should pursue several directions: (i) measure temperature dependences to constrain G_i and assess its contribution, (ii) employ interface engineering (e.g., dusting layers) to tune single vs. double SHE balance, (iii) systematically vary both the HM and MI materials to establish the scope of the single SHE mechanism across different material systems, and (iv) develop a comprehensive microscopic theory for the SSH-HE that is consistent with the experimental trends observed in Fig. 7.9(b,c).

We report experimental evidence that the single SHE mechanism governs the Hall voltage in the SH-HE response and provides the dominant contribution to SH-AHE in YIG/Pt(Ta) bilayers. By jointly analyzing distinct magnetic field regimes (Figs. 7.7–7.8) within Eqs. (7.1)–(7.5), we consistently separate OHE, SH-HE, and SH-AHE signals with consistent sign conventions set by R_H . This simultaneous extraction reduces ambiguities that have conflated single and double SHE-conversion channels in SH-HE/SH-AHE analyses. Using heavy metals with opposite θ_{SH} , the 4 nm series provides particularly clear odd behavior of SH-HE on θ_{SH} because SH-AHE and R_H share the same sign across Pt and Ta, enabling direct sign comparisons across thickness.

Chapter 8

Conclusion and Outlook

8.1 Summary of Main Results

This thesis combines material synthesis, highly sensitive magnetic probes, and quantitative spin transport measurements to build a coherent picture of spin phenomena in oxide heterostructures. The integration of structural control with transport and spectroscopic investigations is crucial for interpreting the observed signals and distinguishing between intrinsic and extrinsic contributions.

RuO₂ Thin Films: Growth, Magnetism, and Spin Transport

The foundation of this work is the successful fabrication of high-quality epitaxial RuO₂ thin films on TiO₂ substrates. Chapter 4 established growth parameters using *in situ* RHEED monitoring to characterize the growth mode and surface structure, complemented by ex-situ X-ray diffraction and other probes to confirm single-crystalline quality and the formation of a single crystallographic variant. Because the magnetic and electronic properties of RuO₂ are highly sensitive to structural domains, strain, and interface composition, this robust structural control is a prerequisite for identifying intrinsic physical phenomena in transport and magnetic measurements [16, 15, 40].

Building on this structural basis, Chapter 5 investigated spin transport in RuO₂/Py bilayers by means of ADMR. The measurements revealed a pronounced, robust negative ADMR signal. Contrary to theoretical expectations for an intrinsic spin splitting originating from the predicted bulk altermagnetic nature of RuO₂, qualitative analysis using drift-diffusion modelling shows that the magnetoresistance is predominantly driven by IGSCs. These IGSCs arise from localized spin-orbit scattering at the RuO₂/Py interface and are converted into a measurable charge signal via the iSHE. The data therefore indicate that extrinsic interfacial spin phenomena overshadow potential intrinsic bulk spin-splitting signatures in the present RuO₂ films [97].

Chapter 6 employed depth-resolved LE- μ SR to clarify the magnetic ground state of RuO₂. The measurements demonstrate that static, inhomogeneous magnetic order is confined predominantly to the near-surface region: the magnetically ordered volume fraction is estimated to be $\sim 8.5\%$ within a top layer of nominal thickness ~ 10 nm. The order shows a gradual temperature dependence and is consistent with an extrinsic origin related to surface termination, defects, or dimensionality effects, rather than a bulk altermagnetic phase [140]. These findings provide a crucial constraint on the location and nature of magnetic moments available for spin coupling and significantly refine the interpretation of the RuO₂-based spin transport experiments.

Benchmarking Spin Transport in YIG-Based Heterostructures

To validate the quantitative spin-transport framework, Chapter 7 examined prototypical YIG/Pt and YIG/Ta bilayers, using magnetotransport measurements to test the key prediction of Ref. [12] regarding the sign of spin Hall angle dependence of the Hall signal. By jointly analyzing distinct magnetic-field regimes, we consistently separated OHE, SH-HE, and SH-AHE contributions with sign conventions fixed by R_H . The experiments provide clear evidence that a single SHE mechanism governs the Hall voltage in the SH-HE response and delivers the dominant contribution to SH-AHE: using heavy metals with opposite spin Hall angles, the 4 nm series reveals that the sign of the measured Hall signal directly follows the sign of the bulk θ_{SH} of the adjacent HM, even though SH-AHE and R_H retain the same sign across Pt and Ta. This behavior is fully consistent with the theoretical description, which considers the combined action of the bulk SHE and spin-dependent interfacial scattering, and it demonstrates that the experimental protocols and analysis strategies developed in this thesis reliably capture interface-sensitive spin phenomena while offering a generalizable platform for engineering and testing spintronic responses in multilayer systems.

Physical Insights and Implications

This section takes a broader view and outlines the main conceptual advances, focusing on how the results refine our understanding of spin transport in RuO_2 and YIG-based systems within the context of oxide spintronics and altermagnetic materials.

RuO_2 as a Platform for Altermagnetic Spintronics

The combination of epitaxial growth, structural characterization, and depth-resolved magnetism measurements establishes RuO_2 as a structurally robust and electronically metallic platform for spintronics. In parallel, three collaborative studies on the same epitaxial $\text{RuO}_2/\text{TiO}_2$ film platform reported altermagnetic-consistent fingerprints, including time-reversal symmetry breaking in the band structure without net magnetization (ARPES), strain-linked dichroic Ru-core-level responses (XMCD/XPS), and a polarization-dependent all-optical spin-polarization response consistent with the proposed d -wave symmetry. Taken together, these results motivate reconciling surface-confined magnetism and interface-dominated transport with spectroscopic/optical signatures of altermagnetic order in strained epitaxial films. LE- μ SR demonstrates

that static magnetic order is predominantly confined to the near-surface region, likely linked to broken symmetry, dimensionality, or defect-related effects, while the film interior remains largely nonmagnetic within experimental sensitivity. At the same time, the rutile structure and high-quality epitaxy provide a stable and reproducible environment for spin transport. A key distinguishing feature of RuO_2 among proposed altermagnets is the coexistence of metallic conduction and magnetic order, allowing spin angular momentum to be carried efficiently by charge carriers [209]. This contrasts with many insulating or semiconducting candidates and positions RuO_2 as an attractive active electrode material.

The observation that RuO_2 can effectively transfer spin angular momentum to an adjacent ferromagnet, even when the dominant mechanism is extrinsic IGSCs, shows that the material can function as a spin-current injector or absorber in practical heterostructures. Future device applications will depend on enhancing and controlling this functionality, for example by exploiting epitaxial strain or stoichiometry to stabilize the intrinsic altermagnetic phase and by refining interface engineering to optimize spin-charge conversion processes. In this sense, RuO_2 serves both as a testbed for altermagnetic concepts and as a realistic oxide electrode for spintronic architectures.

Spin-Current Generation, Transmission, and Detection in Oxide Heterostructures

Across the RuO_2/Py and $\text{YIG}/\text{Pt}(\text{Ta})$ systems, the thesis develops a unified conceptual model for the generation, propagation, and detection of spin currents in oxide-based heterostructures. The experimental results show that the spin-current response of these systems is governed by a competition between intrinsic material properties and extrinsic interfacial effects. In RuO_2/Py , IGSCs arising from localized spin-orbit scattering at the interface dominate the ADMR response and mask the subtle signatures expected from non-relativistic spin splitting in an ideal altermagnetic bulk. In YIG/Pt and YIG/Ta , the sign reversal of the Hall signal tracks the reversal of the bulk θ_{SH} in the HM, in quantitative agreement with the theoretical framework of Ref. [12]. Together, these findings demonstrate that, at the current stage of materials development, macroscopic spintronic functionality in oxide heterostructures is largely dictated by extrinsic interfacial phenomena rather than by intrinsic band-structure effects alone. Consequently, progress in oxide-based altermagnetic spintronics will require a strong emphasis on atomic-scale interface control and defect engineering alongside continued efforts to stabilize intrinsic phases.

Methodology and Limitations

The research establishes several methodological advances that are critical for studying spintronic phenomena in complex oxides. Optimized epitaxial RuO_2 film growth on TiO_2 substrates, monitored by *in situ* RHEED, enables the reproducible realization of high-quality films with a single crystallographic variant. This structural control significantly enhances the interpretability of anisotropic transport signals, which are often obscured in multi-variant films. Robust RuO_2/Py and benchmark $\text{YIG}/\text{Pt}(\text{Ta})$ heterostructures were fabricated and characterized routinely, allowing for systematic application of interface-sensitive spin-transport measurements, in particular ADMR experiments to distinguish IGSCs from potential intrinsic altermagnetic contributions. In parallel, the use of depth-resolved $\text{LE-}\mu\text{SR}$ on sufficiently thick RuO_2 films provided high sensitivity to confined magnetic order, overcoming limitations of earlier $\text{LE-}\mu\text{SR}$ studies on thinner systems.

Despite these advances, several intrinsic limitations constrain the precision of quantitative conclusions. Sample-to-sample variations, especially in interface roughness and subtle deviations from ideal RuO_2 stoichiometry, introduce uncertainties that can influence both magnetic and transport properties; in RuO_2 , defect chemistry is indeed suggested to play a major role. Signal-to-noise constraints, particularly in transport measurements and in muon experiments probing a small magnetically ordered volume fraction ($\sim 8.5\%$ near the surface), limit the ability to resolve very weak intrinsic signals. As a result, the absolute values of extracted parameters carry non-negligible uncertainties, even though the qualitative trends, such as the sign reversal of the Hall signal in $\text{YIG}/\text{Pt}(\text{Ta})$ or the dominance of IGSCs over bulk spin-splitting signals in RuO_2/Py remain robust. These experimental and analytical challenges, especially those related to atomic-scale interface control and sensitivity limitations, motivate the future developments outlined in the following section.

8.2 Outlook

The central open challenge in RuO_2 is the definitive stabilization and detection of the intrinsic altermagnetic phase, which appears highly sensitive to sample properties. Future research should systematically investigate the influence of epitaxial strain, substrate choice, and stoichiometry on the emergence of spin-splitting signals, for example by employing lattice-mismatched substrates or controlled alloying and doping strategies to stabilize the predicted phase. A critical step will be to quantitatively cor-

relate structural metrics such as oxygen/ruthenium stoichiometry, strain, and thickness with spin-transport parameters extracted from ADMR or related measurements. In addition, integrating high-quality RuO₂ films with emerging quantum materials, such as two-dimensional magnets or topological insulators, offers intriguing opportunities to exploit proximity effects and engineer enhanced or novel spin functionalities [209].

The observation of surface-confined magnetism calls for a concerted effort to reconcile spectroscopic and transport perspectives on RuO₂. This is particularly timely, given recent collaborative ARPES, XMCD/XPS, and optical-pump results on the same epitaxial film platform, which report alternamagnetic-consistent signatures. Beyond the present muon study, complementary techniques such as polarized neutron scattering and resonant X-ray scattering will be essential to determine the precise magnetic configuration (including the Néel vector orientation) and the temperature evolution of both surface and potential bulk magnetism. These experimental efforts should be guided by microscopic modelling that explicitly considers the role of defects, interfacial exchange interactions, and surface symmetry breaking in shaping intrinsic and extrinsic spin Hall responses, as well as the effective spin-mixing conductance at RuO₂-based interfaces.

The demonstrated capacity of RuO₂-based heterostructures to generate and absorb spin currents suggests their potential as active elements in future spintronic devices. Near-term work should focus on fabricating optimized spin-current valves, magnetic tunnel junctions, and three-terminal devices to demonstrate reliable, all-electrical manipulation of magnetization in adjacent layers, potentially exploiting the unique spin polarization capabilities envisioned for altermagnets to achieve field-free switching. Key challenges will include ensuring the scalability and long-term stability of the epitaxy-dependent spin response and pushing device operation towards the high frequencies (up to the terahertz regime) that are intrinsic to altermagnetic dynamics. Addressing these issues will be pivotal for translating the material-level insights of this thesis into functional device concepts.

Final Remarks

This thesis provides a comprehensive experimental understanding of spin-current phenomena in rutile RuO₂ and YIG-based heterostructures, highlighting the critical role of extrinsic interfacial effects in comparison to intrinsic bulk responses in the former. Combining controlled epitaxial growth, interface-sensitive spin-transport measurements, and depth-resolved LE- μ SR, the work shows that static magnetic order in RuO₂

thin films is confined to a near-surface region of order 10 nm with a limited volume fraction, and that IGSCs predominantly govern the spin-charge conversion efficiency in RuO₂/Py. In parallel, the same quantitative spin-transport methodologies applied to YIG/Pt and YIG/Ta bilayers confirm the predicted dependence of the nonlocal AHE signal on the sign of the θ_{SH} , thereby validating the underlying theoretical framework and experimental approach.

Beyond these specific systems, the thesis contributes to the broader field of spintronics by advancing the materials science and physical understanding of emerging magnetic phases, particularly altermagnets and complex oxides, that are expected to drive the next generation of spin-based technologies. The questions formulated here and the refined analytical and experimental methods, ranging from structural control to the careful disentangling of intrinsic and extrinsic spin effects in transport, provide a transferable toolbox for investigating magnetic metals and oxide heterostructures more generally. In this sense, RuO₂-containing heterostructures exemplify how oxide platforms can bridge fundamental concepts in altermagnetic spintronics with realistic routes towards energy-efficient device architectures.

Appendix A

RHEED installation to the PLD chamber

This appendix explains the details of the installation of the RHEED to the PLD system and setting it up to record diffraction patterns. Staib instruments supply the high-pressure RHEED with RH 40 electron gun adapted for the double differential pumping and phosphor on a fluorescent screen protruding inside the vacuum chamber. The camera records the diffraction images (k700-12 from k-Space associates) and the process by the Win 10 kSA 400 RHEED System. Installation started with moving the PLD chamber as already discussed in section 3.4.

The new position of the chamber came with the requirement of redesigning and rebuilding the laser safety housing to keep the beam path fully encased during regular laser operation. In the new layout, one less mirror is required, and the laser beam stays in the plane. This change in laser safety housing can be seen in figure 4.1 and the picture of the current state of the PLD with the RHEED System in figure A.1. It is installing the electron gun and pumping unit, which required building, the support pieces, and the adapter ConFlat vacuum flanges. The drawing for the supporting pieces and modification in the laser housing was given to our tool shop, which builds the necessary parts. After testing the RHEED system for diffraction, it was realized that the sample at growth height could not be reached with the limited in-built possible movement of the electron beam. Therefore, it is required to redesign the adapter flanges and the supporting pieces to move the whole electron gun to a suitable position. With re-adapted flanges and supporting pieces, the electron beam can be made to hit the sample at deposition height. The discussed parts are shown in the figure A.1. In the meantime, another challenge was to obtain the diffraction pattern, as it requires

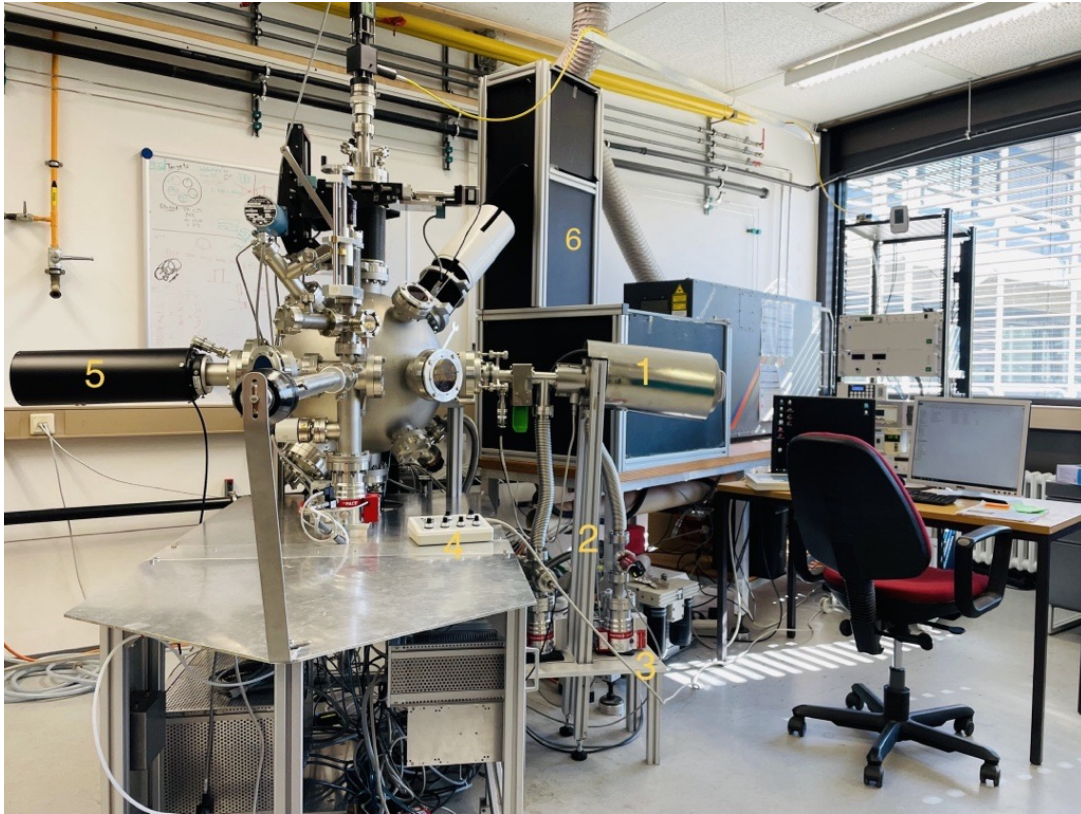


Figure A.1 PLD chamber with RHEED installed with 1) electron gun, 2) support for RHEED gun, 3) support for RHEED's pumping system, 4) e-beam controller, 5) camera setup, and 6) laser safety housing.

meticulous beam alignment to have diffraction and proper camera (k700-12 from k-Space associates Inc.) settings for recording the focused images. The "Beam Finder" feature of the beam controller is proven to be instrumental in locating the beam and ensuring its direct exit from the gun's aperture without reflecting from any surface. With this feature activated, the beam wobbles in one direction and can be manually adjusted perpendicularly.

Appendix B

Use of AI Tools

This appendix documents the use of AI-based tools during the preparation of this thesis. All scientific content, data analysis, interpretations, and conclusions were independently developed by the author. AI tools were used exclusively in a supportive role for language refinement, formatting assistance, code debugging, and conceptual clarification. The author retained full editorial and scientific control at all times, and all AI-generated outputs were critically reviewed, verified, and selectively integrated.

Overview of AI Usage

The primary modes of AI assistance and their specific roles are summarized in Table B.1. In particular, AI tools supported:

- written communication and language quality for a non-native English author,
- code-related debugging and syntax clarification,
- preliminary explanations of complex topics, treated as non-authoritative and independently verified.

AI Tool	Used for	Purpose and Scope	When / Where
ChatGPT	Language refinement, $\text{\LaTeX}$ formatting suggestions, Python debugging, and conceptual clarification	Improving grammar, spelling, clarity, and academic tone; suggesting alternative phrasings for author-developed ideas; resolving syntax-related issues in Python code; providing preliminary explanations of complex concepts, all of which were independently verified and critically assessed	Throughout the thesis
DeepL Translator	Translation of English text into German	Ensuring linguistic precision and stylistic consistency between the English and German versions of the abstract	German abstract
Grammarly	Grammar, punctuation, and stylistic correction	Final proofreading to enhance grammatical accuracy and consistency of academic writing, without altering scientific content or authorial intent	Final proof-reading phase

Table B.1 Overview of AI-based tools used during the preparation of this thesis. AI assistance was limited to supportive tasks; all outputs were reviewed, verified, and incorporated under full authorial control.

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List of Publications

- **A. Akashdeep**, E.M. Ababneh, C. Schmitt, E. Galíndez-Ruales, F. Fuhrmann, T. Kuschel, M. Kläui, V. Amin, and G. Jakob, "Angle-dependent magnetoresistance induced by interface-generated spin current in RuO₂/permalloy heterostructures", *Phys. Rev. Appl.* **24**(5), 054018 (2025).
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- **A. Akashdeep**, D.M. Tran, M. Kläui, G. Jakob, and T. Kuschel, "Single spin Hall anomalous Hall and Hanle effect in YIG/Ta and YIG/Pt bilayers", under preparation (2026).
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Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Removed due to data privacy

Curriculum Vitae

Removed due to data privacy

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