

...A THEORY SHOULD BE AS SIMPLE AS POSSIBLE, BUT NOT SIMPLER. A TRUE UNDERSTANDING OF PHYSICS BEGINS ONLY WHEN YOU CAN EXPLAIN THE SAME PHENOMENON IN SEVERAL DIFFERENT WAYS... A GOOD THEORY SHOULD NOT ONLY EXPLAIN WHAT IS KNOWN, BUT PREDICT WHAT HAS NOT YET BEEN SEEN. THE TASK OF THE PHYSICIST IS NOT TO CATALOGUE NATURE, BUT TO REVEAL THE SIMPLICITY HIDDEN IN ITS COMPLEXITY.

– LEV DAVIDOVICH LANDAU

MECHANISMS OF SPIN AND CHARGE TRANSPORT ACROSS OXIDE AND ALLOY INTERFACES

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By

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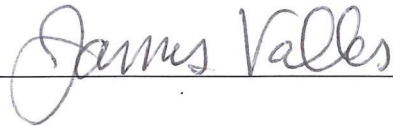
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**Abstract of *Mechanisms of Spin and Charge Transport Across Oxide and Alloy Interfaces*,
by Vineetha S. Bheemarasetty, Ph.D., Brown University, February 2026.**

The control of spin currents through interfacial and spin-orbit-driven effects is central to the development of next-generation spintronic technologies. This dissertation investigates how spin-orbit coupling (SOC) and interfacial symmetry govern the generation, transmission, and conversion of spin currents across three model material systems including half-metallic CrO_2 , $\text{RuO}_2/\text{CrO}_2$ bilayers, and Ru-Pt alloys. These systems were strategically chosen to trace the full pathway of spin manipulation: CrO_2 for intrinsic spin polarization and quantum transport, $\text{RuO}_2/\text{CrO}_2$ for interfacial exchange and spin coupling, and Ru-Pt for extrinsic spin-charge conversion.

Epitaxial CrO_2 thin films serve as a platform for intrinsic spin current generation and quantum transport. Their nearly 100% spin polarization enables robust, coherent spin currents while low-temperature transport measurements reveal quantum oscillations and high mobility, indicative of phase-coherent electronic motion. These results establish CrO_2 as an ideal half-metal for generating spin-polarized currents and exploring SOC-influenced quantum transport phenomena.

$\text{RuO}_2/\text{CrO}_2$ heterostructures exemplify interfacial exchange and spin coupling where an active spin-orbit antiferromagnetic RuO_2 layer interacts with the CrO_2 ferromagnet. Magnetization measurements reveal spin reorientation and exchange bias effects, illustrating how SOC and broken inversion symmetry at the interface can modulate spin alignment, interlayer magnetic interactions, and the transmission of spin angular momentum across oxide heterostructures.

Ru-Pt alloys provide a model for extrinsic spin-to-charge conversion, where spin currents generated in a ferromagnet are converted into measurable charge signals via the spin Hall effect. Systematic mapping of the spin Hall angle across the full composition range demonstrates how crystal structure and SOC control the efficiency of spin-charge interconversion. Beyond conventional impurity tuning, Ru-Pt demonstrates structural phase evolution as an effective tuning parameter and poses as a promising alternative for spin-orbit torque devices. These results highlight how extrinsic SOC effects can be engineered to detect, manipulate, or generate spin currents in metallic systems.

Together, these studies establish a unified framework connecting intrinsic spin generation and interfacial spin coupling to extrinsic spin-to-charge conversion and detection. The results provide insight into design principles for low-power, scalable spintronic devices that exploit SOC and symmetry engineering to control spin currents across diverse material platforms.

Chapter 1 | Fundamentals of Spintronics

SPINTRONICS, OR spin-based electronics, is an emerging field that exploits the intrinsic angular momentum of the electron (i.e. its spin) in addition to its charge. By harnessing this additional degree of freedom, spintronic devices promise enhanced energy efficiency, non-volatility, thermal stability, and faster operation compared to conventional charge-based electronics. The binary nature of electron spin, which can align either “up” or “down,” provides a natural mechanism for encoding digital information, analogous to charge-based logic.

In crystalline solids, the collective behavior of electrons gives rise to magnetic ordering, which forms the foundation for many spintronic phenomena. Effects such as giant magnetoresistance (GMR) and spin-transfer torque (STT) arise from the interplay between spin-polarized currents and magnetization. These have enabled technologies including hard drive read heads and magnetic random-access memory (MRAM). Beyond these classical effects, the interaction between electron spin and orbital motion (i.e. spin-orbit coupling (SOC)) plays a central role in modern spintronics, giving rise to phenomena such as the spin Hall effect (SHE) and spin-orbit torque (SOT), which allow the generation, manipulation, and detection of spin currents in non-magnetic materials.

Spin-dependent scattering mechanisms, including impurity scattering, magnon scattering, and interfacial scattering, further modulate the spin transport and dictate both the efficiency of spin current transmission and the lifetimes of spin-polarized carriers. Together, these fundamental concepts define a cohesive framework for understanding spin current generation, transmission, and conversion in model systems including half-metallic CrO_2 , $\text{RuO}_2/\text{CrO}_2$ heterostructures, and Ru–Pt alloys presented in subsequent chapters.

Magnetism in Crystalline Solids

EXCHANGE INTERACTION

Magnetism in solids originates from the quantum-mechanical exchange interaction which arises from the combination of Coulomb repulsion and the antisymmetric nature of the total electronic wavefunction due to the Pauli exclusion principle. For two electrons with wavefunctions ψ_i and ψ_j , the exchange integral can be expressed as

$$J_{ij} = \int \psi_i^*(\mathbf{r}_1)\psi_j^*(\mathbf{r}_2) \frac{e^2}{4\pi\epsilon_0|\mathbf{r}_1 - \mathbf{r}_2|} \psi_j(\mathbf{r}_1)\psi_i(\mathbf{r}_2) d\mathbf{r}_1 d\mathbf{r}_2. \quad (1)$$

The sign and magnitude of J_{ij} determine whether the spins align parallel ($J_{ij} > 0$) or antiparallel ($J_{ij} < 0$). In transition metals and oxides, exchange coupling is mediated through indirect mechanisms such as super-exchange and double exchange, depending on the overlap between atomic orbitals and the intermediate anions.

A simplified Hamiltonian capturing this interaction is given by the Heisenberg model:

$$\mathcal{H} = - \sum_{i \neq j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (2)$$

where $\mathbf{S}_i$ and $\mathbf{S}_j$ represent localized spin moments. The competition among exchange constants J_{ij} for different atomic pairs gives rise to a variety of magnetic phases and textures observed in crystalline materials.

EXCHANGE SPLITTING

In magnetic materials, the exchange interaction between neighboring electron spins leads to an energy difference between spin-up and spin-down electronic states. This effect is referred to as exchange splitting, which arises from the Pauli exclusion principle and Coulomb repulsion. Together, they favor parallel spin alignment in ferromagnets to minimize the total energy of the system.

The magnitude of exchange splitting, Δ_{ex} , determines the degree of spin polarization at the Fermi level and can be expressed as the energy separation between the majority ($\uparrow$) and minority ($\downarrow$) spin bands:

$$\Delta_{\text{ex}} = E_{\downarrow} - E_{\uparrow}. \quad (3)$$

In a strong FM, Δ_{ex} is sufficiently large that one spin channel becomes completely filled while the other remains metallic, resulting in a half-metallic band structure. This is the case in CrO_2 , where the spin-up channel crosses the Fermi level while the spin-down channel exhibits a bandgap.

Exchange splitting plays an important role in determining the Curie temperature, coercivity, and magnetocrystalline anisotropy of a material. In systems with significant spin-orbit coupling (SOC), complex spin textures, spin-orbit torques (SOT), and anomalous transport behavior can arise.

TYPES OF MAGNETISM

Depending on the balance between exchange coupling, crystal symmetry, and electronic structure, solids can exhibit different forms of magnetic order:

- **Diamagnetism:** A weak, negative susceptibility response arising

from Lenz's law, where induced currents oppose the applied magnetic field. It is a universal property of all materials but typically masked by stronger effects.

- **Paramagnetism:** Describes materials with unpaired spins that align weakly with an applied field, leading to a positive but temperature-dependent susceptibility following Curie's law:

$$\chi = \frac{C}{T - \theta}, \quad (4)$$

where C is the Curie constant and θ represents the Weiss temperature.

- **Ferromagnetism:** Characterized by parallel spin alignment due to positive exchange coupling ($J_{ij} > 0$), resulting in spontaneous magnetization below the Curie temperature (T_C). Examples include Fe, Co, and Ni.
- **Antiferromagnetism:** Neighboring spins align antiparallel ($J_{ij} < 0$), leading to zero net magnetization in the ground state at zero field. The transition temperature is termed the Néel temperature (T_N).
- **Ferrimagnetism:** Similar to antiferromagnetism but with unequal opposing sublattice moments, leading to a nonzero net magnetization. Common in magnetic oxides such as magnetite (Fe_3O_4).
- **Half-metallicity:** One spin channel behaves metallically while the other is insulating, leading to 100% spin polarization at the Fermi level. CrO_2 is a prototypical example, relevant for spintronic applications.

Anisotropy and Magnetocrystalline Effects

MAGNETOCRYSTALLINE EFFECTS

Magnetic anisotropy refers to the directional dependence of a material's magnetic properties, arising from SOC, crystal symmetry, and strain effects. The total anisotropy energy can be decomposed into several contributions:

$$E_{\text{anis}} = E_{\text{magnetocrystalline}} + E_{\text{shape}} + E_{\text{stress}} + E_{\text{surface}}. \quad (5)$$

Magnetocrystalline anisotropy originates from the SOC interaction between electron spins and the crystal electric field, leading to

preferred magnetization directions (easy axes) along certain crystallographic orientations. The energy can be expressed as a series expansion in the direction cosines α_i of the magnetization with respect to crystal axes. For a cubic crystal:

$$E_{\text{MCA}} = K_1(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2) + K_2\alpha_1^2\alpha_2^2\alpha_3^2, \quad (6)$$

where K_1 and K_2 are anisotropy constants determined experimentally or through first-principles calculations.

SHAPE AND SURFACE ANISOTROPY

For thin films and nanostructures, the demagnetizing field leads to shape anisotropy that favors magnetization in the plane of the film. The surface or interface contribution becomes significant at reduced dimensions, where broken symmetry and strain alter the local electronic environment. These effects play a crucial role in determining switching behavior and spin-orbit torque efficiency in heterostructures.

MAGNETOELASTIC COUPLING

Strain-induced changes in magnetic anisotropy, described by magnetoelastic coupling, can modulate magnetic states dynamically. This effect forms the basis for straintronics and magnetoelectric coupling, where electric fields or lattice distortions control magnetization without direct current flow.

Overall, understanding the balance between exchange interactions and anisotropy energies is central to describing spin textures, domain formation, and magnetization dynamics in low-dimensional systems. These principles provide the theoretical basis for interpreting spin-orbit-driven phenomena such as SOT and the spin Hall effect (SHE) discussed in subsequent sections.

MAGNETORESISTANCE (MR)

MR refers to the change in a material's electrical resistivity in response to an applied magnetic field. It arises from the coupling between charge carrier motion and the magnetic structure or spin configuration of the material. In simple metals, MR originates from the Lorentz force deflecting charge carriers, leading to a classical quadratic dependence on field. In magnetic materials, however, spin-dependent scattering plays a dominant role, where electrons with spins parallel or antiparallel to the local magnetization experience different scattering probabilities. This gives rise to effects such as anisotropic magnetoresistance (AMR), giant magnetoresistance (GMR), and tunneling magnetoresistance (TMR). This phenomenon is the basis for various magnetic sensors, including those used in

hard drives, electronic compasses, and current sensors, which exploit the change in resistance to detect and measure magnetic fields. It is measured as

$$\text{MR} = \frac{\rho(B) - \rho(0)}{\rho(0)} \times 100\% \quad (7)$$

where $\rho(B)$ is the resistivity in an applied magnetic field B and $\rho(0)$ is the resistivity at zero field ($B = 0$).

MR measurements are widely used to probe electronic band structure, spin polarization, and scattering mechanisms. The sign, magnitude, and field dependence of MR can reveal information about carrier mobility, magnetic ordering, and quantum transport phenomena such as weak localization or Shubnikov-de Haas oscillations. In spintronic devices, MR serves as a key readout mechanism, directly linking electrical transport to spin configuration and magnetic switching behavior.

ANISOTROPIC MAGNETORESISTANCE (AMR)

The AMR effect describes how the resistivity of a ferromagnetic material changes with the angle between the current and magnetization due to SOC. It arises from the scattering probability being affected by the rotating magnetic field. The magnitude of the total AMR effect can be quantified as the ratio

$$\frac{\Delta\rho}{\rho} = \frac{\rho_{||} - \rho_{\perp}}{\rho_{\perp}}, \quad (8)$$

where $\rho_{||}$ and $\rho_{\perp}$ refer to the two extreme cases when the current is oriented parallel or perpendicular to the applied field, respectively. Typically, the AMR exhibits a sinusoidal dependence on the angle (ϕ) between field and current, given by

$$\rho_{\phi} = \rho_{\perp} + (\rho_{||} - \rho_{\perp}) \cos^2(\phi). \quad (9)$$

From this, it follows that the minima are usually located at $\phi = 0^\circ$ and $\phi = 180^\circ$ while the maxima occur at $\phi = \pm 90^\circ$.

Spin-Orbit Phenomena

SPIN-ORBIT COUPLING (SOC)

SOC is one of the fundamental interactions that bridges an electron's intrinsic spin with its orbital motion around the nucleus. It originates from relativistic effects when an electron moving through an electric field experiences an effective magnetic field in its rest

frame. The magnetic field interacts with the electron's magnetic moment, giving rise to an energy term of the form

$$H_{\text{SOC}} = \lambda \mathbf{L} \cdot \mathbf{S}, \quad (10)$$

where λ is the strength of the spin-orbit interaction, and $\mathbf{L}$ and $\mathbf{S}$ are the orbital and spin angular momentum operators, respectively.

In solids, SOC plays a significant role in determining the electronic band structure, especially in materials containing heavy-metals (HM) where relativistic effects are more pronounced. SOC lifts the spin degeneracy in momentum space and leads to spin-splitting in the band, even in the absence of an external magnetic field. Such spin-momentum locking supports many emergent phenomena in condensed matter physics including the quantum spin Hall effect (QSHE), Rashba effects, and topological insulators.

For ferromagnetic systems like CrO_2 , SOC couples the magnetic and crystalline anisotropies, defining the easy and hard magnetization axes. This influences the magnetocrystalline anisotropy energy. In non-magnetic HMs such as Pt, Ta, and β -W, SOC enables efficient conversion between charge and spin currents through mechanisms like the SHE. In hybrid heterostructures that combine ferromagnets (FMs) with HMs, SOC gives rise to SOTs which enables the direct electrical manipulation of the magnetization. Understanding SOC is critical for interpreting both the spin transport phenomena and the magnetization dynamics explored in this thesis.

SPIN-ORBIT TORQUE (SOT)

SOC couples an electron's spin with its orbital motion in a crystal lattice, enabling the conversion between charge and spin angular momentum. This coupling forms the physical basis for SOT, which allows the electrical control of magnetization dynamics in adjacent magnetic layers.

When a spin current generated by the SHE or the Rashba-Edelstein effect (REE) is injected into an adjacent FM, the transferred spin angular momentum exerts a torque on the local magnetization. This phenomenon is known as SOT which enables current-driven control of magnetization without the need for an external magnetic field. SOT has two major components:

1. Field-like torque (FLT):

$$\tau_{\text{FL}} \propto \mathbf{M} \times \boldsymbol{\sigma} \quad (11)$$

which behaves like an effective magnetic field oriented along the spin polarization direction $\boldsymbol{\sigma}$. Here $\mathbf{M}$ is the unit magnetization vector.

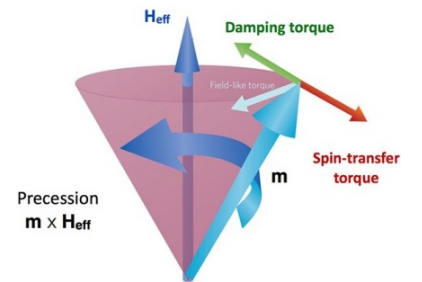


Figure 1: Schematic of field-like and damping-like torques [4].

2. Damping-like torque (DLT):

$$\tau_{DL} \propto \mathbf{M} \times (\mathbf{M} \times \sigma) \quad (12)$$

which modifies the magnetization damping and is effective in switching and domain wall motion.

The efficiency of SOT-driven switching depends on various factors including the spin Hall angle (SHA) of the HM, the transparency of the interface, and the spin diffusion length within the FM. Materials with strong SOC such as Pt and its alloys are particularly effective at generating large DLTs. This thesis leverages such materials to understand how alloy composition modulate the SOT strength and magnetization dynamics.

SPIN-TRANSFER TORQUE (STT)

Another mechanism through which spin-polarized currents can manipulate the magnetization of FM layer is spin-transfer torque (STT). Unlike SOT which arises from the SOC at interfaces, STT occurs when a spin-polarized current directly passes through a FM. The interaction between the spin current and the local magnetization results in a torque given by

$$\tau_{STT} \propto (\mathbf{J}_s \cdot \Delta) \mathbf{M}, \quad (13)$$

where $\mathbf{J}_s$ is the spin current density.

The STT mechanism enables the operation of STT-MRAM and current-induced magnetization switching in nano-pillars. Its efficiency depends on the degree of spin polarization of the current and the spin relaxation time within the FM. As such, half-metals provide an ideal platform for maximizing STT efficiency. In CrO_2 -based systems, the intrinsic half-metallicity ensures that spin angular momentum is transferred with minimal dissipation, making these materials valuable for low-power spintronic devices.

COMPARISON OF SOT AND STT DEVICES

While STT devices rely on a spin-polarized current passing directly through a magnetic layer, SOT devices achieve magnetization switching through the transverse spin current generated in an adjacent nonmagnetic HM with a large SHA. This distinction provides several practical advantages. First, because SOT devices only require a nonmagnetic layer with strong SOC to generate the spin-polarized current, they eliminate the need for a fixed magnetic reference layer, thereby simplifying device design and improving material flexibility. Second, the SOT geometry enables independent read and write

paths, typically realized in two-terminal configurations, which reduces power dissipation and enhances device endurance compared to the three-terminal STT structures. Additionally, SOT switching is inherently faster, since the torque acts directly on the interfacial spins rather than relying on spin accumulation through a tunnel barrier. It further allows for field-free and deterministic switching when combined with structural asymmetry or interfacial exchange bias. Collectively, these advantages make SOT-based architectures highly promising for next-generation nonvolatile memory and neuromorphic spintronic applications.

Hall Effects

HALL EFFECT

The Hall effect originates purely from the Lorentz force, where electrons move in curved paths described by $\vec{F} = \vec{v} \times \vec{B}$ in the presence of an external magnetic field. This creates a charge imbalance accumulating at the edges of the sample that results in a voltage difference along the transverse direction.

SPIN HALL EFFECT (SHE)

The SHE describes the generation of a transverse spin current in the form a longitudinal charge current in materials with strong SOC. When an electrical field is applied, electrons with opposite spins experience opposing transverse forces due to spin-dependent scattering or band-structure effects. This leads to spin accumulation at the sample's edges with no measurable potential difference. The resulting spin current can be quantified by the spin Hall angle (SHA):

$$\theta_{\text{SH}} = \frac{J_s}{J_c}, \quad (14)$$

where J_s and J_c denote the spin and charge current densities, respectively. The sign of the SHA denotes the polarization direction of the spin current. There are two primary microscopic origins of the SHE...

Intrinsic Berry-Phase Mechanism

The intrinsic contribution to the SHE originates from the geometric properties of electronic wavefunction encoded in the Berry curvature, which acts as a momentum-dependent magnetic field deflecting spins of opposite orientations in opposing directions. When an external electric field is applied, electrons acquire an anomalous transverse

velocity proportional to the Berry curvature of the occupied states. In systems with strong SOC, this curvature becomes spin-dependent, leading to opposite deflections for spin-up and spin-down electrons even in the absence of impurity scattering. This gives rise to a pure transverse spin current without net charge flow. The Berry curvature, defined as

$$\Omega_n(\mathbf{k}) = i \langle \nabla_{\mathbf{k}} u_{n\mathbf{k}} | \times | \nabla_{\mathbf{k}} u_{n\mathbf{k}} \rangle, \quad (15)$$

with $u_{n\mathbf{k}}$ representing the periodic Bloch functions, describes how the phase of an electron wavefunction evolves in reciprocal space. In materials with avoided band crossings or topologically nontrivial band structures such as heavy-element alloys or correlated oxides, the Berry curvature can be sharply peaked near regions of strong spin-orbit hybridization. This results in a large intrinsic spin Hall conductivity that is a direct manifestation of the band topology. Because it depends only on the geometric structure of the electronic bands, the intrinsic contribution is robust against moderate disorder and is therefore a fundamental property of clean, crystalline systems where coherence is preserved. This geometric origin provides a powerful framework for understanding and predicting large SHAs in complex materials such as Ru–Pt alloys and transition-metal oxides.

Extrinsic Scattering Mechanisms

In contrast to the intrinsic Berry-phase mechanism, the extrinsic contribution to the SHE originates from spin-dependent scattering off of impurities and defects in the crystal lattice. These processes are typically classified as skew scattering, where SOC introduced as asymmetry in the scattering cross section, and side-jump scattering, where electrons experience a transverse displacement during scattering events. The total spin Hall conductivity is therefore a sum of intrinsic and extrinsic contributions, reflecting both the underlying band topology and the impurity landscape of the material.

Materials such as Pt, Ta and β -W exhibit large SHAs, often referred to as giant SHE materials, due to strong SOC and high scattering rates. This promotes them as promising candidates for spin current sources. In this work, the Ru–Pt binary alloy is explored as a model platform for probing the interplay between intrinsic and extrinsic spin-charge conversion mechanisms. Its tunable composition and structural phase evolution offer control over both electronic coherence and scattering environments, while maintaining excellent chemical stability and compatibility with FM interfaces. Understanding these competing mechanisms provides valuable insights into the

design of efficient spin-charge conversion materials for developing robust, scalable spintronic materials for next-generation devices.

INVERSE SPIN HALL EFFECT (ISHE)

The ISHE is the reverse of the SHE in that it is a spin-to-charge conversion phenomenon in which a spin current flowing through a non-magnetic material with strong SOC generates a transverse charge current. Microscopically, the ISHE arises because electrons with opposite spins are deflected in opposite directions due to spin-dependent scattering or intrinsic spin-orbit interactions, producing a measurable voltage perpendicular to both the spin polarization and the spin current. Like the SHE, ISHE is also widely used to detect spin currents and quantify the SHA of materials, making it a cornerstone in the characterization of spintronic devices and SOTs.

ANOMALOUS HALL EFFECT (AHE)

The AHE is observed in FMs due to the imbalance of spin up and spin down states. This imbalance manifests as a transverse Hall voltage (R_H) independent of an applied field. This effect can be used to probe the out-of-plane component of the magnetization vector: $R_H \propto M_z$. Unlike the ordinary Hall effect, which arises purely from the Lorentz force on charge carriers, the AHE is dominated by spin-dependent mechanisms, including intrinsic contributions from the Berry curvature of electronic bands and extrinsic contributions from skew scattering and side-jump processes.

Scattering Mechanisms

The transport properties of magnetic conductors are strongly influenced by different kinds of scattering processes that determine the charge carrier relaxation time, mean free path, and ultimately the electrical resistivity. When an electric field drives electrons through a solid, their momentum is continuously randomized by collisions with imperfections or excitations in the lattice. Charge carrier scattering arises from several distinct microscopic mechanisms, each exhibiting a characteristic temperature dependence. The total resistivity can be expressed as

$$\rho(T) = \rho_0 + \rho_{\text{ph}}(T) + \rho_{\text{mag}}(T) + \rho_{\text{anom}}(T), \quad (16)$$

where ρ_0 represents the residual resistivity due to static disorder, ρ_{ph} the phonon contribution, ρ_{mag} the magnon (spin-wave) contribution, and ρ_{anom} terms arising from spin-orbit-coupled scattering, which are particularly relevant to the AHE and SOT.

These scattering events can be classified into three main types:

- **Impurity (Elastic) Scattering:** This type of scattering occurs from defects, vacancies, grain boundaries, or impurity atoms in the crystal lattice. Since these centers are static, the energy of the electron is conserved during scattering, but its momentum changes direction. This gives rise to a temperature-independent term ρ_0 , which sets the baseline resistivity. Impurity scattering dominates at low temperatures where phonon excitations are frozen out.
- **Phonon (Inelastic) Scattering:** At elevated temperatures, electron-phonon interactions become significant. Phonons act as perturbations of the lattice potential, enabling inelastic processes where electrons exchange energy with lattice vibrations. The relaxation time thus decreases with increasing temperature, leading to a resistivity that scales approximately as $\rho \propto T^5$ in the low-temperature regime and linearly as $\rho \propto T$ at higher temperatures.
- **Magnetic (Spin-Dependent) Scattering:** In FMs and other magnetically-ordered systems, the exchange interaction between conduction electrons and localized magnetic moments introduces an additional scattering channel. Spin-flip scattering occurs when an electron reverses its spin due to interactions with spin disorder, magnons, or domain walls. The resulting resistivity contribution depends strongly on the spin polarization and the degree of magnetic order, often giving rise to phenomena such as AMR and GMR.
- **Spin-Orbit Coupled Scattering:** SOC introduces additional asymmetries in the scattering of spin-polarized electrons, leading to two key mechanisms that contribute to both the longitudinal and transverse (Hall) resistivities:
 - **Skew scattering:** This mechanism arises from asymmetric scattering of spin-up and spin-down electrons by impurities in the presence of spin-orbit interaction. The resulting resistivity contribution scales linearly with the longitudinal resistivity, i.e.,

$$\rho_{\text{skew}} \propto \rho_{xx} \propto T, \quad (17)$$

reflecting its extrinsic origin tied to the impurity concentration and the temperature-dependent scattering rate. The chiral features of disorder scattering make are responsible for this sort of scattering, where the transition probability depends on whether it is left-handed or right-handed with respect to the magnetization direction.

- **Side-jump scattering:** This effect originates from a lateral displacement of the electron trajectory during each scattering event, caused by the spin-orbit-induced anomalous velocity. More specifically, it is due to the displacement of the wave vector when the wave packet is scattering by an impurity. The side-jump contribution scales quadratically with the longitudinal resistivity,

$$\rho_{\text{sj}} \propto \rho_{xx}^2 \propto T^2, \quad (18)$$

and is generally less sensitive to impurity concentration than skew scattering.

- **Magnon Scattering in Half-Metallic Ferromagnets:** In conventional metallic FMs, both spin channels are partially occupied at the Fermi level. At finite temperatures, conduction electrons can scatter by emitting or absorbing magnons (spin-wave excitations). The simplest process is one-magnon scattering, which flips the electron's spin and changes its momentum. The magnon population scales with temperature as $n_m(T) \propto T^{3/2}$, following Bose-Einstein statistics for low-energy magnons. Combining this with the available electronic phase space leads to a resistivity contribution that scales approximately as

$$\rho_{1\text{-mag}} \propto T^2, \quad (19)$$

in the low-temperature limit.

In contrast, half-metallic FMs exhibit metallic behavior in only one spin channel, while the other spin channel possesses a gap at the Fermi energy, such that E_F lies within a band gap for one spin channel. This electronic structure prohibits the typical one-magnon spin-flip processes, since transitions into the gapped minority spin band are energetically forbidden at low temperatures.

As a result, two-magnon scattering becomes the dominant mechanism for spin-dependent resistivity in half-metals. In this higher-order process, an electron scatters while simultaneously creating/annihilating two magnons, thereby conserving the total spin without requiring a spin-flip transition. Although such scattering events are much weaker, they exhibit a stronger temperature dependence due to the quadratic dependence on magnon population.

The temperature dependence of two-magnon scattering can be derived by considering the magnon density of states $D(\epsilon) \propto \sqrt{\epsilon}$, together with the quadratic magnon dispersion $\epsilon_k = Dk^2$. Integration over the joint two-magnon density of states yields a resistivity

contribution of the form

$$\rho_{2\text{-mag}} \propto T^{4.5}. \quad (20)$$

This $T^{4.5}$ dependence is the defining feature of half-metallicity, reflecting the suppression of single-magnon spin-flip scattering due to the minority spin gap. Double-magnon scattering is confirmed by the results pursued in this thesis.

Mechanism	Magnetic System	Process Type	Resistivity Dependence
1-magnon scattering	Metallic ferromagnet	Spin-flip	T^2
2-magnon scattering	Half-metal	Spin-conserving	$T^{4.5}$
Phonon scattering	Any metal	Lattice vibrations	T^5 (low T)
Impurity scattering	Any metal	Elastic (defects)	T^0 (constant)

Table 1: Summary of dominant scattering mechanisms and their characteristic temperature dependence in magnetic conductors.

Spin-Dependent Conduction

One of the most important requirements for efficient spintronic operation is the ability to generate, manipulate, and detect spin currents. Spin currents can arise from (a) spin-polarized charge currents flowing through a FM, (b) spin pumping from precessing magnetization, or (c) SOC phenomena such as the SHE and REE.

In thin-film heterostructures, such as HM/FM bilayers, spin currents generated by the SHE in the HM (i.e. Pt, Ta, or Ru-Pt alloys) can diffuse into the FM layer and exert torques on its magnetization. Materials with a large SHA are promising candidates for energy-efficient spintronic devices, including magnetic memories, spin-orbit oscillators, and logic elements.

The spin Hall conductance (SHC) therefore has multiple contributions from the side-jump and skew scattering as well as the intrinsic mechanisms:

$$\sigma_{xy}^H = \sigma_{xy}^{int} + \sigma_{xy}^{sj} + \sigma_{xy}^{skew}. \quad (21)$$

In the ideal case of a single, perfect crystal the intrinsic mechanism is the dominant contribution to the SHE. Materials with little scattering contributions are advantageous to spintronic applications as they correspond to fewer impurities and lower resistivities.

The field of spintronics has evolved from early discoveries of spin-polarized transport in metals and semiconductors to the exploration of complex materials with novel spin textures such as half-metals,

topological magnets, and altermagnets (AM). These systems propose enhanced spin polarization and intriguing mechanisms for generating and controlling spin currents. Understanding the fundamental physics governing SOC and spin-momentum locking is crucial for interpreting experimental observations and for designing next-generation spintronic architectures.

Spin Polarization

A central concept in modern spintronics is the control of spin polarization in electronic transport. In most conventional metals, both spin-up and spin-down electrons contribute to conduction which results in only a partial spin polarization. The spin polarization, p , can be quantified as the asymmetry of spin-up and spin-down states in a FM:

$$P = \frac{n_{\uparrow} - n_{\downarrow}}{n_{\uparrow} + n_{\downarrow}} \quad (22)$$

where $n_{\uparrow}$ and $n_{\downarrow}$ are the density of spin-up and spin-down states at the Fermi level, respectively. In contrast, half-metallic ferromagnets (HMF) exhibit a unique band structure where one spin channel behaves metallically, contributing to all conduction properties, while the other is insulating or semiconducting at the Fermi level [5]. This leads P to approach 100% in HMFs, making them ideal candidates for spin-injection and spin filtering applications. As a result, HMFs serve as nearly ideal sources of spin-polarized current for STT and SOT experiments.

Spin polarization has several key implications for spintronic transport:

- It ensures that only one spin species contributes to the charge current. This effectively eliminates spin-flip scattering and allows for efficient spin injection into adjacent NM or HM layers.
- The strong exchange splitting in HMFs stabilizes long-range FM order, leading to a high T_C and reduced thermal depolarization.

Despite their advantages, realizing and maintaining half-metallicity in the low-dimensional limit of thin films poses several challenges. Structural imperfections, interfacial disorder, and chemical non-stoichiometry can introduce mid-gap states that partially depolarize the spin current. Additionally, interfacial hybridization between HMFs and HMs can modify the density of states (DOS) at the Fermi level. In some cases, this can suppress full spin polarization. These effects also make the synthesis of high-quality samples, successfully

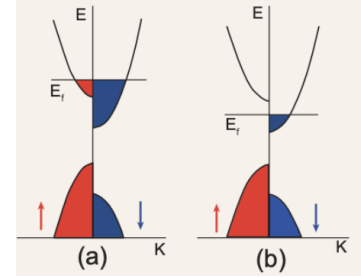


Figure 2: Schematic of (a) ferromagnetic and (b) half-metallic band diagrams [5].

achieved in this work, a critical step toward accurately probing intrinsic spin transport phenomena.

In addition to their technological relevance, HMFs provide an excellent platform for studying fundamental spin-dependent transport phenomena such as Shubnikov-de Haas (SdH) oscillations, quantum interference effects, and AMR. Because only one spin channel participates in conduction, any observed oscillatory or magnetoresistive behavior can be directly attributed to the spin-polarized band structure and its interaction with external magnetic fields.

THE CASE OF CrO_2

The concept of half-metallicity was discovered in 1982 while studying the conduction properties of NiMnSb , a Heusler alloy. Since then, various materials including perovskite oxides, sulfides, and zinc-blende structure compounds have been explored for their potential half-metallic behavior [6]. Among these, chromium dioxide (CrO_2) stands out as one of the few experimentally verified HMFs with robust, near-perfect spin polarization persisting up to room temperature [7].

Band structure calculations and photoemission experiments confirm the majority-spin band crossing at the Fermi level while the minority-spin band exhibits a clear gap [9, 10]. The DOS for majority and minority spin states is shown [8]. The d band of the minority spin state shifts due to exchange splitting such that the Fermi level lies between the oxygen p states and the chromium d states.

Spin polarization in HMFs is, however, temperature-dependent. At low temperatures, $P \approx 1$, but at finite temperatures it can decrease due to thermal fluctuations which mix spin-up and spin-down states at the Fermi level. This gradual depolarization of CrO_2 can be measured by the drop in net magnetization at elevated temperatures. As shown in this work, the magnetization of bulk CrO_2 films decreases notably from 100% at 300 K to $\sim 70\%$ at 2 K.

In addition to its unity spin polarization, CrO_2 has several desirable properties including a high Curie temperature of $T_C \approx 390$ K, large magnetic moment of $2 \mu_B$ at 10 K [11], and a rutile crystal structure. CrO_2 is also a metastable FM which can reduce to Cr_2O_3 in oxygen-poor environments and becomes an insulating antiferromagnet (AFM) [12].

In this context, CrO_2 offers a clean and well-defined system for exploring quantum transport in the half-metallic limit, serving as a benchmark for theoretical models and a foundation for investigating more complex spin textures in emerging materials like AMs and AFMs.

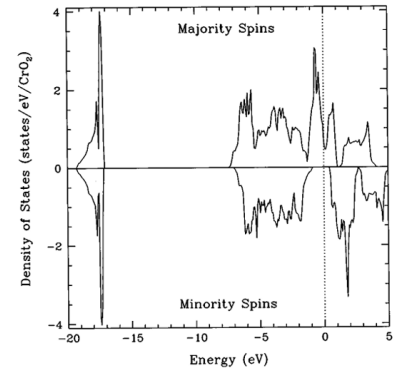


Figure 3: Calculated density of states for CrO_2 . Dashed line indicates Fermi level [8].

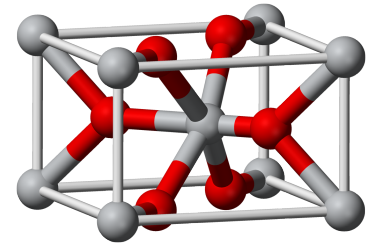


Figure 4: Schematic of rutile structure. CrO_2 forms a body-centered tetragonal lattice surrounded by octahedra of oxygen atoms.

Quantum Transport in Metals

LANDAU QUANTIZATION

When a magnetic field is applied to a conducting material, the motion of charge carriers becomes quantized due to the Lorentz force which confines their trajectories into circular cyclotron orbits. The quantization of these orbits gives rise to discrete energy levels known as Landau levels (LLs). For a free electron in a uniform magnetic field $\mathbf{B}$ applied along the z -axis, the energy spectrum is given by

$$E_n = \left(n + \frac{1}{2}\right) \hbar \omega_c + \frac{\hbar^2 k_z^2}{2m^*}, \quad (23)$$

where $n = 0, 1, 2, \dots$ is the Landau level index, $\omega_c = \frac{eB}{m^*}$ is the cyclotron frequency, m^* is the effective mass of the carrier, and k_z is the wave vector along the direction of the magnetic field.

Each LL represents a highly degenerate set of states corresponding to electrons with quantized transverse motion but free motion along $\mathbf{B}$. When the magnetic field is sufficiently strong or the temperature sufficiently low, the discrete nature of these levels becomes experimentally observable.

At the Fermi level, oscillations in the DOS occur whenever a LL passes through the Fermi energy. These oscillations manifest as periodic modulations in a variety of measurable quantities such as resistivity, magnetization, and heat capacity, thereby leading to quantum oscillations.

SHUBNIKOV-DE HAAS OSCILLATIONS

The Shubnikov-de Haas (SdH) effect refers to the quantum oscillations in electrical resistivity observed under high magnetic fields and low temperatures. It arises from the periodic variation of the electronic DOS at the Fermi level due to Landau quantization. When a LL crosses E_F , the number of available conduction states changes, modulating the scattering rate and thus the longitudinal resistivity (ρ_{xx}).

Experimentally, the SdH oscillations are periodic in the inverse magnetic field ($1/B$) rather than B itself. The background oscillatory component of the resistivity can be expressed as

$$\Delta\rho_{xx} \propto \cos \left[2\pi \left(\frac{F}{B} + \gamma \right) \right], \quad (24)$$

where F is the oscillation frequency (in Tesla) and γ is a phase factor containing information about the Berry phase and Fermi surface topology. The frequency F is directly related to the extremal cross-sectional area A_F of the Fermi surface perpendicular to the magnetic

field through the Onsager relation:

$$F = \frac{\hbar}{2\pi e} A_F. \quad (25)$$

This relation forms the basis for mapping the geometry of the Fermi surface from SdH measurements. In materials with simple, isotropic Fermi surfaces, A_F corresponds to a circular cross-section. In contrast, anisotropic or multi-band systems such as CrO_2 produce multiple frequencies corresponding to different extremal orbits.

LIFSHITZ-KOSEVICH FORMALISM

The amplitude of SdH oscillations carries rich information about the quasiparticle dynamics and scattering mechanisms. The temperature and field dependence of the oscillation amplitude is described by the Lifshitz-Kosevich (LK) formalism, which incorporates both thermal damping and impurity scattering:

$$\Delta\rho_{xx} \propto R_T R_D \cos \left[2\pi \left(\frac{F}{B} + \gamma \right) \right], \quad (26)$$

where

$$R_T = \frac{\alpha T m^*/B}{\sinh(\alpha T m^*/B)} \quad \text{and} \quad R_D = \exp \left(-\frac{\alpha T_D m^*}{B} \right) \quad (27)$$

are the temperature and Dingle damping factors, respectively, with $\alpha = 2\pi^2 k_B m_0 / e\hbar$. Here, T_D is the Dingle temperature related to the carrier scattering time τ through

$$\tau = \frac{\hbar}{2\pi k_B T_D}. \quad (28)$$

At cryogenic temperatures, the dominant scattering mechanism transitions from phonon to impurity and magnetic scattering, leading to long carrier lifetimes and mean free paths sufficient to resolve Landau quantization and SdH oscillations. The T_D extracted from the LK analysis quantifies the degree of disorder-induced broadening of LLs, directly related to the total scattering rate. A lower T_D therefore indicates a longer relaxation time and higher sample purity, both of which are prerequisites for observing quantum oscillatory phenomena. In systems with strong spin polarization or reduced dimensionality such as half-metals, magnetic heterostructures, or two-dimensional electron gases, the interplay of these scattering mechanisms determines not only the resistivity but also the efficiency of spin-charge conversion and the stability of spin textures.

By fitting experimental data to the LK formula, one can extract fundamental material parameters such as:

- Effective mass m^* of charge carriers,
- Dingle temperature T_D and corresponding scattering time τ ,
- Fermi wave vector k_F , velocity v_F , and carrier density n ,
- Berry phase ϕ_B , which distinguishes between trivial and topologically nontrivial band structures.

Overall, Landau quantization and SdH oscillations provide a sensitive window into the interplay between spin, charge, and topology in correlated electron systems, making them central to understanding magnetotransport in low-dimensional and spintronic materials.

Emerging Concepts

ALTERMAGNETISM

Altermagnets (AMs) are characterized by a compensated magnetic structure with zero net magnetization, similar to AFMs, yet exhibit a symmetry-protected spin splitting of electronic bands comparable to that of FMs. This phenomenon arises from non-relativistic exchange interactions combined with specific crystalline symmetries that break spin degeneracy without requiring SOC. In contrast to traditional AFMs, where opposing spins belong to inversion-related sublattices, AMs possess magnetic sublattices connected by rotation or mirror symmetries that preserve overall time-reversal breaking while maintaining zero total moment. The result is a large spin polarization in momentum space despite a vanishing macroscopic magnetization, enabling efficient spin-charge conversion and magnetoresistive effects without stray fields. This unique symmetry-derived property positions AMs as promising candidates for next-generation spintronic devices, bridging the gap between ferromagnetic and antiferromagnetic functionalities [13].

Chapter 2 | Materials & Methods

TO RESOLVE the magnetization dynamics of spins in the various systems studied in this work, a fundamental understanding of the physics and mechanisms behind different fabrication and characterization techniques is crucial. A discussion of these techniques is included below to guide the reader in gauging the significance of the resultant research projects.

Sample Fabrication

This section highlights the basic methodologies used for fabricating the samples studied in this work.

WAFER PREPARATION

Silicon wafers are the predominant substrate used for performance device fabrication. Thermal oxidation of silicon produces an extremely high quality Si/SiO₂ interface with very low defect density. This passivation layer stabilizes the electrical properties of the silicon surface and protects the device from environmental damage and contamination. Therefore, bare silicon wafers are baked in an oxygen-rich environment to facilitate the growth of this desirable native oxide.

Procedure to bake bare Si wafers in dry oxidation furnace:

1. Obtain a 25-pack of 2-inch P/B (100) single-side polished (SSP) prime Si wafers from WaferPro LLC with 1-10 Ω -cm resistivity and $279 \pm 25 \mu\text{m}$ thickness.
2. Turn on the breaker and throttle switch located in the chase at the back of the furnace.
3. Open the Ni bottle and valve to purge the tube.
4. Turn on the top tube furnace and set the temperature to 1050°C.
5. In the meantime, arrange the wafers into the slots of the quartz carriers such as to maximize the exposure of the polished side.

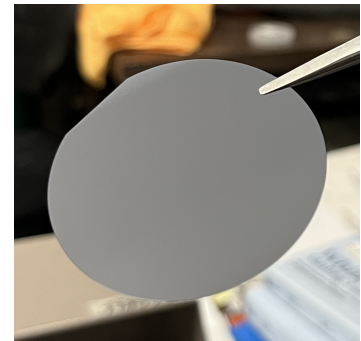


Figure 5: Image of bare Si wafer.

6. Once the temperature is stabilized, load the wafer carriers carefully into the CENTER of the tube using a quartz stick.
7. Close the Ni valve and open the oxygen valve to flow research-grade O_2 with a flow rate of 0.5 sccm/sec. Check that the oxygen flow rate in Channel 2 reads 1.5.
8. Let the wafers bake for approximately 48 hours or until the color changes from gray to teal.

This is a very standard, repeatable, and cost-effective procedure which allows for precise control over the oxide's thickness and quality and is preferred to purchasing oxidized wafers directly.

Magnetron Sputtering

Magnetron sputtering is a physical vapor deposition technique used to deposit sub-micron thin films on silicon wafers. It is a room-temperature deposition technique which requires a chamber capable of reaching ultra-high vacuum ($\sim 10^{-8}$ Torr) to ensure a pristine deposition environment. A cryopump held at ~ 15 K removes gaseous impurities such as adsorbed water vapor and He atoms in the system by use of activated charcoal. A rotary chain allows the silicon wafer substrate holder to rotate from above from one target gun to another, allowing us to build multilayer stacks of various materials.

Standard approach for depositing thin-film stacks using sputtering systems:

- Check integrity of the shutters for all guns. Check targets are sufficiently thick. Clean shutter undersides if build-up visible. Blow targets with Ni gun to remove flakes and particles.
- Load up to 5 Si wafers onto the substrate holders (positions 3, 6, 8, 10, 12). Blow with Ni gun.
- Close the chamber, pump down with mechanical pump and open gate valve. Keep baffle at 100% OPEN.
- Turn on the heater tape and bake the system at 80°C for at least one day, pumping for an additional 2-3 days until base pressure $\sim 10^{-8}$ Torr.
- OPEN Ar bottle, turn ON chiller for guns, ensure all power supplies are ON. Set baffle value to 30% OPEN.
- As per calibration, set Ar pressure to 4 mTorr for all targets except for MgO which should be set to 1.6 mTorr.

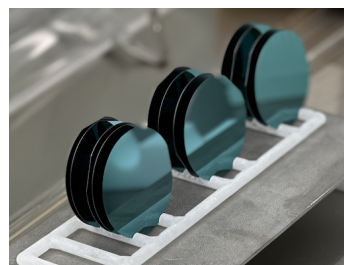


Figure 6: Image of oxidized Si wafer.

For best results, use wafers within 1-2 months of oxidation. This prevents surface degradation, poor adhesion from reduced surface energy, and increased surface roughness from airborne particles.

The flow rate of Ar can be manually adjusted to account for slight deviations of the baratron gauge. The baratron should be zeroed to ensure an accurate reading prior to deposition.

In the case of plasma ignition trouble: increase Ar pressure, increase dc output power, check that copper ring is tight and in good contact with target, if gun is dc try to light with rf.

- With global rotation ON, pre-sputter targets of interest for 5 min (new targets should be pre-sputtered for at least 2 hours.) This step ensures that any gettered impurities are removed prior to deposition. Ensure that plasma ignites (soft purple plume).
- Type in desired recipe for multilayer stack and appropriate target.
- Once deposition finishes, turn OFF Ar valve, OPEN baffle back to 100%, turn OFF chiller. Let system stabilize for at least 1 hour before removing wafers.
- As per sample documentation, label each wafer with initials, sample run number, substrate index, and date.

The efficacy of magnetron sputtering relies on the physical bombardment of energetic Ar ions with the target material atoms due to a voltage difference [14]. A sputtering source (the target) is charged with a negative voltage while a substrate (Si wafer) holder facing the target is kept at ground. The large potential difference between the target and substrate ionizes the Ar atoms by accelerating free electrons which collide with Ar atoms. This, in addition to secondary electron emission, creates a self-sustaining plasma in which the Ar ions are accelerated toward the negatively charged target to sputter atoms off its surface. These free atoms finally condense on the substrate and forms thin films.

An electrically grounded aluminum shield with an opening at the center is placed on top of the target, encasing the copper ring holding down the target. The shield maintains a small gap with the target's surface, enabling ionization to happen at a much lower voltage.

A strong permanent magnet (~ 1.5 kG) is placed below the target to generate a magnetic field parallel to the substrate surface. The Lorentz force causes secondary electrons to spiral, which significantly increases their chances of collision. The magnetic field of the gun pulls electrons away, bending their paths and confining the electron density near the target. Consequently, lower sputtering pressure and power are required to sustain the plasma. Typically, dc sputtering is used for metallic and magnetic targets and rf sputtering for insulating targets, like MgO, since rf fields alternately charge and discharge the surface.

The sputtering yield, defined as the number of ejected atoms per incident Ar ion, depends on target atom mass and ion energy. For a target made up of multiple elements (i.e. CoFeB), elements with higher sputter yield will be ejected more than others. This changes the elemental composition at the target surface. Eventually a steady

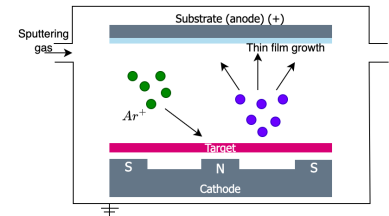


Figure 7: Schematic of magnetron sputtering illustrating bombardment of Ar ions with target atoms.

It is recommended that routine maintenance and deposition rate calibrations are performed to ensure high quality and reproducibility of samples.



Figure 8: Magnetron sputtering system used for sample fabrication. Important components are labeled.

state is reached where the stoichiometry of ejected atoms equals that of the target. To ensure uniformity, substantial pre-sputtering is needed.

During deposition, the silicon wafers are self-rotating and off-centered relative to the gun. This allows for a more uniform film with fewer impurities. With this system, films as thin as 1 nm can be deposited with angstrom-level precision. A custom Python GUI was created for user system control and operation to reliably automate the sputtering process and minimize user error across batches. This GUI may also be used manually for troubleshooting and maintenance procedures.

Chemical Vapor Deposition (CVD)

Chemical vapor deposition is used when depositing thin films that must have high epitaxial integrity. This technique requires a precursor material (typically in powder or pellet form), a carrier gas, and a single-crystal substrate which determines the orientation of the film. When the precursor is heated above a critical temperature, it gains a significant vapor pressure and can evaporate. It is then transported by the carrier gas and deposited on the substrate.

We employ a two-zone tube furnace to independently control the precursor temperature (239°C) and the single-crystal TiO_2 substrate temperature (370°C). Deposition is carried out in an oxygen (O_2)-rich atmosphere to support a stable deposition rate and good film quality.

WARNING: Practice extreme caution when handling the precursor. Wear all necessary PPE (lab coat, respirator, double-layered gloves: nitrile + green). Use designated spatula to scoop precursor into boat. Do NOT dump excess precursor into the sink, instead dissolve in chromium etchant and pour into chemical waste bottle.



Figure 9: Chemical vapor deposition system used for epitaxial sample fabrication. Important components are labeled. O_2 bottle not pictured.

1. Ensure all components are cleaned with chromium etchant/DI solution (100mL:300mL) and subsequently rinsed with methanol, DI water. Of the 400mL etchant solution, use 200mL for cleaning the boat and substrate holder in a wide-mouth beaker and the remaining 200mL to clean the tube.
2. Pretreat the 25 mm^2 single-crystal TiO_2 substrates with acetone (5 min), IPA (5 min), and diluted hydrofluoric acid (20 min) to create a clean, activated surface for film growth. Use within 48 hours for reliable deposition.

3. Load the substrates on the sample carrier at the 370°C end, push to center of furnace, and seal the tube on both sides.
4. Turn on both heaters as well as the Omega heater (set to 240°C located behind Heater 1. Start the flow of 100 sccm of O₂. Wait $\approx$ 1 hour until furnace temperatures stabilize. The TiO₂ substrates should turn yellow-orange in color.
5. Carefully fill the hexavalent chromium precursor into circular boat. Open the 239°C end of the tube, load the boat and push into center of furnace. Seal the tube.
6. Time the deposition for desired film thickness. (Approximately linear following an initial activation time $\approx$ 10-12 min).
7. Turn off O₂ flow and all 3 heaters. Open the furnace hoods and let system cool to room temperature before removing residual precursor.
8. Reseal the tube, turn on all 3 heaters and anneal for additional 1 hour at 370°C under 100 sccm O₂ flow.

Check thickness of films prior to patterning using VSM (use the lowest temperature to get the most accurate reading of magnetic moment – see Bloch's law.

Steps 7 and 8 are crucial for high-quality samples. After deposition, the films are heated again. This step improves the film by stabilizing CrO₂, which is metastable and tends to reduce to Cr₂O₃ or CrO₃. The oxygen anneal preserves the Cr⁴⁺ oxidation state, limits the formation of non-stoichiometric defects, and enhances epitaxial crystallinity by reducing structural imperfections.

Photolithography

Once the thin films have been deposited, they are patterned into devices. This may entail a single patterning step or multiple, depending on the complexity of the device and the subsequent characterization technique(s) in mind. The Heidelberg Instruments MLA 150 Maskless Aligner is used to pattern discrete features as small as 2 μ m. Maskless patterning involves a 375 nm laser write head which rasters across the entirety of the wafer, denaturing the photoresist (PR) polymer only where exposed. It follows that a more complex pattern spanning a larger area of the wafer will take longer to pattern.

1. Coat the wafer evenly with spin-on PR: AZ 5214-E (kept at 6°C) using a clean dropper (do not use PR at bottom of bottle).
2. For $\approx$ 1.2 μ m thick PR, set spin-coater speed to 4,000 rpm for 30 seconds.
3. Bake the wafer on a hot plate for 2 minutes at 108°C.

Due to the viscoelasticity of the PR, the edge of the wafer will likely have thicker PR and thereby yield non-uniformity in patterning steps. Avoid placing important device features at the edges.

4. Let cool for 30 seconds and place in MLA for patterning. Make sure the flat edge of wafer is flush with the aligner and centered. The laser is sensitive to height non-uniformity so ensure the wafer is clean and there is no PR residue on backside.
5. Set MLA parameters: 375 nm laser, 200 mJ/cm² dose.
6. Upload appropriate AutoCAD design file. Check if inversion required (exposes all BUT patterned area – used for ion milling step).
7. After patterning complete, develop in undiluted AZ 917MIF until PR dissolves and reveals pattern. Development time varies based on feature size and complexity. Verify in microscope.

For the works detailed in this thesis, the MLA was used to pattern all electrical leads, VSM circles, Hall crosses, and quarter sectors for XRD/XRR. The standard Hall cross dimensions are 55 μm $\times$ 20 μm .

Etching

(1) Physical ion milling is used to define patterns on films and multilayer stacks deposited with magnetron sputtering. This type of etching involves accelerating highly-energetic Ar atoms at the wafer surface, causing bombardment and release of metal atoms. To ensure good collimation of the incident beams, a screening grid is placed after the acceleration grid, which accelerates the Ar ions to a high energy. This technique is relatively insensitive to the stack's composition and yields a rather uniform milling rate of about 15 nm/min. To avoid charge accumulation, a neutralizer filament is placed in between the ion source and substrate which emits electrons to neutralize the Ar ions. A filament is also used as the cathode of the plasma generator where hot electron emission initializes and stabilizes the plasma. Constant in-plane rotation and a 45° offset between the wafer and Ar beam ensures clean edges, sidewall cleanliness, and uniformly-milled surfaces. Strong thermal contact between the wafer and wafer plate is necessary to prevent overheating and hard-baking of PR. The substrate holder is water-cooled and can rotate from 0° to 90°.

(2) Reactive ion etching (RIE) was used specifically for the CrO₂ samples deposited using the CVD technique. RIE is a dry chemical etching technique that relies on the chemical reactivity of the precursor gases to the film composition. The plasma in RIE is generated by rf power supply between two parallel metal plates from a reactive gas or mixture of reactive and inert gas. The discharge generates highly

Signs of underdevelopment (keep developing): rainbow colors inside pattern, absence of dark outline at edge of pattern. Signs of overdevelopment (must re-pattern): curved corners, lack of sharp edges.

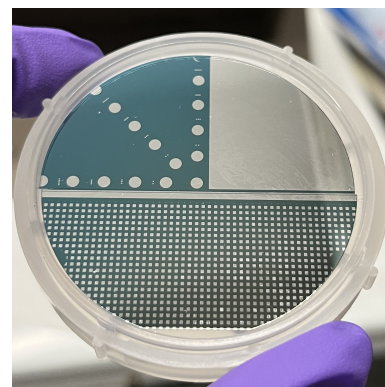


Figure 10: Image of patterned wafer after physical ion milling and photoresist wash-off. Circles used for VSM, film for XRD/XRR, Hall bars for resistivity.

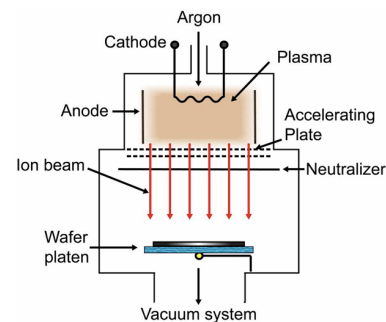


Figure 11: Schematic of physical ion milling method used to etch samples [15].

reactive radicals that chemically etch the sample, while the electric field between the metal plates also accelerates the ions to bombard the sample surface. This is a highly selective etching technique in which the precursor gas must be chosen carefully such as to etch only the film of interest. For the case of CrO_2 , the films were etched for 60 seconds using a mixture of 80 sccm CF_4 and 40 sccm O_2 gases on a PlasmaTherm Vision 320 RIE system.

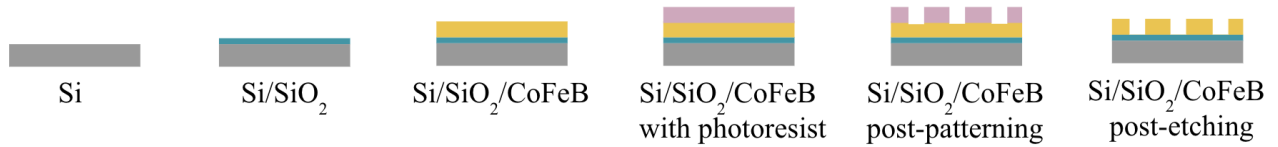


Figure 12: Schematic of sample process flow from bare Si wafer to patterned device.

Characterization

This section briefly explains the fundamental concepts and techniques used for characterizing the samples studied in this work. The experimental results of these techniques can be found in the relevant chapters.

X-ray diffraction (XRD)

XRD is a characterization technique used to determine the crystal structure and grain size of thin films. Monochromatic $\text{Cu-K}\alpha$ X-rays are scattered by the regularly spaced atoms in the crystal lattice. At specific angles, the scattered waves align perfectly, resulting in constructive interference and a strong amplified signal. These conditions are described by the Bragg condition:

$$n\lambda = 2d \sin(\theta) \quad (29)$$

where n is a positive integer, θ is the Bragg angle, $\lambda = 1.5406\text{\AA}$ is the characteristic X-ray wavelength of $\text{Cu-K}\alpha$, and d is the interplanar spacing of the lattice. A detector measures the intensity of the diffracted X-rays at various angles, producing a diffraction pattern. A Bruker D8 Discovery High Resolution X-ray Diffractometer is used to collect this information.

1. Load and secure the sample, flush to the mount in the path of X-rays.
2. Set the detector to Lynxeye oD (HiRES). Choose the slit size to be 1.2 mm. The X-rays should be set to 40 kV at 40 mA.

3. Zero all parameters and set Primary Rot Absc to 58.22.
4. Perform a Z-scan (mode: STEP, time/step: 0.1s, range: -1.3mm to 1.9mm with step size 0.01mm). Double-click the Z value at half the max counts, this will fix the optimal Z-height of the sample.
5. Perform a rocking curve (mode: CONTINUOUS, time/step: 0.1s, range: -5.0° to $+5.0^\circ$ with step size 0.01°). Double-click the center of the Gaussian, this will fix the optimal value for Ω .
6. Repeat the Z-scan.
7. Perform the 2θ - Ω scan (time/step: 0.2s). Set the detector to Lynx-eye 1D. Set the Primary Rot Absc to 1. Choose the desired range for diffraction scan (typically $20^\circ - 90^\circ$)

As a check, look for Si (004) peak from substrate at $2\theta = 69^\circ$.

X-ray Reflectometry (XRR)

XRR can be used to non-destructively extract the film thickness using the same XRD machine. It works by shining the X-rays at very shallow, or "grazing", angles. Due to X-rays having a refractive index slightly less than 1 in most materials, they undergo total external reflection at these shallow angles. A portion is reflected from the surface, while another part penetrates slightly. If the sample has multiple layers with different electron densities, X-rays reflect off each interface. These reflected beams interfere, meaning they can either reinforce (constructive interference) or cancel each other out (destructive interference). This constructive and destructive interference creates a distinctive oscillation pattern, known as interference fringes, in the measured reflectivity data.

1. Load and secure the sample, flush to the mount in the path of X-rays.
2. Set the detector to Scinti. Choose the slit size to be 0.2 mm. The X-rays should be set to 40 kV at 40 mA.
3. Zero all parameters and set Primary Rot Absc to 58.22.
4. Perform a Z-scan.
5. Perform a rocking curve.
6. Repeat the Z-scan.
7. Set θ_2 -H to 0.5° . Repeat rocking curve.

8. Perform the 2θ - Ω scan (time/step: 0.1s). Set the Primary Rot Absc to 1. Set 2θ range from 0.0 to 5.0° with step size 0.005° and time/step: 1.0s). Change counts to log-scale to resolve fringes.

A rough, back-of-the-envelope calculation can be used to estimate the thickness of the film prior to a more detailed Fourier analysis:

$$t_{\text{film}}(\text{\AA}) = \frac{88.3}{\Delta 2\theta_{\text{fringe}}} \quad (30)$$

where $\Delta 2\theta_{\text{fringe}}$ is the spacing between two consecutive peaks. XRR data can also be analyzed carefully to gauge the surface roughness at the interface between the film and substrate.

Energy Dispersive X-ray Spectroscopy (EDS)

EDS was used extensively to quantify the chemical compositions of various alloys. It is a material analysis technique that identifies elements in a sample by bombarding it with an electron beam, causing the atoms to emit characteristic X-rays at specific energies. The incident beam ejects an electron from an atom's inner electron shell, creating a "hole". An electron from a higher-energy outer shell then moves to fill this hole. The energy difference between the shells is released as an X-ray photon. The energy of these emitted X-rays is unique to the specific element and the energy level transition involved. A detector then measures the energy of these X-rays and their number and provides a spectrum of X-ray intensity and characteristic energy.

Transmission Electron Microscopy (TEM)

TEM is a powerful imaging characterization technique that uses an electron beam to study the internal structure of ultra-thin films. The beam of electrons interacts with the specimen and transmit through the sample. The image is then magnified and focused onto a fluorescent screen. Due to the small de Broglie wavelength of electrons, TEM can provide much higher resolution image than optical microscopy.

In this case, 10 nm-thick films were deposited on 25 nm-thick, 3.05 mm O.D. pure-carbon films (no Formvar) supported by 400-mesh copper grid TEM disks. This mesh size allows for a uniform film with structural integrity while allowing transmission of electrons through the film for imaging.

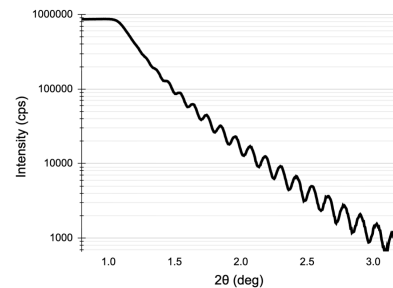


Figure 13: Example XRR scan of Ru-Pt alloy. Spacing of fringes used to extract thickness.

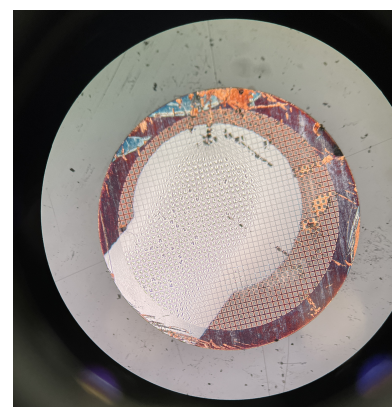


Figure 14: Optical image of TEM copper grid with 10 nm thick $\text{Ru}_{78}\text{Pt}_{22}$ deposited by magnetron sputtering.

The same instrument, JEOL 2100F - S(TEM), was used to collect diffraction patterns of the film, illustrating the degree of polycrystallinity. These rings represent the accumulation of diffraction spots from each tiny crystallite, with different crystals contributing reflections at various angles, which, when averaged, create continuous rings rather than discrete spots. The radii of these rings are measured and analyzed to determine the d-spacings and crystal structure of the material using Bragg's Law, similar to XRD.

PHYSICAL PROPERTY MEASUREMENT SYSTEM (PPMS)

The EverCool II Physical Property Measurement System (PPMS) from Quantum Design is a versatile cryogen-free characterization piece of equipment with various measurement modules, full temperature-field control and is designed to perform a variety of automated measurements.

Ultrahigh-purity He gas is condensed into a liquid by a compressor and is collected in a dewar. The superconducting coil can provide up to 12 T in uniaxial magnetic field strength and is immersed in the liquid He, while evaporating He gas from the bottom of the dewar provides cooling of the sample space.

The most commonly used modules relevant to this work are discussed below:

Vibrating Sample Magnetometer (VSM)

The VSM module is used to collect magnetization hysteresis (MH) loops as a function of field and temperature. These sorts of measurements provide insight into the magnetic ordering of spins in a material and the resulting moment of the sample.

VSM measures the magnetic properties of a material by oscillating it at a certain frequency within a magnetic field. The vibrating sample, moving through pickup coils, induces a voltage according to Faraday's Law. By measuring this induced voltage and relating it to a known standard, the VSM can determine the sample's magnetic moment and other magnetic characteristics.

1. Identify whether an in-plane (IP) or out-of-plane (OOP) sample holder is required depending on the sample and property of interest. IP sample holder is quartz, OOP is brass.
2. Adhere the sample to the sample rod using varnish and let dry for at least 1 hour. Ensure that the sample is within the bounds of the holder and not protruding.

To reliably detect the transmitted electrons in TEM, the sample should be less than 100 nm thick.

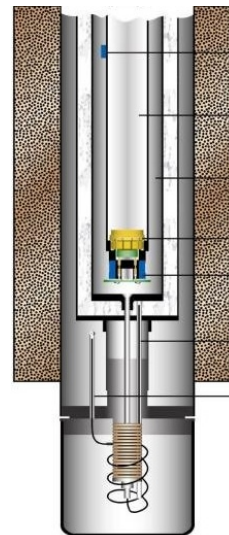


Figure 15: Schematic of PPMS dewar [16].

The field inside the PPMS is oriented **perpendicular** to the ceiling.

Verify using the provided Pd standard that the VSM is well calibrated (account for any field drift). This should be done every 3 months or after 10 consecutive field sweeps above 5 Tesla.

The PPMS limits are 2 K – 400 K & ± 9 T. Do not exceed or approach these limits for extended periods of time. Do not exceed a temperature rate change of ± 12 K/min.

3. Install the VSM module:

- (a) Vent the chamber. Ensure all modules are deactivated.
- (b) Insert the temperature sensor using the puck insertion tool.
- (c) Insert guide rod (hollow plastic rod).
- (d) Install the VSM motor carefully and clamp the flange.
- (e) Connect the VSM module cable (box) and the motor cable.
- (f) Activate the VSM option. Make sure the motor (yellow position indicator moves to top) and the coil thermometer are initialized properly.

4. Screw the sample into the sample rod and insert at the top of the motor. Replace the cover.

5. To locate the sample, apply a small field (100 Oe) and follow instructions on the MultiView software to complete the setup.

The MH curves, field-cool (FC), and zero-field cool (ZFC) measurements presented in this thesis are all performed using this module.

He gas is used up faster for high-field scans ($> 3\text{T}$) than for low-temperature scans. Be sure to maintain He levels above 70%.

DO NOT purge the system if the black seal at top of motor is not put back.

You must perform an extended purge if cooling below 250 K.

BREAKING DOWN A MAGNETIZATION LOOP

The key features of a typical magnetization (MH) curve are identified and explained below:

- M_S is the saturation magnetization, which can be found by normalizing the total magnetic moment of the sample when all magnetic moments are aligned with the field to a unit of volume. The units are typically $[\text{emu}/\text{cm}^3]$.
- H_S is the saturation field, intuitively the field at which M_S is reached.
- H_c is the coercive field with units of Oersted [Oe]. This quantity refers to the magnitude of the external magnetic field required to demagnetize a FM after it has been driven to magnetic saturation. Typically, it is the width of the MH loop at $M = 0$ and calculated as the average of the positive and negative x-intercepts of the MH loop:

$$H_c = \frac{|H_{c+}| + |H_{c-}|}{2} \quad (31)$$

Materials with low coercivities are considered magnetically soft while those with large coercivities are magnetically hard (i.e. permanent magnets).

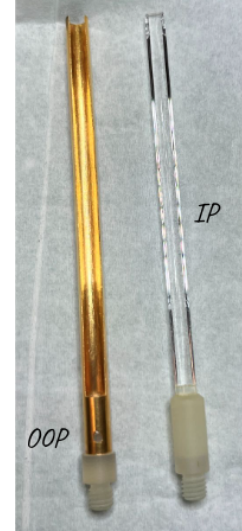


Figure 16: Out-of-plane (OOP) and in-plane (IP) samples holders used for VSM measurements.

- H_k is the kink field, also with units of [Oe], and is given by

$$H_k = \frac{2K_u}{\mu_0 M_S} \quad (32)$$

where K_u is magnetocrystalline anisotropy energy density and μ_0 is the vacuum permeability. In thin films or anisotropic materials, it typically corresponds to the field required to rotate the magnetization from the easy axis toward the hard axis. If the field is applied along a direction that is not the easy axis, H_k marks the onset of significant magnetization rotation toward the field direction. In materials with large K_u , H_k indicates the field strength needed to start reorienting spins against the anisotropy energy. For multi-domain systems, the kink can represent the field at which domain walls start to move significantly. Hence, H_k is strongly direction-dependent, reflecting the in-plane versus out-of-plane anisotropy.

- H_{EB} is the exchange bias field in units of [Oe]. It arises due to exchange coupling at FM/AFM interfaces. This parameter is depicted as a horizontal shift of the MH loop and quantifies the bias imposed by the AFM on the FM.

$$H_{EB} = -\frac{H_{c+} + H_{c-}}{2} \quad (33)$$

It is temperature-dependent and typically decreases with increasing temperature, vanishing above the AFM's T_N . It can also be training or history-dependent due to domain reconfiguration.

- M_{rem} is the remnant magnetization in units of [emu/cm³]. It quantifies the residual magnetization remaining after the external field is reduced to zero. For soft magnets, M_{rem} is low whereas for hard magnets it is high.

Electron Transport (Resistivity)

The resistivity module provides temperature- and field-dependent four-probe characterization of a sample. This requires wire-bonding the sample to a sample chuck and applying a current while measuring the voltage difference between leads. This module can be used for measuring both transverse (or Hall) (R_{xy}) and longitudinal (R_{xx}) resistances depending on the wire bonding geometry. Additionally, van der Pauw measurements can also be performed on more ambiguous samples with atypical geometries.

1. Adhere the sample to the sample puck using varnish and let dry for at least 1 hour. You may bond up to 3 different samples on a single puck.



Figure 17: PPMS resistivity sample chuck. Image shows 2 separate bonded samples.

2. Install the resistivity module:

- (a) Vent the chamber. Ensure all modules are deactivated.
- (b) Insert the sample at the bottom using the puck insertion tool.
Make sure to rotate and confirm it is inserted properly by aligning the notch.
- (c) Insert baffle rod (thin plastic rod) and clamp the flange.
- (d) Connect the resistivity module cable.
- (e) Activate the resistivity option.

3. Check the bridge configuration and confirm connectivity of all pins and contacts for each channel.

The MR and longitudinal transport measurements presented here are all performed using this module.

Additionally, a rotator mount was employed to measure angle-dependent R_{xx} between the sample's surface normal and the applied magnetic field. This enables 360° freedom in sample rotation, enabling precise AMR measurements.

Magneto-Optic Kerr Effect (MOKE) Microscopy

MOKE microscopy is a nondestructive technique which can be used to visualize the magnetic domain configurations inside a magnetic material. Its setup requires a high-output-power optical source, a high extinction ratio of polarizers, a large bit-depth camera (i.e. low noise level), and sufficiently high spatial resolution ΔX given by:

$$\Delta X = \frac{C\lambda}{A_N} \quad (34)$$

where $C = 0.61$, A_N is the numerical aperture of the objective lens, and λ is the laser wavelength.

Linearly polarized incident light becomes elliptically polarized after reflection. As the light reflects, the material's magnetization causes the light's polarization to change. If the direction of magnetization in the material is reversed, the direction of the polarization rotates and the sense of the ellipticity also reverses. The polarized light is then focused onto the magnetic sample through a beam splitter. The reflected light beams passes through an analyzer, which is oriented almost 90° to the incident polarizer such as to filter the unpolarized light after reflection. This light is reflected back through the objective lens and the analyzer before being focused on the CCD detector.

For standard measurements, set the control limits to 10 μA , 10 mV and 100 μW . If the applied current is significantly less than the control limit, try reducing the limit or check the connectivity of the wire bonds.



Figure 18: PPMS resistivity rotator mount used for angle-dependent R_{xx} measurements.

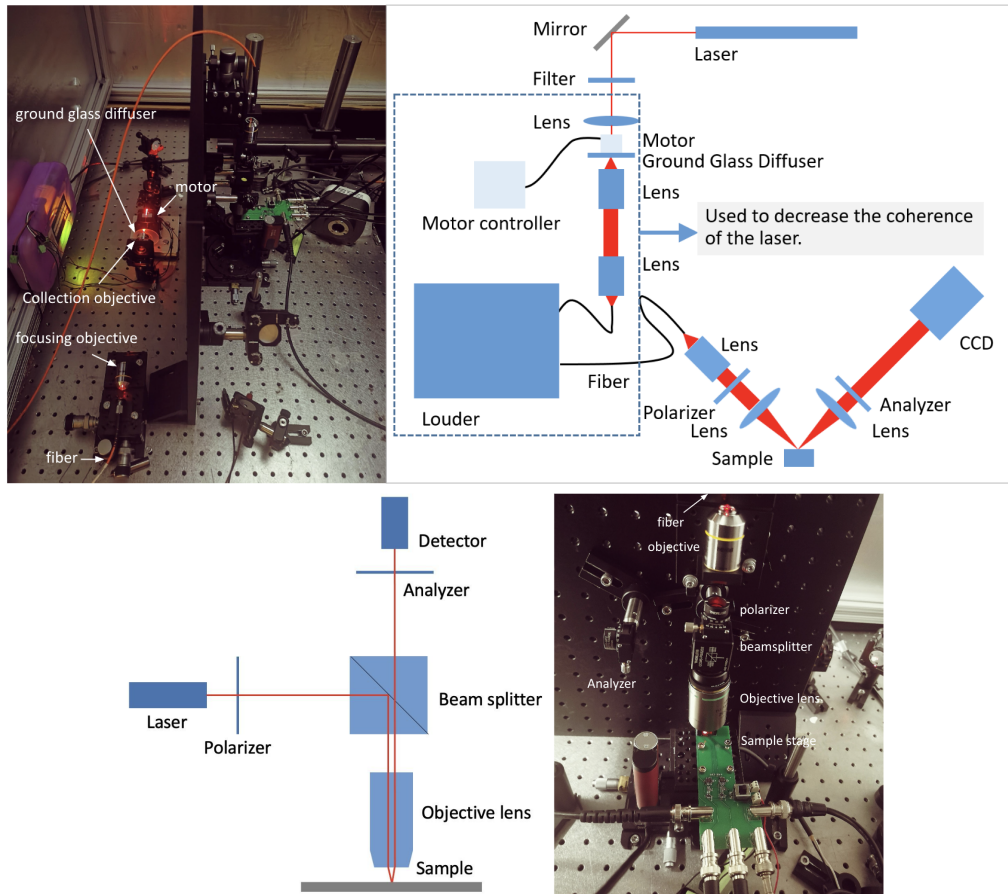


Figure 19: Schematics and photographs of polar-MOKE setup with various optical component labeled.

Contrast in a MOKE image comes from magnetization polarization, not depth contrast as in optical images. MOKE is categorized by the direction of the magnetization vector relative to the sample surface and the plane of incidence of the light. The three main types are polar, longitudinal, and transverse. Polar-MOKE is predominantly used in this work, which detects the out-of-plane magnetization.

Ferromagnetic Resonance (FMR)

FMR describes the phenomenon in which the magnetization of a ferromagnetic material absorbs energy from an applied microwave field when their frequencies match, causing the magnetization to precess (wobble) at a specific resonant frequency determined by the material's properties and an applied external magnetic field. Spin-torque FMR (ST-FMR) is a widely-used technique to determine the SHA of a material. It has the added advantage of not requiring perpendicular magnetic anisotropy (PMA) of the FM layer to maximize the out-of-plane magnetization response. This is a limitation that fol-

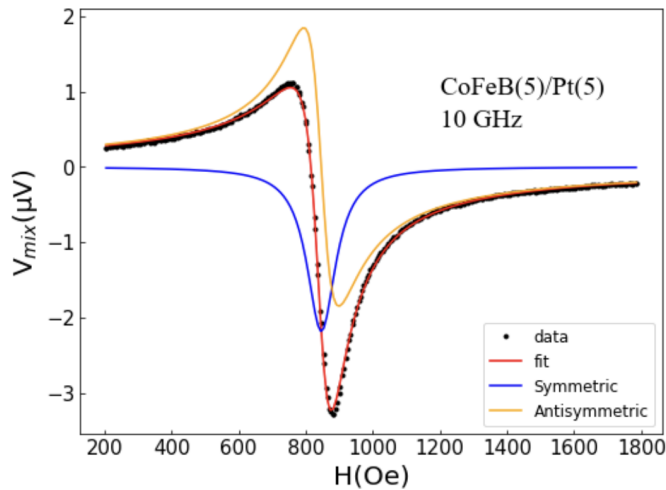
lowers directly from the shape and surface anisotropy favoring in-plane magnetization of spin in a thin film.

A non-magnet/FM bilayer is required for this measurement. A constant magnetic field is applied to set the magnetization of the magnetic layer. A radio frequency (rf) current is then applied, causing precession of the material's magnetization. This allows one to measure the oscillating spin torque generated by the spin current through FMR.

Due to strong SOC in the non-magnetic heavy metal layer, a transverse oscillating spin current is generated by the SHE. This spin current is then injected into the adjacent FM layer, which exerts an oscillating spin torque on it. This spin torque induces magnetization precession of the spin in the FM. The spin dynamics in the FM layer can be described by the well-known Landau-Lifshitz-Gilbert (LLG) equation:

$$\frac{d\mathbf{M}}{dt} = -\gamma\mathbf{M} \times \mathbf{H}_{\text{eff}} + \alpha\mathbf{M} \times \frac{d\mathbf{M}}{dt} + \gamma \frac{\hbar}{2e\mu_0 M_s t} \times J_s(\mathbf{M} \times \boldsymbol{\sigma} \times \mathbf{M}) - \gamma\mathbf{M} \times \mathbf{H}_{\text{rf}}. \quad (35)$$

Important terms include the torque from the effective field $\mathbf{H}_{\text{eff}} \approx \mathbf{H}_{\text{ext}} - 4\pi\mathbf{M}_{\text{eff}}$ for thin films with in-plane magnetization, the torque induced by the spin current J_s injected into the FM layer, and the torque induced by the Oersted field $\mathbf{H}_{\text{rf}}$ generated from the rf current. Here, γ is the gyromagnetic ratio, α is the Gilbert damping coefficient which accounts for how fast the precessing spin dampens to the applied field direction, μ_0 is the permeability of free space, M_s is the saturation magnetization of the FM, t is the thickness of the FM, and $\boldsymbol{\sigma}$ is the spin direction.



Due to the AMR effect in the FM layer, the resistance of the bilayer

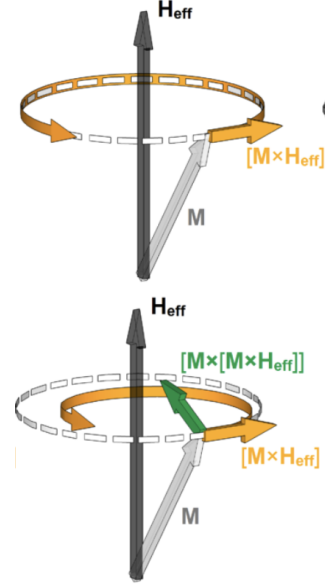


Figure 20: Spin precession and damping about a constant external applied field direction in response to an applied rf current.

Figure 22: FMR signal of CoFeB/Pt bilayer used for system calibration indicating symmetric and antisymmetric contributions to V_{mix} [17].

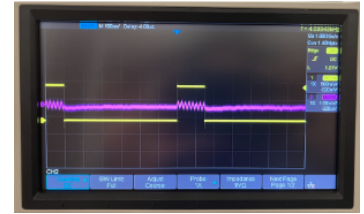


Figure 21: Screen capture of pulsed rf signal for FMR measurements. Signal produced by modulating reference square pulse from signal generator (yellow) with rf GHz signal (purple).

oscillates with the angle of the applied external field. This resistance mixes with the rf current and generated a dc voltage across the sample (V_{mix}) in response to the torques.

This voltage can be detected using a lock-in amplifier and manifests as a dip in the voltage signal as a function of applied field, resembling a superposition of a symmetric (S) and antisymmetric (A) Lorentzian function. By analyzing these two contributions, the SHA can be derived as

$$\theta_{\text{SH, FMR}} = \frac{S}{A} \frac{e\mu_0 M_s t d}{\hbar} \left(1 + \sqrt{\frac{4\pi M_{\text{eff}}}{H_{\text{ext}}}}\right). \quad (36)$$

Crucial components of the FMR setup include: an electromagnet to provide the static magnetic field, an rf probe with a ground-signal-ground (GSG) configuration to match the coplanar waveguide geometry of the sample, an rf generator to supply the rf current at a sufficiently large power ($\sim +10$) dBm, a signal generator to pulse the rf current using a square wave to stimulate spin dynamics in the ferromagnet/heavy metal bilayer, a bias tee to split the dc from the rf component of the reflected voltage signal, and a lock-in amplifier to detect the small dc component, typically on the scale of a few microVolts.



Figure 23: Ferromagnetic resonance measurement setup. Important components are labeled.

The sample geometries are carefully chosen such that they are impedance matched to $\sim 50 \Omega$ to maximize the signal quality without unwanted losses.

The G-S-G sample configuration is crucial for several reasons:

- It produces a coplanar transmission line with well-defined electromagnetic field confinement between the signal line and the surrounding grounds.
- It provides additional degrees of freedom to carefully tune the sample dimensions which allows precise matching to the desired characteristic impedance.
- The two grounds on either side of the signal line produce a symmetric electric and magnetic field profile. This symmetry prevents unwanted electric field coupling and ensures that the generated microwave magnetic field is localized and uniform across the sample area directly above the signal line.
- The grounded planes confine the field, reducing stray radiation which can enhance the signal-to-noise ratio of the FMR signal.

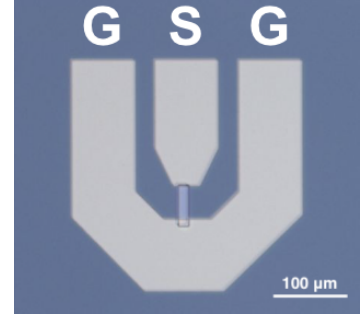


Figure 24: Optical image of coplanar waveguide device used for ST-FMR measurements. Ru-Pt rectangular strip is shown along with G-S-G Cr/Au leads for rf probe contacts.

Chapter 3 | Intrinsic Spin Polarization & Quantum Transport in CrO_2

A CENTRAL challenge in spintronics is the efficient generation of spin-polarized currents, which serve as the foundation for downstream spin transmission and manipulation. Half-metallic FMs, where conduction occurs predominantly through a single spin channel, provide an almost ideal platform for this purpose. Epitaxial CrO_2 thin films are a canonical example, combining robust ferromagnetism, near-perfect spin polarization, and high crystalline quality. This makes CrO_2 an ideal material for exploring fundamental spin-dependent transport phenomena. Understanding the intrinsic transport behavior of CrO_2 is therefore critical for both developing new spintronic devices and establishing benchmarks for interfacial spin transmission.

In this chapter, the low-temperature, high-field magnetotransport properties of CrO_2 thin films with a particular focus on phenomena that reveal the quantum-coherent motion of spin-polarized carriers. Angle-dependent anisotropic magnetoresistance (AMR) measurements probe the symmetry and orientation of the electronic density of states, offering insight into the interplay between crystal structure and spin-dependent scattering. The first experimental observation of Shubnikov–de Haas (SdH) oscillations in CrO_2 provides a direct window into its Fermi surface geometry, effective masses, and quantum coherence of the conduction electrons. The results discussed in this chapter have been recently published in [18]. The combination of these techniques allows us to link the half-metallic nature of CrO_2 to its ability to generate highly spin-polarized currents, establishing a firm foundation for the subsequent study of spin transmission across interfaces such as in $\text{RuO}_2/\text{CrO}_2$ bilayers.

Placing CrO_2 within the broader context of spin generation, spin transmission, and spin-charge conversion, this work emphasizes its role as a model spin injector, highlighting the intrinsic transport properties that enable efficient spin current flow and setting the stage for understanding interfacial phenomena in complex oxide heterostructures.

MOTIVATION

Chromium dioxide (CrO_2) is an exemplary half-metallic oxide [19] that has attracted considerable interest due to its exceptionally high spin polarization at the Fermi level, approaching $P = 96 - 98\%$ [8, 20–24]. It also possesses a relatively T_C of approximately 390 K [25]. Its ferromagnetic ordering above room temperature, driven by

the double-exchange mechanism, makes CrO_2 a strong candidate for spintronic devices that demand reliable control of spin states. In addition, CrO_2 exhibits a large bulk saturation magnetization, reaching about 480 emu/cm^3 at room temperature [26] and increasing to nearly 640 emu/cm^3 at low temperatures [27].

Due to the single-spin conduction channel in half-metals, there are distinct technological advantages available to enhancing magnetotransport properties. The oxide's structural compatibility with rutile altermagnets (XF_2 , where $X = \text{Mn, Fe, Co, Cu}$) as well as with other unconventional spin-torque materials such as RuO_2 and IrO_2 [28, 29] further extends its applicability [30–33]. In addition to these electronic benefits, CrO_2 is chemically stable and straightforward to synthesize, making it well suited for robust, long-term applications. A defining characteristic of its half-metallic nature is the large MR it exhibits, a property of central importance for magnetic sensing technologies.

Despite these attractive properties, the quantum electronic and magnetotransport behavior of CrO_2 , especially in the thin film limit, has not been thoroughly investigated. Most of the prior studies on this topic have concentrated on bulk films, microcrystals, or polycrystalline samples, often restricted to low-field or high-temperature regimes [25, 34, 35]. In this chapter, the high-field and low-temperature MR of epitaxial CrO_2 thin films is examined in the sub-100 nm limit. The findings address several gaps in the current understanding of the quantum transport properties of CrO_2 thin films, and further highlight their promise for spintronic and magnetotransport applications.

FABRICATION

CrO_2 films were epitaxially grown via CVD. TiO_2 substrates with a (100) orientation, measuring 25 mm^2 in lateral area were used. These substrates were specifically chosen to minimize lattice mismatch and reduce the effects of strain on the magnetocrystalline anisotropy in CrO_2 films. For transport studies, the CrO_2 films were patterned into $55 \mu\text{m} \times 20 \mu\text{m}$ Hall bar devices using photolithography techniques. These Hall bars were designed to be oriented along both the b ([010]) and c ([001]) crystallographic directions to allow simultaneous transport measurements under identical conditions. The devices were etched out by reactive ion etching. Following photoresist removal, the Hall bars were wire-bonded for electrical measurements.

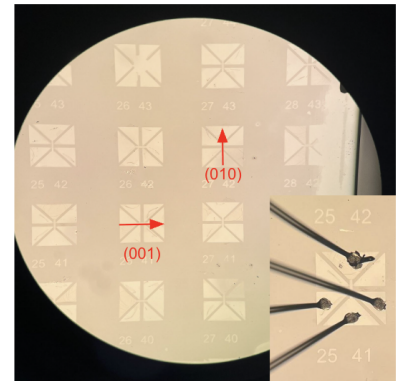


Figure 25: Optical image of CrO_2 Hall bars oriented along [010] and [001] directions. Inset shows wire-bond geometry for ρ_{xx} measurements.

Structural Characterization

Surface Oxidation

It is known that CrO_2 reduces to Cr_2O_3 in an oxygen-deprived environment (i.e. under ambient conditions). Cr_2O_3 is an AFM and is undesirable for this study on CrO_2 . Using a wide-range XRD scan, it was confirmed that these films do not reduce to the native oxide. A 50-day old, 9 nm-thick CrO_2 film is measured, which is thin enough to prevent saturation of the Cr_2O_3 signal from bulk CrO_2 beneath it. The absence of peaks at the expected 2θ positions for Cr_2O_3 confirms that the films, given the resolution of the instrument, have less than a nanometer of Cr_2O_3 at the surface, if at all.

X-Ray Strain Analysis

Structural characterization was carried out using high-resolution X-ray diffraction (HRXRD) on a Rigaku Smartlab diffractometer equipped with a Ge (220) double-bounce monochromator. The epitaxial quality of the 53 nm sample used for the transport experiments below was confirmed [36, 37]. A wide-angle symmetric θ - 2θ scan shows the film has only one out-of-plane crystal phase which matches that of the substrate.

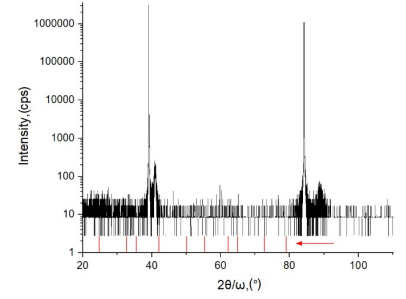
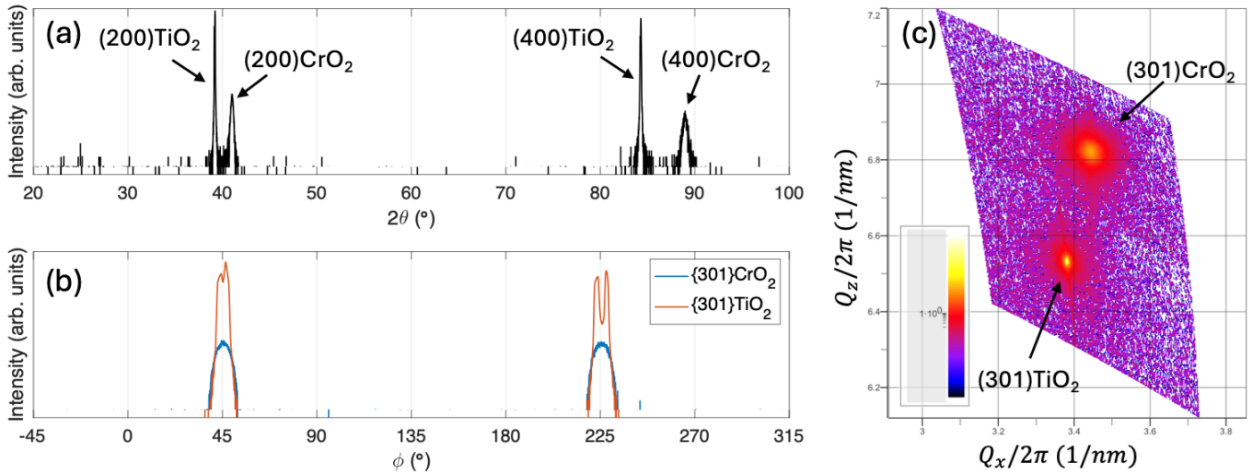


Figure 26: XRD scan of 9 nm CrO_2 film after 50 days in ambient conditions. No Cr_2O_3 peaks are observed (expected at red markers), indicating little to no oxidation.



A full-range ϕ -scan of the asymmetric 301 planar family for both the CrO_2 film (blue) and the TiO_2 substrate (orange) was collected, which confirms that there is also only one in-plane crystal phase of the film that matches the rutile structure of the substrate. The diffraction results indicate an isostructural rutile phase consistent

Figure 27: Structural characterization of 53 nm CrO_2 film by high resolution X-ray diffraction. (a) Symmetric $\theta/2\theta$ scan, (b) ϕ scan of asymmetric 301 planar families, and (c) reciprocal space map of asymmetric (301) plane. Scans performed by David T. Plouff and collaborators at the University of Delaware.

with the TiO_2 substrate, with relatively small compressive strains of 0.48% and 0.38% along the a and c lattice parameters, respectively. Furthermore, a reciprocal space map (RSM) of the asymmetric (301) peak illustrates that the film is relatively unstrained, with extracted lattice parameters $a=4.398 \text{ \AA}$ and $c=2.904 \text{ \AA}$. When comparing these lattice parameters to those of an unstrained powder reference sample (using PDF Card 01-071-0869), strain values of -0.48% and -0.38% are obtained for the a and c lattice parameters, respectively.

Magnetic Characterization

For this project, several CrO_2 films of varying thicknesses were fabricated. Thickness calibrations were performed on as-deposited, unpatterned films using VSM and further confirmed with ellipsometry. The advantage of using VSM here is that it is a non-destructive measurement technique that relies on the strong FM moment of the CrO_2 films. The net saturation magnetization (M_s) can then be normalized by the area of the film to calculate its thickness with high precision.

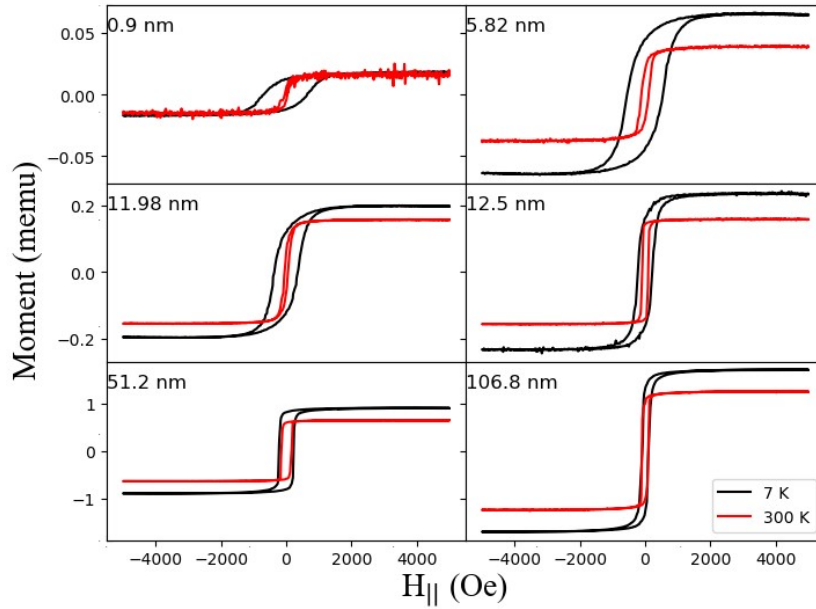


Figure 28: Magnetization loops measured using VSM for six different CrO_2 samples at 7 K and 300 K along [010], with calculated thickness denoted.

Thickness-Dependent Fit to Bloch's Law

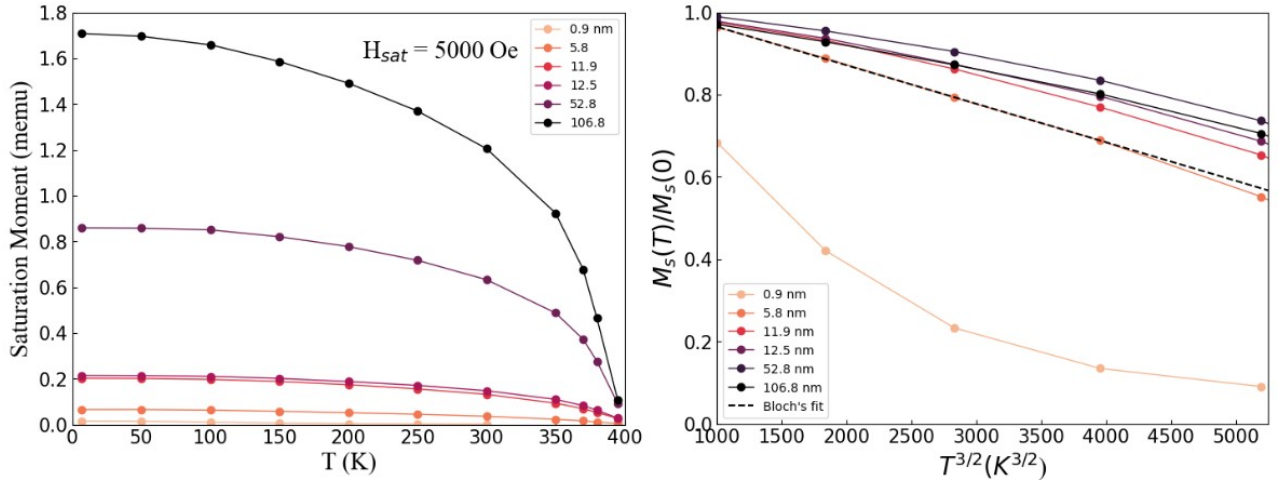
Magnetization (MH) loops are collected at both room temperature (300 K) and at low temperature (7 K) using VSM. Only the M_s at 7 K, however, is used to extract the film thickness data. This is because

FMs follow Bloch's law at low temperatures, meaning their magnetization is closer to the intrinsic $M_s(0)$ at 0 K. This behavior is captured by the equation

$$M_s(T) = M_s(0)[1 - BT^{3/2}], \quad (37)$$

where B is the magnitude of the external applied field and T is the temperature. Additionally, lower temperatures are marked by reduced thermal noise and minimized diamagnetic background from the substrate or sample holder.

Bloch's law describes how the M_s of a FM decreases with increasing temperature, valid for $T < T_C$.



As shown, Bloch's law describes the data well for samples thicker than 5 nm, as expected for FMs in the bulk regime. A notable deviation from Bloch's $T^{3/2}$ fit is observed for samples with thicknesses below 5 nm, approaching 1 nm. This points to a possible 2D transition for CrO₂ in the ultra-thin regime and is worth exploring in future studies.

A JA Woollam M-2000 Variable Angle Research Grade ellipsometer was also used to characterize these films by analyzing changes in the polarization state of light as it reflects off or passes through the sample. It is a non-destructive optical technique employed to determine the thickness and optical constants of surfaces and thin films. Both approaches, VSM and ellipsometry, provide good agreement on the thicknesses of the CrO₂ films studied here.

Analysis of MH Loops

The magnetization loops are also analyzed to extract key parameters including the coercive field (H_c) and to map the M_s to the deposition time of CVD.

Figure 29: Deviation from Bloch's law. (Left) Saturation magnetization plotted as function of temperature for CrO₂ films of varying thicknesses. (Right) Normalized data replotted as a function of $T^{3/2}$.

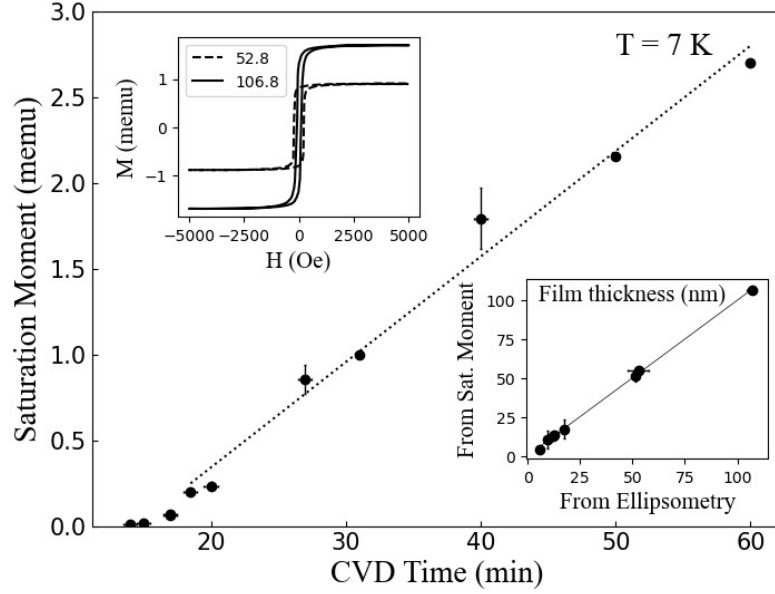


Figure 30: Saturation magnetization (M_s) determined from VSM measurements plotted as a function of CVD time. Top left inset illustrates differences in M_s for 52 nm and 106 nm films. Bottom right inset shows strong agreement in calculated film thicknesses from ellipsometry data with M_s extrapolation from MH curves.

It is found that M_s increases linearly with CVD time with an initial "activation" time of approximately 12 minutes, during which no deposition occurs. The coercive field is also found to be strongly thickness-dependent, following a clear $1/\sqrt{t_{\text{CrO}_2}}$ trend. This is consistent with previous studies on CrO_2 [36] as well as other FM systems [38].

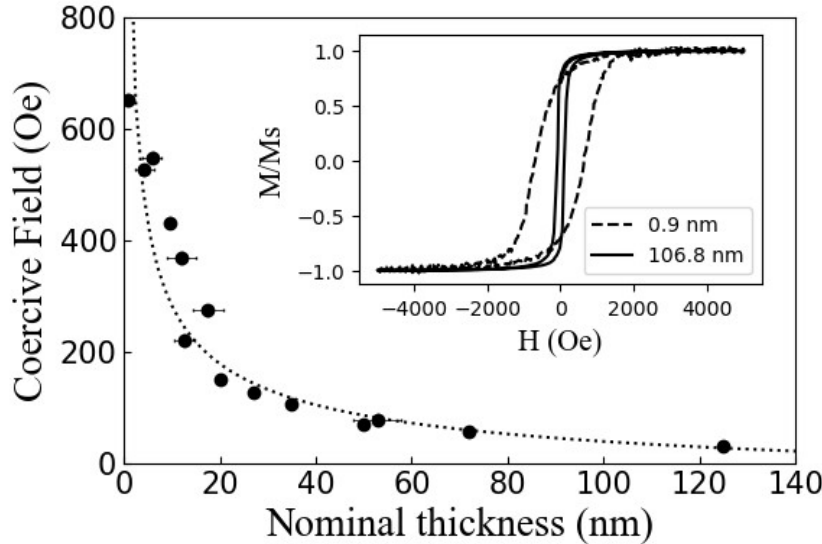


Figure 31: Coercive field (H_c) extracted from VSM measurements plotted as a function of CrO_2 film thickness. Inset illustrates differences in the normalized H_c for 1 nm versus 106 nm thick films.

Electron Transport Characterization

Longitudinal Resistivity

The patterned films are wire-bonded and loaded into the resistivity module in the PPMS to collect electron transport properties. First, the temperature-dependent longitudinal resistivity (ρ_{xx}) of the CrO₂ films with various thicknesses is compared, with current applied along the two in-plane orthogonal crystallographic axes.

Overall, the resistivity increases with decreasing film thickness, as expected. It follows a quadratic trend with temperature, pointing to the typical scattering mechanisms predicted to dominate in metallic systems. The low-temperature resistivities along [010] are almost always higher than those along [001]. This observation can be explained by the cross sections of the crystallographic unit cell along these directions. More scattering centers (i.e. atoms) are present within a smaller volume in the (010) plane as compared to the (001) plane. This breaks down once the film becomes sufficiently thin, approaching the 2D limit, where the low-temperature resistivities converge while the high-temperature values deviate notably.

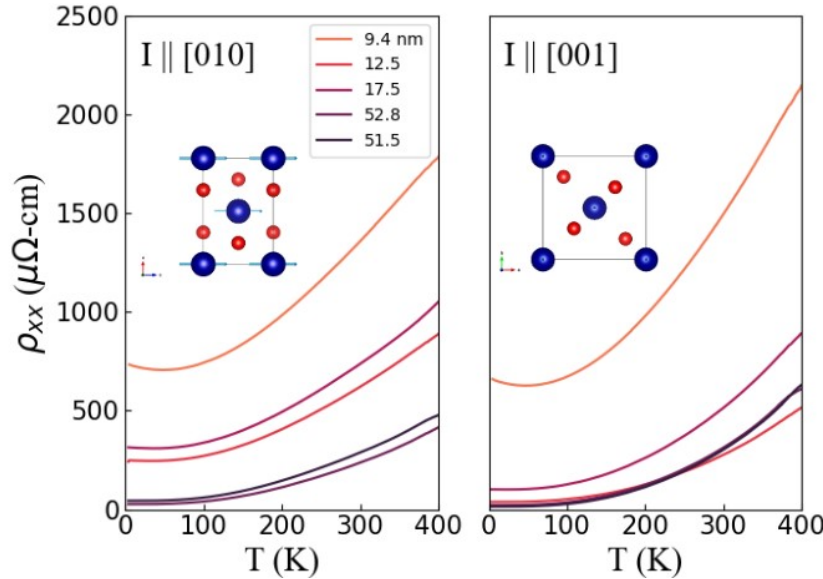


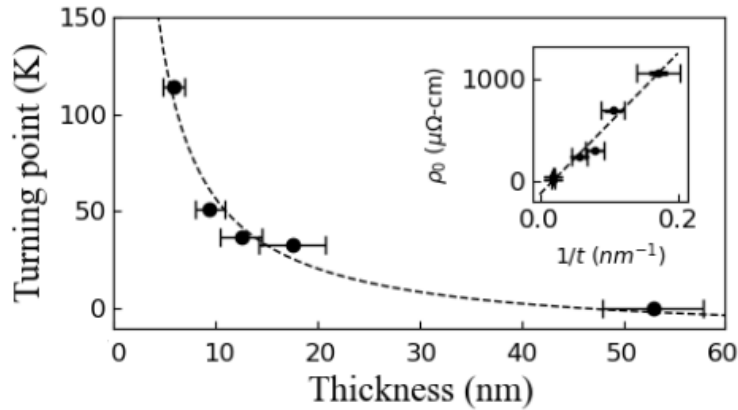
Figure 32: Longitudinal resistivity of CrO₂ films of varying thicknesses along b ([010]) and c ([001]) axes. Insets show cross section of crystallographic unit cell along these directions.

The residual resistivity ratio (RRR), defined as $\rho(400 \text{ K})/\rho(2 \text{ K})$, is a useful metric to quantify the electronic quality of the samples. Further analysis of the temperature-dependent ρ_{xx} data reveals a strong anisotropy in RRR values along the [010] and [001] directions. The results show that the RRR along [010] exceeds that along [001] for all the film thicknesses, in some cases by as large as 150 times

for bulk samples. The RRR reaches a maximum of 45 along the [001] direction for the 53 nm CrO_2 film, reflecting the high crystalline. For comparison, reported RRR values for CrO_2 thin films span roughly 6 to 50, depending on substrate orientation, composition, and film thickness [8, 39–41].

Interestingly at low temperatures, the resistivity of CrO_2 thin films shows a slight upturn that becomes more pronounced in thinner films. When the characteristic temperature of this upturn is plotted as a function of film thickness t_{CrO_2} , a clear $1/t_{\text{CrO}_2}$ decay of the turning point temperature is observed.

This behavior reflects the increasing influence of surface and interfacial scattering as the film thickness decreases. In thick films, the conduction is dominated by bulk metallic behavior, and resistivity decreases monotonically with temperature due to reduced phonon scattering. However, as the film becomes thinner, electron motion becomes increasingly confined, and the contribution from surface and grain boundary scattering (which are temperature-independent) begins to dominate at low temperatures. The crossover temperature where this becomes significant scales inversely with the effective thickness of the conducting channel.



Additionally, CrO_2 is a HMF, so the low-temperature upturn may also involve quantum corrections to conductivity such as weak localization or electron-electron interaction effects, which are enhanced in reduced dimensions. These effects become more pronounced when the electron mean free path approaches the film thickness, again consistent with the inverse scaling behavior.

In this context, the observed resistivity upturn is best understood as a manifestation of finite-size and interfacial scattering effects, marking a transition from bulk-dominated to surface-dominated transport as the film thickness decreases. This scaling thus provides a quantitative signature of the dimensional confinement of charge

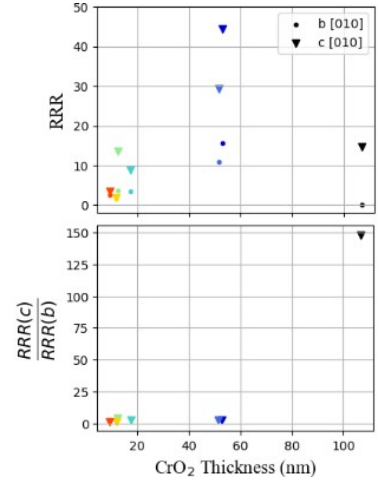


Figure 33: Summary of residual resistivity ratios (RRR) calculated along the b [010] and c [001] current directions.

Figure 34: Plot of upturn (turning point) temperature in resistivity as a function of film thickness, t . Inset shows $1/t$ dependence of residual resistivity, ρ_0 .

carriers in epitaxial CrO₂ films.

Characterization of Scattering Mechanisms

For subsequent discussions and analysis, the focus is placed on the temperature-dependent ρ_{xx} data of the 53 nm film, with current applied along the two crystallographic axes. As expected, T_C at approximately 396 K. Interestingly, a crossover between the ρ_{xx} curves along the two current directions occurs near 195 K, in good agreement with earlier studies [34]. A logarithmic plot better illustrates the low temperature scaling.

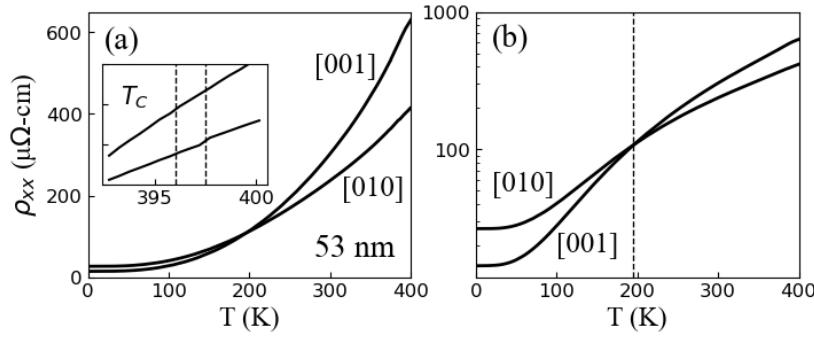


Figure 35: (a) ρ_{xx} of 53 nm CrO₂ film with current applied along the [010] and [001] directions. (b) Data replotted with logarithmic scaling to show details at low temperature.

First, ρ_{xx} is extrapolated to 0 K which enables the extraction of the residual resistivity (ρ_0). These values are low, comparable to those from prior studies [42, 43], and determined with very high precision. The temperature dependence of ρ_{xx} is then modeled using a power law of the form

$$\rho_{xx}(T) = \rho_0 + AT^n, \quad (38)$$

where n characterizes the scattering processes.

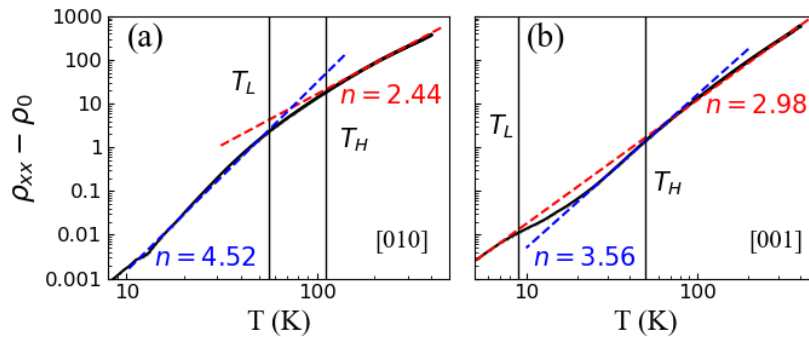


Figure 36: Extraction of scattering exponent n . Low (blue) and high (red) temperature regimes indicated by verticals T_L and T_H , respectively. Dashed lines are linear fits along (a) [010] and (b) [001] with corresponding slopes denoted by n .

Temperature-dependent resistivity is presented by $(\rho_{xx} - \rho_0)$ on a logarithmic scale, enabling the precise determination of the critical

temperatures T_H and T_L which are used to distinguish between the high- and low-temperature regimes, respectively. Linear fits yield the exponent n . This provides insight into the underlying transport mechanisms.

Axis	ρ_{300} ($\mu\Omega\text{cm}$)	ρ_0 ($\mu\Omega\text{cm}$)	RRR	T_C (K)	T_L (K)	T_H (K)	$n(T_L)$	$n(T_H)$
[010]	237	26.4191 ± 0.0004	16	397	56	112	4.52	2.44
[001]	303	14.1752 ± 0.0010	45	396	9	50	2.98	2.98

Table 2: Summary of transport parameters of 53 nm CrO_2 film along [010] and [001] current directions.

Along the [010] axis, two clear transport regimes are observed: an exponent $n = 2.44$ for temperatures above $T_H = 112$ K and $n = 4.52$ below $T_L = 56$ K. Models of spin-fluctuation scattering typically predict values of n in the 2–3.5 range [44, 45]. Other studies propose that conduction is curtailed by Anderson localization of the minority-spin channel, producing a resistivity dependence of $\rho_{xx} \propto T^{2.5}$ above roughly 60 K [46], which matches the temperature span of the observed crossover. At temperatures above 100 K, spin-disorder scattering is likely the primary mechanism, which yields $n \approx 2 - 2.5$. Because HMFs lack accessible minority-spin states, single-magnon scattering is suppressed, leaving double-magnon scattering dominant at low temperatures. This is indeed consistent with the low-temperature slope of $n = 4.5$ observed in this data [47–51]. Similar behavior has also been described within a rigid-band framework [51].

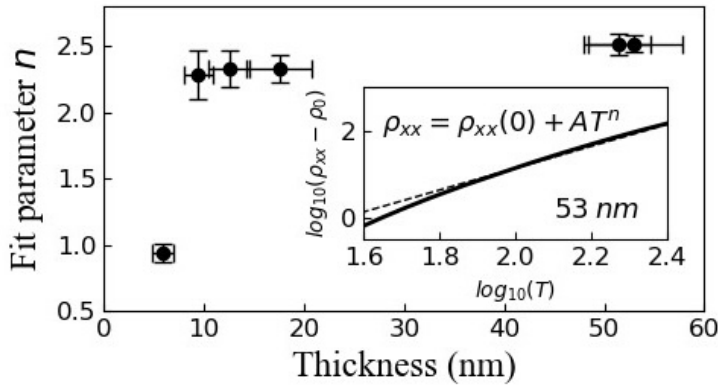


Figure 37: Plot of scattering exponent n as a function of thickness, with inset showing example of fit for 53 nm sample.

In contrast, the [001] direction shows a nearly constant exponent of $n = 2.98$ across the full temperature range from 5–400 K range, with a brief increase to $n = 3.56$ between 9 K and 50 K. Because this deviation occurs over such a limited interval, additional measurements are needed to determine whether the trend is truly monotonic. An exponent near 3 has been reported before and linked to non-rigid-band effects [52], likely stemming from unconventional one-magnon

scattering [48]. The relatively weak temperature dependence suggests that magnetic order and interactions are more uniform along the $[001]$ axis.

A thickness-dependent study of the scattering exponent n was also performed using this same approach. A very clear transition from $n = 1$ to $n \approx 2.5$ is observed for samples above 10 nm in thickness. The dependence of n on thickness is clearly binary, with a notable threshold value observed within the measured range of thicknesses in this study.

Band Structure & Theory

Half-metals possess complete spin polarization at the Fermi level and a gap in the minority-spin band. As such, their charge-carrier scattering is especially sensitive to spin-dependent interactions and electronic anisotropy. Coupling between spin, charge, and lattice degrees of freedom is therefore a defining feature of many HMFs.

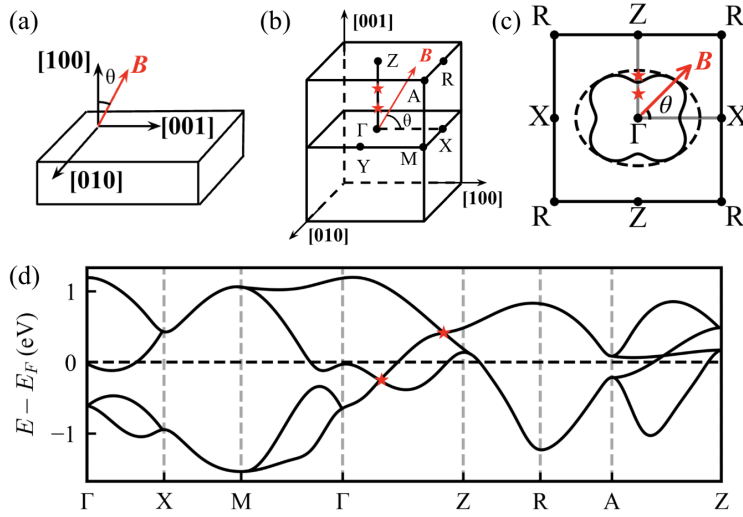


Figure 38: Electronic structure of CrO_2 . (a) Sample orientation used for measurements. (b) Reciprocal space bulk Brillouin zone of CrO_2 with high-symmetry points labeled. (c) Cross-sectional rendering of Fermi surface along the $k_x k_z$ plane (adapted from [44, 53]). (d) Band structure of spin majority carriers in CrO_2 near Fermi level. Triple point crossings indicated as red stars.

The directional anisotropies observed in ρ_{xx} point to variations in the band curvature and dispersion near the Fermi energy along the $[010]$ and $[001]$ axes. Crystallographically, $[010]$ corresponds to the $M - \Gamma$ path in the first Brillouin zone (BZ), while $[001]$ corresponds to the $\Gamma - Z$. Previous works report significant band crossings along $[001]$ [44, 53], including the presence of two triple points (TPs). This indicates a more complex band structure compared to $[010]$, where only single bands appear with no notable degeneracies [54].

TPs enhance the DOS, leading to more intricate and less temperature-dependent spin scattering. This is consistent with the nearly constant n along $[001]$ over a wide temperature range. In contrast, the ab-

sence of TPs along [010] results in simpler transport behavior with clear temperature-dependent transitions. The fewer bands and simpler electronic structure along [010] suggest that temperature has a stronger influence on scattering mechanisms, as seen in the pronounced transition of n from 4.5 at low temperatures to 2.5 at room temperature.

Notably, these TPs lie slightly below the Fermi energy (within about 0.5 eV). However, they can shift closer to E_F under external perturbations and thereby influence the electronic transport behavior. Low-temperature double-magnon scattering, observed above, could alter the spin-wave interactions and lead to an adjustment of the TP energies and potentially bring the nearby electronic states closer to the Fermi level [8, 55]. Additionally, Zeeman splitting between spin channels under strong magnetic fields combined with substantial SOC [56] could lift the TP degeneracies and produce significant energy shifts.

MAGNETOTRANSPORT CHARACTERIZATION

Magnetoresistance ($\theta_B = 90^\circ$)

Magnetotransport characterization was performed using a PPMS which enabled measurements of ρ_{xx} as a function of magnetic field orientated parallel to the film's surface normal.

The magnetic field, $\mathbf{B}_\perp$, is oriented perpendicular to the sample's surface. By sweeping the field at low temperature, MR curves for CrO_2 films of various thicknesses are obtained. An important observation is the thickness-dependent crossover from negative to positive MR at high fields. This is fairly common in conducting FM films. Several intertwined mechanisms can explain why thicker CrO_2 films show larger positive MR, while thinner films display weaker, negative MR.

The evolution from negative to positive MR with increasing film thickness can be understood as a shift in the dominant scattering mechanisms. In ultra-thin films strong interface disorder, epitaxial strain, and grain-boundary or domain-wall scattering enhance spin-disorder and magnon-related processes. When a magnetic field aligns spins and suppresses these fluctuations, the resistivity drops, producing negative MR.

Thin films can also exhibit weak localization and other quantum corrections that favor a negative field response. As the film thickness increases, remnant strains relax and the average crystalline quality improves, leading to higher carrier mobility and a longer mean free path. Under these conditions, classical orbital MR arising from the

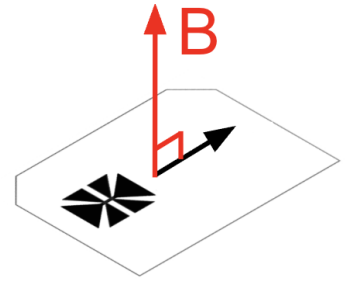


Figure 39: Orientation of magnetic field, B , relative to in-plane crystallographic axes of CrO_2 .

Lorentz force that bends charge carriers becomes the primary contribution and scales positively with field. The net effect is a gradual crossover: negative MR weakens as spin-disorder and localization diminish while positive orbital MR strengthens, resulting in the observed increase in positive MR for thicker films and the corresponding decrease in negative MR for thinner ones.

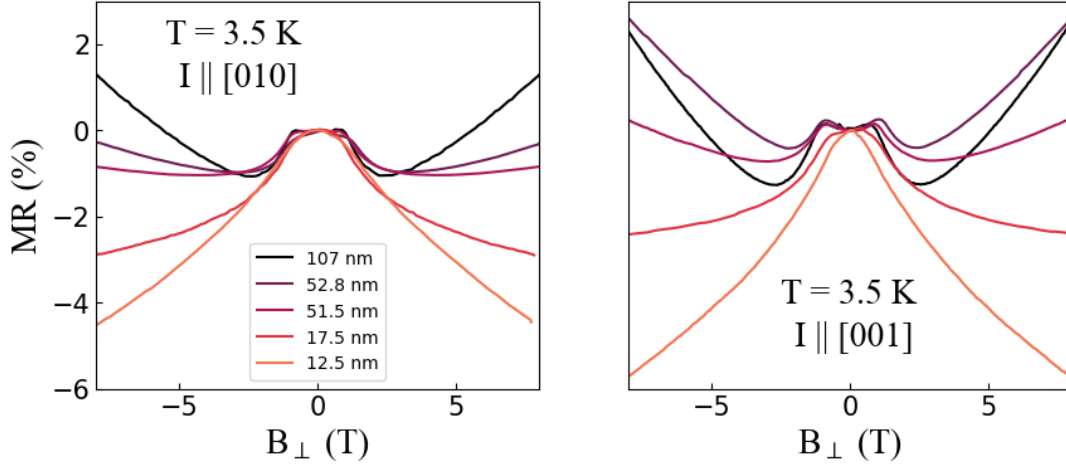


Figure 40: Magnetoresistance measurements along [010] and [001] current directions at 3.5 K for CrO_2 films of varying thicknesses.

As expected, the MR is also strongly orientation dependent. Replotting this data more directly to compare the MR along the b and c axes illustrates a strong axis-dependence anisotropy that disappears for sufficiently thin films. Thicker films in the bulk regime always exhibit higher, positive MR along the c axis. Below a certain critical thickness, the MR shifts to being negative. For very thin samples ($t_{\text{CrO}_2} \leq 10$ nm), the MR curves along the b and c axes converge, illustrating isotropic magnetotransport behavior.

Plotting the maxima MR achieved at 8 T along the [010] and [001] current directions better depicts this divergence with increasing thickness. The dashed lines represent an exponential growth in MR with CrO_2 film thickness.

Anisotropic Magnetoresistance ($\theta_B \neq 90^\circ$)

More interesting phenomena appears when the applied magnetic field is oriented at an arbitrary angle relative to the sample's surface normal with a nonzero in-plane component. For these measurements, the magnetic field, $\mathbf{B}$ is oriented at some angle θ relative to the surface normal. The schematics show how the same angle is oriented differently along the b and c in-plane crystallographic axes.

Angle-dependent ρ_{xx} measurements were performed to investigate the influence of temperature and magnetic field on the 53 nm CrO_2

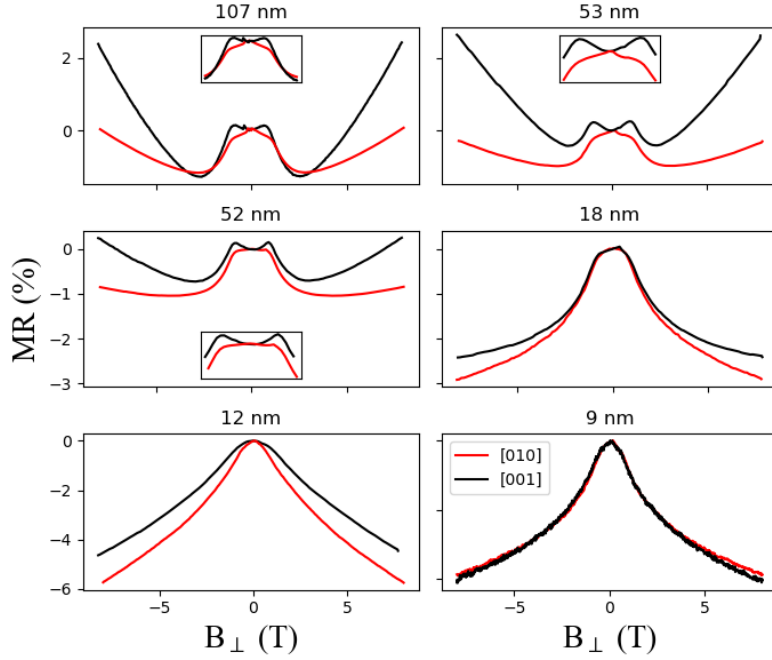


Figure 41: Magnetoresistance measurements replotted along the b [010] and c [001] current directions at 3.5 K for CrO_2 films of varying thicknesses.

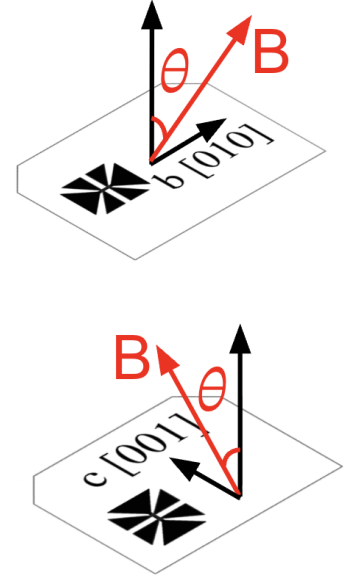


Figure 42: Orientation of magnetic field, B , relative to in-plane crystallographic axes of CrO_2 .

film with current applied along the [001] and [010] crystallographic directions. The focus was on the out-of-plane angular dependence of the anisotropic magnetoresistance (AMR), defined as

$$\text{AMR}(\theta) = \frac{\text{MR}(\theta) - \text{MR}(90^\circ)}{\text{MR}(90^\circ)}, \quad (39)$$

where θ is the angle between the applied magnetic field $\mathbf{B}$ and the film's surface normal.

As $\mathbf{B}$ is rotated from an in-plane orientation parallel to the current to an out-of-plane configuration perpendicular to the current at $T = 1.8$ K, the AMR reaches its maximum value of approximately 2.4% near $\theta = 0^\circ, 180^\circ$, and 360° , with minima at $\theta = 90^\circ$ and 270° . This produces a characteristic four-lobed "bow-tie" or butterfly-like pattern. This AMR magnitude is among the highest reported for epitaxial CrO_2 films, highlighting the importance of spin-dependent scattering in this highly spin-polarized system [40].

This behavior suggests a strong link between the electronic band structure and spin-dependent scattering mechanisms. Three-fold band degeneracy points (i.e. TPs) can enhance the SOC via increased orbital hybridization, which promotes anisotropic spin-dependent scattering and contributes to the observed AMR. Single-crystalline CrO_2 has been reported to exhibit strong direction-dependent SOC along the [010] and [001] axes [56], and the intrinsic anisotropy of the

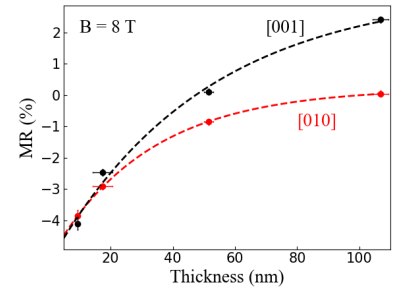


Figure 43: Summary of magnetoresistance values measured at $B_\perp = 8$ T and $T = 3.5$ K along the b [010] and c [001] current directions.

Fermi surface near TPs likely plays a key role in the AMR patterns [57].

In conventional metals, scattering between states is restricted by momentum and energy conservation. Near a TP, the unique band topology opens additional scattering channels, which can either enhance or suppress certain processes, giving rise to anisotropic transport.

The evolution from a sharp, cusp-like feature in the AMR at low fields (< 2 T) to a double-humped shape at higher fields likely reflects the strong magnetic anisotropy between the in-plane ($[010]$ and $[001]$) and out-of-plane ($[100]$) directions. VSM measurements along these orientations were performed on the bulk, unpatterned 53 nm film to illustrate this directional anisotropy when tilting the magnetic field out of the plane. The low-field VSM results highlight the directional magnetic anisotropy of the CrO_2 film and the associated variations in the DOS and scattering pathways. The differences in the MH loops along the $[010]$ and $[001]$ in-plane directions highlight the anisotropy observe in other measurements.

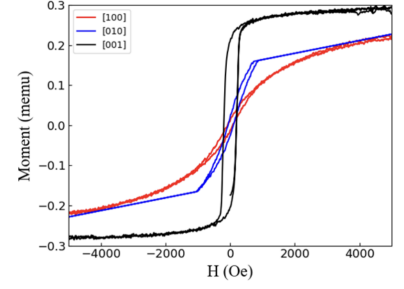


Figure 44: VSM analysis of the unpatterned 53 nm CrO_2 film along the $[001]$, $[010]$, and $[100]$ crystal orientations.

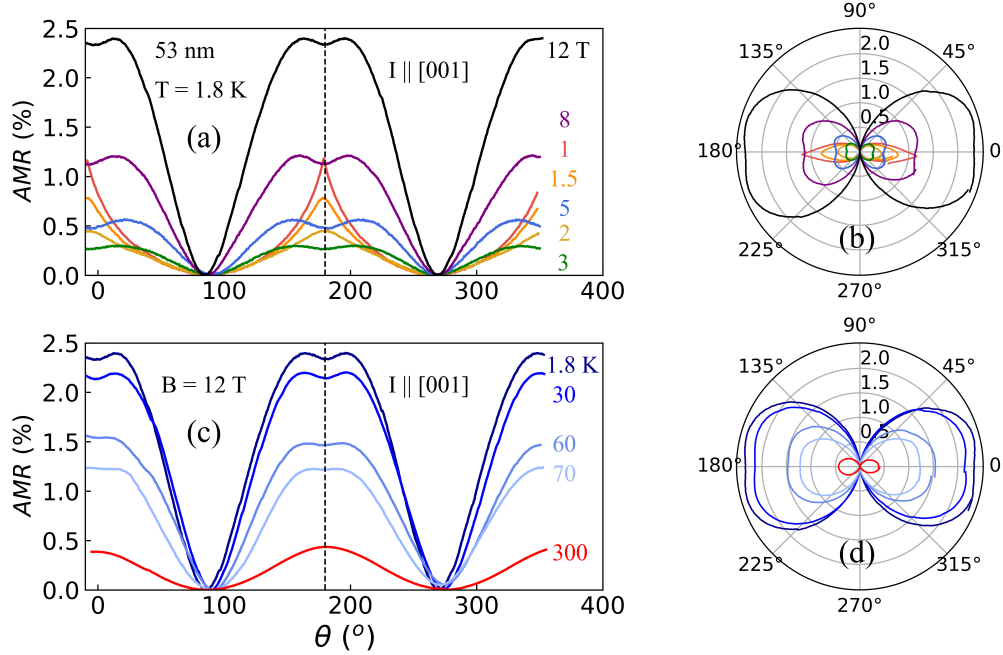


Figure 45: Anisotropic magnetoresistance (AMR) along $[001]$. (a) Field-dependent AMR at 1.8 K with polar plot representation in (b). (c) Temperature-dependent AMR at 12 T with corresponding polar plot in (d). Dashed lines at $\theta = 180^\circ$.

The AMR of CrO_2 exhibits a strong dependence on the applied magnetic field, reflecting both magnetic anisotropy and electronic structure effects. At low fields (below 2 T), the AMR is primarily governed by the magnetic anisotropy imposed by the crystal symmetry. This is observed as a sharp, elongated teardrop-like profile in the angular polar plot, highlighting the strong directional dependence

of scattering. At higher fields (above 3 T), the electronic structure increasingly dictates the AMR response. As shown in the earlier schematic, the electron Fermi surface of CrO_2 forms a pillbox around the zone center, and the $k_x k_z$ cross-section closely resembles the bow-tie AMR pattern seen experimentally at high fields. Slight deviations from this idealized shape arise from in-plane warping of the Fermi surface induced by the applied field which modifies the available electronic states.

The [001] crystallographic direction in CrO_2 shows enhanced spin-dependent scattering, which can be linked to a higher projected DOS (PDOS) along this axis. Both theoretical calculations and experimental results indicate that [001] is the easy magnetization direction, correlating with an increased number of electronic states at the Fermi level [58]. This abundance of states facilitates spin-dependent scattering and leads to a larger AMR amplitude along [001]. In contrast, the [010] direction exhibits a significantly smaller AMR, approximately one-third of that along [001]. This suppression is associated with a lower PDOS along [010], meaning fewer available scattering sites. Bands that are steeply dispersing along [010] contribute to this effect, as they provide a smaller number of states within a given energy range, resulting in weaker scattering and a reduced AMR response.

Triply degenerate points near the Fermi level further influence the transport properties. TPs enhance magnon-mediated scattering by increasing the PDOS at specific energies, leading to more frequent scattering events. They also strengthen SOC through enhanced orbital hybridization, producing strongly anisotropic spin-dependent scattering that directly impacts the AMR. The interplay between SOC and magnon scattering near TPs also plays a critical role in the temperature dependence of the resistivity [59]. Specifically, double-magnon scattering processes are amplified by the presence of TPs as they increase the DOS at energies that contribute to magnon-mediated transport.

AMR scans performed at high fields (12 T) show that the AMR is strongest at low temperatures, where the conduction electron spin polarization is most robust. With increasing temperature, the AMR magnitude decreases due to weakening of the spin-dependent scattering. The characteristic bow-tie or butterfly-like AMR pattern remains evident, similar to observations in type-II Weyl semimetals that also possess highly-anisotropic electron Fermi surfaces [57].

Angle-dependent AMR measurements along the orthogonal [010] direction confirm that the AMR amplitude is much smaller, reaching a maximum of only $\sim 0.7\%$, about one-third of the [001] response. Despite the reduced amplitude, the general trends and qualitative

polar plot shapes are consistent with those observed along $[001]$. The suppression along $[010]$ can be understood as a consequence of aligning the magnetic field with directions of lower PDOS, reducing the number of available electronic states for scattering and thus decreasing the AMR. In this context, a low-PDOS direction refers to bands that are steeply dispersing, which provide fewer states within a given energy range, resulting in weaker spin-dependent scattering and a smaller AMR signal.

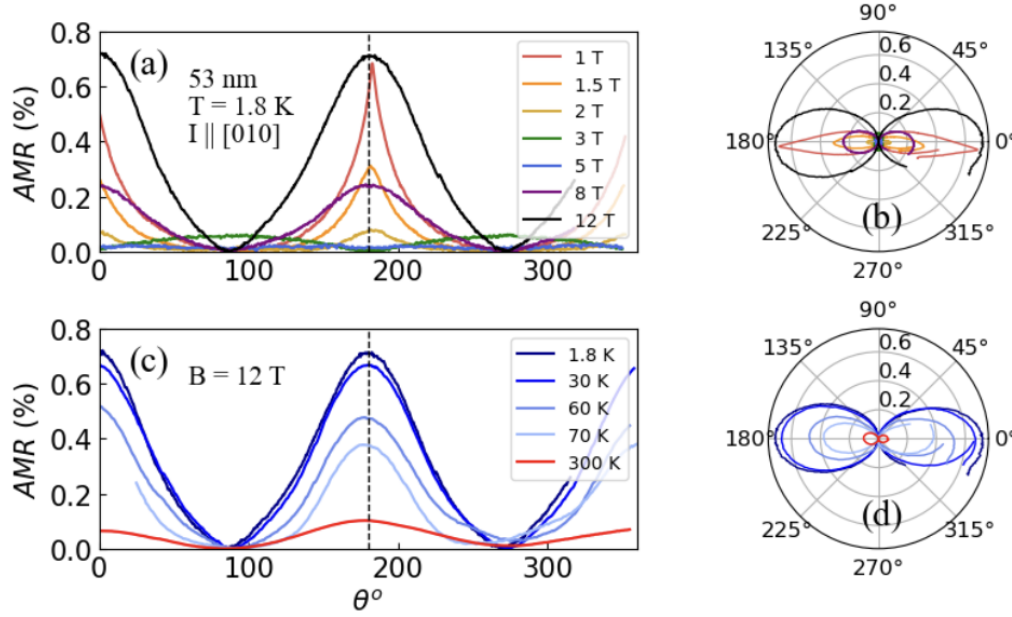


Figure 46: Anisotropic magnetoresistance (AMR) along $[010]$. (a) Field-dependent AMR at 1.8 K with polar plot representation in (b). (c) Temperature-dependent AMR at 12 T with corresponding polar plot in (d). Dashed lines at $\theta = 180^\circ$.

Experimental ferromagnetic resonance (FMR) measurements indicate a large anisotropy of the effective Gilbert damping in CrO_2 [56], supporting the strong directional magnetic anisotropy along the $[010]$ and $[001]$ axes observed in the AMR. Similarly, studies on thermal magnon transport suggest that factors such as next-nearest neighbor hopping, easy-axis anisotropy, and the presence of Dzyaloshinskii-Moriya interactions (DMI), which are manifestations of SOC, stabilize FM order and influence magnon dynamics [60]. These effects are expected to be enhanced by TPs, which amplify SOC-driven, angle-dependent spin scattering.

Field-Dependent Background Magnetoresistance

Longitudinal resistivity measurements were carried out as a function of magnetic field strength on the same 53 nm CrO_2 film at 1.8 K over a broad range of tilt angles. Using Hall-bar devices patterned along

both [010] and [001] crystallographic axes, the MR is determined according to

$$\text{MR}(B) = \frac{R_{xx}(B) - R_{xx}(0)}{R_{xx}(0)}. \quad (40)$$

By extending the magnetic field range compared to earlier reports [39, 40, 47, 61], significantly higher MR values are achieved. A second-order polynomial was fitted to the high-field regime at $\theta = 0^\circ$ and extrapolated back to zero field, yielding a maximum MR of 6.58% [47, 62, 63].

The MR originates from a combination of classical and quantum effects. In the high-field limit, the MR follows a quadratic dependence on the magnetic field ($\propto B^2$), consistent with a classical orbital contribution. In this regime, the cyclotron radius becomes smaller than the electronic mean free path, and the Lorentz force acting on charge carriers produces a non-saturating MR response.

This represents the largest MR reported to date for epitaxial CrO_2 thin films!

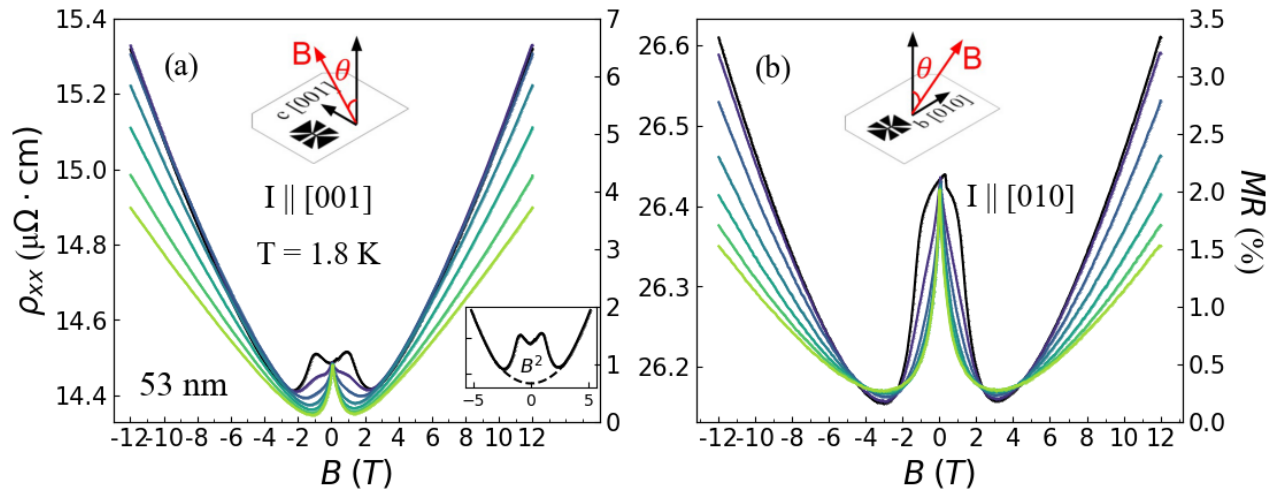


Figure 47: Angle-dependent magnetoresistance (MR) of bulk CrO_2 . MR measurements collected at 1.8 K along (a) [001] and (b) [010]. Dashed line in inset of panel (a) is quadratic fit to $\theta = 0^\circ$ data.

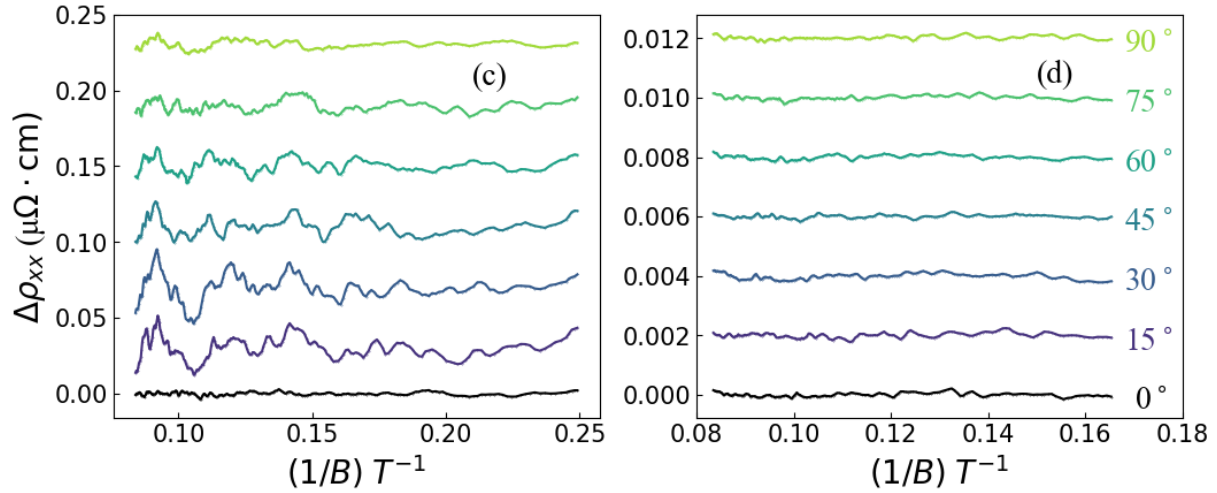
At lower fields, additional quantum contributions emerge due to interference effects in the presence of SOC. Along the [001] current direction, a small positive MR is observed in the range of 1–2%, which can be attributed to weak localization arising from the breaking of time-reversal symmetry by the applied field. This feature is gradually suppressed with increasing magnetic field strength. Such behavior is consistent with spin-dependent weak antilocalization effects previously reported in other HMFs [64, 65].

Oscillatory Behavior in the High-Field Regime

Quantum oscillations in ρ_{xx} are a powerful probe of the Fermi surface, yet they are notoriously difficult to observe in materials with large carrier densities or strong scattering. In the case of CrO_2 , theory has long suggested that Shubnikov–de Haas (SdH) oscillations may be detected given the relatively low carrier concentration and high carrier mobility [47]. However, the observation of such oscillations has remained elusive, in part due to the large Fermi surface which tends to mask the periodic component of the MR.

In the measurements described here, the high crystalline quality of the epitaxial films combined with exceedingly low temperatures and strong applied magnetic fields makes it possible to detect these subtle quantum oscillations. After removing the smoothly varying background MR with a second-order polynomial fit, periodic modulation in the longitudinal resistivity is uncovered, denoted as $\Delta\rho_{xx}$. This data is plotted as a function of inverse magnetic field, as SdH oscillations are typically depicted. The periodicity and field dependence of these oscillations are consistent with SdH behavior, providing direct evidence of Landau level (LL) quantization in CrO_2 .

This constitutes the first clear experimental demonstration of SdH oscillations in a half-metallic ferromagnet!



The oscillations are most clearly resolved when current flows along the $[001]$ axis. Systematic measurements as $\mathbf{B}$ is tilted between $\theta = 0^\circ$ and 90° reveal a pronounced angular dependence. The oscillation amplitude decreases continuously with tilt, with strong suppression both near the out-of-plane (90°) and in-plane (0°) geometries. The reduction at $\theta = 90^\circ$ is expected in thin film systems, where

Figure 48: (c) and (d) illustrate underlying quantum oscillations $\Delta\rho_{xx}$ after subtracting smoothly-varying polynomial background from data in (a) and (b) above.

the Lorentz force acting on carriers becomes ineffective and orbital motion is minimized. More surprising is the suppression at $\theta = 0^\circ$, which suggests that the extremal orbits responsible for the SdH effect are absent when the field is strictly aligned along the symmetry axis. In anisotropic Fermi surfaces, extremal cross-sections occur only at certain field orientations, and if the orbit at $\theta = 0^\circ$ is non-extremal (i.e. not a true minimum or maximum cross-sectional area), quantum oscillations will vanish even though LLs still form.

The observed behavior therefore provides insight into the topology of the CrO_2 Fermi surface. The fact that oscillations appear most strongly at intermediate tilt angles indicates that high-mobility electronic pockets dominate transport away from the high-symmetry orientations. This angular selectivity reflects the warped and anisotropic nature of the Fermi surface, where only certain orbits contribute to quantum oscillations.

From a broader perspective, the MR of CrO_2 is shaped by both classical orbital effects and quantum corrections arising from Landau quantization. Since electronic transport is governed by carriers near the Fermi energy, the angular dependence of ρ_{xx} can be directly tied to the Fermi surface geometry and the DOS along different crystallographic directions. The ability to observe oscillations in these measurements highlights the combination of high film quality, low scattering rates, and the extreme experimental conditions employed.

The anisotropy between crystallographic axes is striking. Along $[001]$, the SdH oscillations are robust, consistent with a smaller effective mass, higher carrier mobility, and a lower scattering rate compared to $[010]$. Along the $[010]$ axis, by contrast, the oscillatory signal is nearly absent. This asymmetry points to a Fermi surface that is strongly direction-dependent: warped in such a way that extremal orbits exist primarily along $[001]$. The presence of subtle subharmonic features even after background subtraction further supports the idea of a complex and anisotropic Fermi surface, consistent with prior theoretical reconstructions of the electronic structure of rutile CrO_2 [53, 66].

Further enhancement of field strength or reduction of temperature would likely amplify the oscillatory component, making it an even more powerful probe of the electronic structure.

The detection of SdH oscillations in CrO_2 not only establishes the presence of Landau quantization in a HMF but also provides new insight into its highly anisotropic electronic structure. The above results emphasize that quantum transport in CrO_2 is dominated by a small number of high-mobility carriers associated with extremal Fermi surface pockets, whose contribution is highly sensitive to the field orientation.

Landau Quantization

To further analyze the oscillatory contribution in $\Delta\rho_{xx}$, a fast Fourier transform (FFT) of the data is performed. At $\theta = 30^\circ$, the spectrum reveals a dominant SdH oscillation frequency of $f = 48 \pm 6$ T, indicated by the dashed guidelines.

In principle, each extremal cross-sectional area of the Fermi surface produces its own oscillation frequency. Since band structure calculations predict a rather intricate Fermi surface for CrO_2 , one might expect several discrete peaks. However, in this case, only a single well-resolved frequency is visible with indications of additional, unresolved side components. The absence of clearly separated secondary frequencies is attributed to experimental limitations in the lowest temperature (1.8 K) and maximum field strength (12 T) achieved.

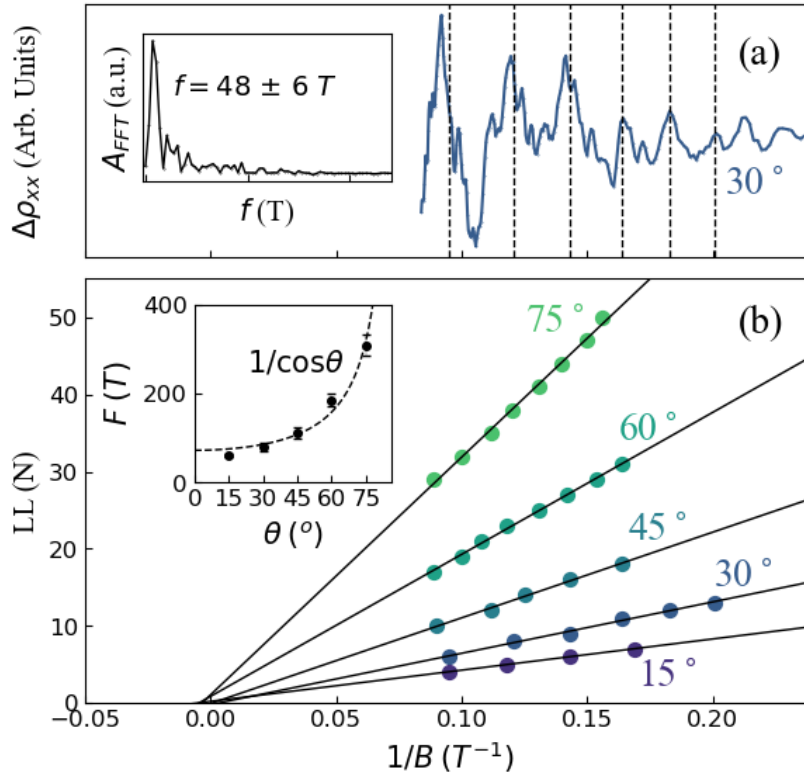


Figure 49: Analysis of Shubnikov-de Haas (SdH) oscillations. (a) Data along [001] after background subtraction at $\theta = 30^\circ$. Inset shows corresponding FFT spectrum. (b) Landau level fan diagram of peak positions (indicated by dashed lines in (a)) at various tilt angles. Linear fits with slopes F plotted as a function of tilt angle in inset of (b).

The origin of the oscillations is understood within the framework of Landau quantization, where electronic states condense into discrete LLs in a magnetic field. To analyze these levels, the peak positions of $\Delta\rho_{xx}$ are extracted as a function of inverse field, $1/B$ and used to construct LL fan diagrams for several tilt angles. The linear

Extending measurements to even lower temperatures and stronger magnetic fields should enhance the oscillation contrast and may reveal further frequency components predicted by band structure calculations.

fits are described by the simplified expression

$$N = \frac{F}{B} + \frac{\phi_B}{2\pi} - \delta, \quad (41)$$

where F denotes the oscillation frequency, N is the LL index number, and δ encodes any relevant phase shifts.

The intercepts of these LL linear fits provide insight into the Berry phase ($\phi_B = 2\pi\beta$), where $\beta = 1/2$ corresponds to a topologically non-trivial Berry phase ($\phi_B = \pi$) associated with Dirac or Weyl fermions. Using the convention in [67], we conclude a mixed 2D- and 3D-like Fermi surface, where carriers accumulate a Berry phase as they traverse closed orbits [57]. The nontrivial Berry phase extracted from the LL fan diagram points to additional topological corrections to the MR.

The sharpness of the observed LLs further underscores the high crystalline quality and low impurity content of the 53 nm epitaxial film studied here. The presence of TPs and spin-polarized conduction in CrO₂ could introduce Berry-curvature-driven transport anisotropies, further modifying MR behavior.

Angle-Dependent Frequency Analysis

The angular dependence of the oscillation frequency F is explored next. As the tilt angle is increased from $\theta = 15^\circ$ to $\theta = 75^\circ$, the SdH oscillation frequency shifts from $f = 41 \pm 3$ T to 308 ± 22 T, following a trend consistent with $F \propto 1/\cos \theta$. This scaling indicates that the oscillations are governed primarily by the perpendicular component of the magnetic field $B_\perp = B \cos \theta$, which is typical of quasi-2D transport. In a perfectly 2D system, $F(\theta)$ would diverge as $\theta \rightarrow 90^\circ$, when the field is fully in-plane. In the measurements shown here, however, finite oscillations persist even at $\theta = 90^\circ$, demonstrating that the system retains strong three-dimensional bulk contributions.

This angular response reflects the highly anisotropic Fermi surface of CrO₂. Elongated pockets along the [001] direction can generate quasi-2D oscillation signatures as the magnetic field is tilted away from high-symmetry axes. Although the film thickness exceeds typical confinement length scales, partial confinement effects cannot be excluded. Instead, the dominant mechanism is more plausibly the intrinsic Fermi surface anisotropy. The persistence of the SdH signal across a wide range of θ combined with residual oscillations at full in-plane fields strongly supports the conclusion that the oscillations are bulk in origin rather than arising from surface states [67, 68].

Thermal Damping

Beyond the angular dependence of ρ_{xx} , the temperature sensitivity of the SdH oscillations is also studied. At the base measurement temperature of 1.8 K, oscillatory features are clearly resolved. As the temperature is increased, however, the oscillation amplitude diminishes rapidly and becomes indiscernible. Even with advanced signal processing and careful background subtraction, no reproducible oscillatory features could be detected at higher temperatures, indicating that thermal broadening of LLs is the dominant suppression mechanism. In the framework of Lifshitz-Kosevich (LK) theory, this thermal damping is captured by the factor

$$R_T = \frac{\alpha T m^* / B}{\sinh(\alpha T m^* / B)}, \quad (42)$$

where $\alpha = 2\pi^2 k_B / (\hbar e)$, m^* is the cyclotron effective mass, T is the temperature, and B is the applied magnetic field. As the temperature increases, smearing of the Fermi-Dirac distribution reduces the contrast between filled and empty LLs, leading to a rapid suppression of the oscillation amplitude.

Extraction of Material Parameters

In practice, the LK formalism provides a powerful tool for extracting the carrier effective mass. By measuring the oscillation amplitude at a fixed magnetic field and fitting its decay with $R_T(T)$, one can determine m^* with high accuracy. However, in the case of CrO₂, the oscillations become indiscernible already above $T = 1.8$ K, making such an analysis challenging with the present dataset. This rapid suppression nevertheless implies that the contributing extremal orbits correspond to carriers with a relatively small effective mass, which makes them especially sensitive to thermal broadening.

To quantify these oscillations at 1.8 K, a modified thermodynamic LK fit of the normalized oscillatory component $\Delta\rho_{xx}/\rho_{xx}(0)$ for $\theta = 30^\circ$ along [001] is utilized [68–71]. It can be expressed as

$$\frac{\Delta\rho_{xx}}{\rho_{xx}(0)} \propto \sqrt{\frac{B}{2F}} \frac{\alpha T m^*}{m_0 B \sinh\left(\frac{\alpha T \mu}{B}\right)} \exp\left(-\frac{\alpha T_D m^*}{B m_0}\right) \cos\left(\frac{\alpha g m^*}{2m_0}\right) \cos(2\pi N), \quad (43)$$

where m_0 is the free electron mass, k_B is Boltzmann's constant, $\hbar$ is the reduced Planck's constant, and g is the electron g -factor. The Dingle temperature T_D captures the effects of impurity scattering.

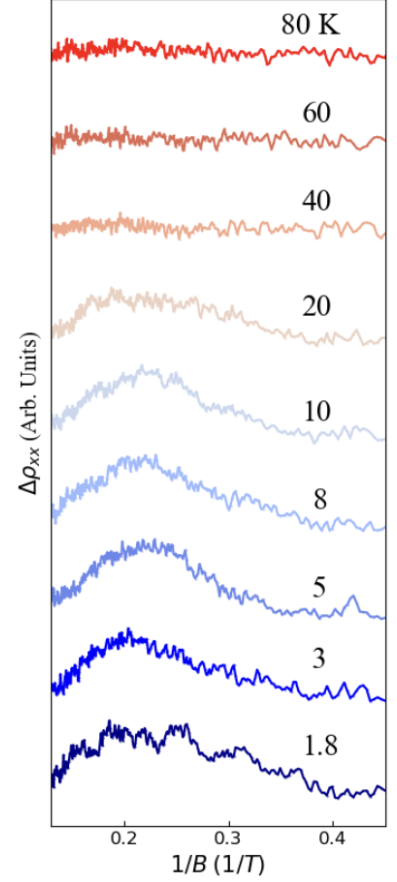


Figure 50: Temperature dependence of SdH oscillations. Low-temperature characterization of a 53 nm sample from 1.8 K to 80 K at a fixed angle of $\theta = 17^\circ$. Data shown is post-background subtraction.

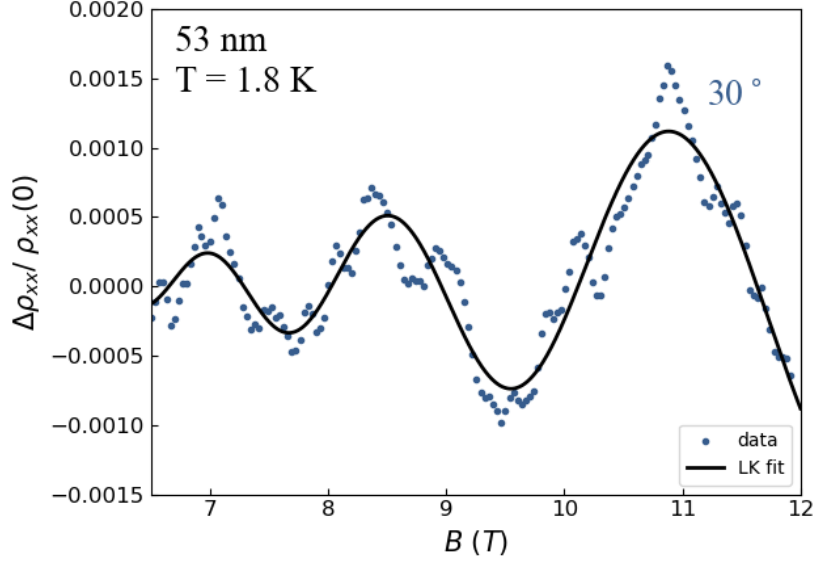


Figure 51: Magnetic field-dependent data along [001] at $\theta = 30^\circ$ fit to the modified Lifshitz-Kosevich (LK) model.

This fit allows extraction of several key physical parameters. The scattering time is determined from $\tau = \hbar/(2ek_B T_D)$, which in turn enables calculation of the carrier mobility $\mu = e\tau/m^*$ and mean free path $l = \tau v_F$. The fit also provides Fermi surface statistics including the Fermi wavevector $k_F = \sqrt{2eF/\hbar}$, Fermi velocity $v_F = \hbar k_F/m^*$, and the cross-sectional area of the Fermi pocket $A_F = \pi k_F^2$. The values obtained from these fits are in good agreement with both theoretical predictions and other experimental studies performed using angle-resolved photoemission spectroscopy (ARPES) [72, 73], further corroborating the high quality and low impurity content of the CrO_2 films.

Parameter	Symbol	Measured	Units
Oscillation frequency	f_{SDH}	48 ± 6	T
Effective mass	m^*	0.53 ± 0.05	m_0
Scattering time	τ	$(3.3 \pm 0.8) \times 10^{-13}$	s
Dingle temperature	T_D	3.6 ± 0.5	K
Carrier mobility	μ	0.12 ± 0.02	m^2/Vs
3D carrier density	n_{3D}	$(2.8 \pm 0.4) \times 10^{23}$	cm^{-3}
Fermi wave vector	k_F	1.21 ± 0.08	$\text{\AA}^{-1}$
Fermi velocity	v_F	$(0.82 \pm 0.13) \times 10^5$	m/s
Fermi pocket area	A_F	4.6 ± 0.6	$\text{\AA}^{-2}$
Mean free path	l	27 ± 8	nm
Dimensionless parameter	$\omega_c \tau$	1.31 ± 0.34	-

Table 3: Summary of extracted material parameters for CrO_2 from Lifshitz-Kosevich fit.

The Dingle temperature T_D provides a measure of the damping of SdH oscillations due to scattering and captures the effective thermal broadening of LLs. In CrO₂, the extracted Dingle temperature is relatively low compared to other materials exhibiting SdH oscillations [67, 69, 70], indicating minimal thermal broadening. This property is particularly advantageous for applications that require thermal stability across a wide temperature range including optical and electronic devices. The mean free path derived at 1.8 K is consistent with prior experimental measurements at 2 K [23, 33, 40, 62, 63] and aligns with theoretical predictions for single-crystal CrO₂ at 0 K [8], further corroborating the high quality of the films.

The scattering time τ , determined from T_D , lies within the expected range of 10^{-14} to 10^{-12} s that is typical of transition metal oxides [41]. Likewise, the electron effective mass m^* extracted from these fits agrees with theoretical calculations at high-symmetry points on the Fermi surface [33, 42, 53, 74]. To further characterize quantum transport, the cyclotron frequency $\omega_c = eB/m^*$ is computed, which describes the orbital motion of electrons in a magnetic field. In the quantum regime, where $\omega_c\tau \gg 1$, electrons complete multiple cyclotron orbits before scattering, giving rise to well-resolved SdH oscillations. The clear observation of such oscillations in the data confirms that $\omega_c\tau > 1$ for the relevant carrier pockets in CrO₂.

Carrier mobility in bulk CrO₂ thin films has been previously reported to reach several hundred cm²/Vs at low temperatures [33, 41, 47]. The extracted mobility values fall within this expected range, consistent with the observation of strong SdH oscillations, since higher mobility directly enhances oscillation amplitude. Using the standard single-band model $n = k_F^3/3\pi^2$, with k_F obtained from the SdH frequency assuming a spherical Fermi surface, the carrier density can be estimated. This approach provides an approximate upper bound because it neglects anisotropic Fermi surface effects, which have been theorized to exist in CrO₂ and are shown experimentally in the angular-dependent measurements above. Therefore, comparisons to carrier densities derived from more complex multi-band analysis should be made cautiously [47].

The findings provide insight into the longitudinal MR, which exhibits a positive B^2 scaling consistent with classical orbital contributions. The extracted 3D carrier density exceeds previously reported values [66], in agreement with the enhanced DOS along [001] and the pronounced visibility of SdH oscillations. Additionally, the Fermi wave vector [75] and Fermi velocity [10, 44, 63, 76] derived from the SdH analysis are in excellent agreement with theoretical calculations

and prior experimental reports. The consistency between these extracted parameters and earlier studies validates the modified LK fitting procedure and demonstrates its reliability for characterizing the electronic structure and quantum transport properties of high-quality CrO_2 thin films.

Key Findings

This study marks the first experimental observation of Shubnikov–de Haas (SdH) oscillations in a half-metallic oxide, marking a significant step in probing quantum transport in ferromagnetic oxides beyond the ultra-thin limit. These oscillations, observed even in bulk films, establish CrO_2 as a promising platform for exploring quantum phenomena in half-metallic ferromagnets. Complementary measurements reveal temperature-dependent double-magnon scattering and pronounced, axis-dependent AMR, consistent with strong SOC and band anisotropy. The results are quantitatively described by the Lifshitz–Kosevich formalism and Landau level indexing, yielding material parameters that closely match previous studies. Together, these findings underscore CrO_2 's unconventional electronic structure and highlight its potential as a model system for spintronic and half-metallic quantum materials research.

Looking Ahead

The transport behavior of CrO_2 offers more than just a window into its electronic structure, it represents a platform for generating highly spin-polarized currents at the source. The half-metallic nature of CrO_2 makes it ideal for probing spin-dependent transport phenomena and for injecting spin currents into adjacent layers. These findings position CrO_2 as a model system for exploring how spin-polarized carriers propagate, interact, and couple across interfaces. This is a central theme that motivates the following discussion on interfacial exchange and spin transmission in $\text{RuO}_2/\text{CrO}_2$ heterostructures.

Chapter 4 | Interfacial Exchange & Spin Coupling in $\text{RuO}_2/\text{CrO}_2$

INTERFACES BETWEEN antiferromagnetic and ferromagnetic oxides provide a versatile platform for exploring interfacial spin coupling, exchange phenomena, and spin-orbit effects, which are central to the design of next-generation spintronic devices. In the context of the spin current pipeline, where CrO_2 serves as a robust source of highly spin-polarized carriers, $\text{RuO}_2/\text{CrO}_2$ bilayers allow us to probe how these spin currents interact, transmit, and are modified at interfaces. By combining a half-metallic ferromagnet with an antiferromagnetic oxide, this system provides a controlled environment to study the interplay between spin polarization, magnetic anisotropy, and interfacial symmetry. We gain insight into how these factors govern spin alignment, exchange interactions, and magnetization dynamics.

Magnetization measurements reveal a distinct exchange bias shift in the hysteresis loops of CrO_2 induced by the RuO_2 capping layer. This demonstrates unidirectional interfacial coupling that is strongly temperature dependent and disappears well below the Néel temperature of RuO_2 . Zero-field-cooled (ZFC) and field-cooled (FC) magnetization measurements show magnetic irreversibility and low-temperature bifurcation, indicative of interfacial spin freezing or reorientation. A pronounced field-dependent kink in the high-temperature regime reflects competing exchange interactions. Furthermore, notable deviations from Bloch's law in the saturation magnetization further point to modified spin-wave excitations and enhanced magnon scattering across this interface.

Taken together, these observations highlight the critical role of interfacial exchange and SOC in mediating the transmission of spin-polarized currents from the ferromagnet to the adjacent oxide layer. By providing a tunable and well-characterized platform for studying spin-orbit-driven interfacial phenomena, $\text{RuO}_2/\text{CrO}_2$ bilayers bridge the gap between intrinsic spin generation in CrO_2 and spin-charge conversion in heavy-metal systems.

MOTIVATION

Ruthenium dioxide (RuO_2) has recently attracted significant attention as a correlated metallic oxide that combines high electrical conductivity, chemical stability, and rich SOC effects. Unlike conventional noble metals, RuO_2 exhibits an unconventional collinear antiferromagnetic ground state with compensated spin moments on the Ru sublattices, yet it remains metallic at room temperature.

This duality of metallic conduction coexisting with antiferromag-

netic order places RuO_2 among a new class of itinerant AFMs that have been proposed as active layers in SOT and exchange-bias heterostructures [77]. Its noncentrosymmetric rutile crystal structure further allows for moment canting and anisotropic spin transport, which can give rise to phenomena such as altermagnetism and symmetry-protected spin polarization even in the absence of net magnetization.

A Controversial Altermagnet

The rutile-structured oxide has attracted significant attention in spintronics because it was proposed to belong to the emergent class of altermagnets (AMs). These are materials that combine zero net magnetization (like AFMs) with spin-band splitting (like FMs). In one line of work, theoretical symmetry-analysis and experiments such as ARPES claimed that RuO_2 exhibits momentum-dependent spin band splitting and time-reversal symmetry breaking in its electronic structure, which are characteristics of altermagnetic ordering [13, 79]. Earlier neutron diffraction studies even inferred small antiferromagnetic moments and lattice distortions that seemed to support itinerant antiferromagnetism up to at least 300 K [80, 81].

Recent studies with more sensitive bulk probes such as polarized neutron diffraction, muon spin-rotation, and terahertz spectroscopy, however, have found no evidence for long-range magnetic order or significant local moments in stoichiometric bulk RuO_2 crystals [82–84]. These studies argue that earlier signals may have arisen from defects, secondary phases, or misinterpreted diffraction features. The discrepancy has led to a reassessment of RuO_2 's status. Rather than a prototypical AM, it may instead be a nonmagnetic metal whose spin-splitting effects may stem from subtle structural or surface-confinement effects.

The current consensus is that while RuO_2 remains a very interesting material owing to its metallicity, high crystalline quality, and compatibility with spintronic heterostructures, the claim of intrinsic altermagnetic order in the bulk remains unproven and highly contested. This debate has in fact sharpened research interest: on the one hand to clarify whether altermagnetism can be realized in metallic oxides at all, and on the other to explore how strain, dimensional confinement, doping or interface engineering might drive RuO_2 , or RuO_2 -derived systems, into an altermagnetic or spin-polarized regime.

Extending the work on CrO_2 , this chapter investigates the integration of RuO_2 with CrO_2 which offers a unique model system where a metallic AFM is exchange-coupled to a HMF, a configuration rarely

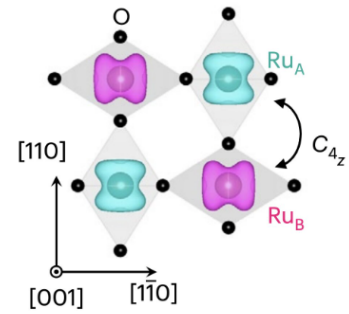


Figure 52: Spin density isosurfaces of RuO_2 . Black atoms are oxygen; antiferromagnetically-ordered Ru atoms in blue and pink [78].

explored experimentally.

In this bilayer geometry, the RuO₂/CrO₂ interface can mediate an (EB) effect, observable as a horizontal shift in the magnetic hysteresis loop. Such a shift arises from interfacial exchange coupling between the uncompensated spins at the RuO₂ surface and the ferromagnetic moments in CrO₂. This coupling effectively breaks time-reversal symmetry at the interface, leading to a unidirectional anisotropy that pins the CrO₂ magnetization during reversal. Importantly, the temperature dependence and magnitude of this bias field provide microscopic insight into the nature of the interfacial spin alignment and the magnetic order in RuO₂. Observing a measurable exchange bias in this system is thus highly significant, not only as a confirmation of magnetic exchange coupling between an itinerant AFM and a half-metal, but also as supporting evidence to push for RuO₂-based heterostructures in all-oxide spintronics and SOT devices.

Origins of Exchange Bias

The exchange bias (EB) effect arises in heterostructures that combine a FM and an AFM through interfacial coupling. When such a bilayer is cooled through the Néel temperature (T_N) of the AFM in the presence of an external magnetic field, exchange interactions at the FM/AFM interface lead to a preferential orientation of the interfacial AFM spins. This interfacial spin alignment creates a unidirectional anisotropy that manifests macroscopically as a shift in the MH loop. In practice, the EB effect stabilizes one direction of magnetization, enabling applications such as magnetic memory pinning, spin-valves, and tunneling magnetoresistance (TMR) sensors.

At the microscopic level, the exchange bias originates from interfacial exchange coupling energy between uncompensated AFM spins and the FM spins at the interface. The coupling energy density can be expressed as

$$E_{\text{ex}} = -J_{\text{int}} \mathbf{M}_{\text{FM}} \cdot \mathbf{M}_{\text{AFM}} \quad (44)$$

where J_{int} is the interfacial exchange constant and $\mathbf{M}_{\text{FM}}$ and $\mathbf{M}_{\text{AFM}}$ represent the local magnetization vectors of the FM and AFM layers, respectively. The sign and magnitude of J_{int} determine whether the exchange coupling is ferromagnetic or antiferromagnetic, and thus whether the resulting bias is positive or negative. The existence of uncompensated spins at the AFM interface, which comes from surface roughness, atomic steps, or spin canting, is crucial for establishing the bias.

The temperature dependence of the exchange bias provides valuable insight into the interfacial spin structure and the strength of

coupling. The EB field typically decreases with increasing temperature and vanishes above a characteristic temperature known as the blocking temperature, T_B . This temperature represents the point at which the AFM spin structure becomes thermally unstable and can no longer maintain a fixed orientation relative to the FM layer.

In general, T_B lies below T_N of the AFM since only a subset of interfacial spins contribute to the bias effect. There are several reasons for this:

- T_N is an intrinsic property of the AFM while T_B depends on the stability of the AFM/FM interfacial spin configuration under thermal stress. Hence, even if the AFM remains ordered up to T_N , the exchange coupling to the FM may collapse earlier.
- In the RuO₂/CrO₂ bilayer, EB originates from interfacial exchange coupling between Ru⁴⁺ moments and the half-metallic FM Cr⁴⁺ sublattice. Spin frustration or canting at the interface can cause the spins to become thermally unstable. The strong ferromagnetic ordering of CrO₂ relative to the weak itinerant AFM of RuO₂ can also cause spin dragging that can destabilize AFM pinning when heated.
- In particularly thin RuO₂ films below 30 nm, strain from lattice mismatch ($\approx 2\text{--}3\%$ with CrO₂) can suppress the AFM order parameter. Interfacial roughness and further weaken the exchange coupling. This finite-size effect can push T_B down to 150–250 K, even if the intrinsic T_N of RuO₂ is closer to 400 K.

In the limit that $T \ll T_B$, the interfacial spins in the AFM are rigidly pinned, resulting in a maximum bias field. As temperature increases, thermal activation allows partial reorientation or depinning of these spins, gradually reducing H_{EB} . Near T_B , stochastic spin fluctuations dominate, leading to the complete disappearance of the bias. This behavior can be described by a power-law decay:

$$H_{EB}(T) \propto \left(1 - \frac{T}{T_B}\right)^m \quad (45)$$

where the exponent m reflects the origin and mechanism of the interfacial coupling and spin anisotropy [85, 86].

Bilayer Architecture

The RuO₂/CrO₂ bilayers of interest were fabricated on single-crystal TiO₂ substrates with a (111) orientation.

Very thin RuO₂ films, 10 nm in thickness, were deposited using magnetron sputtering of a Ru target in an oxygen-rich environment

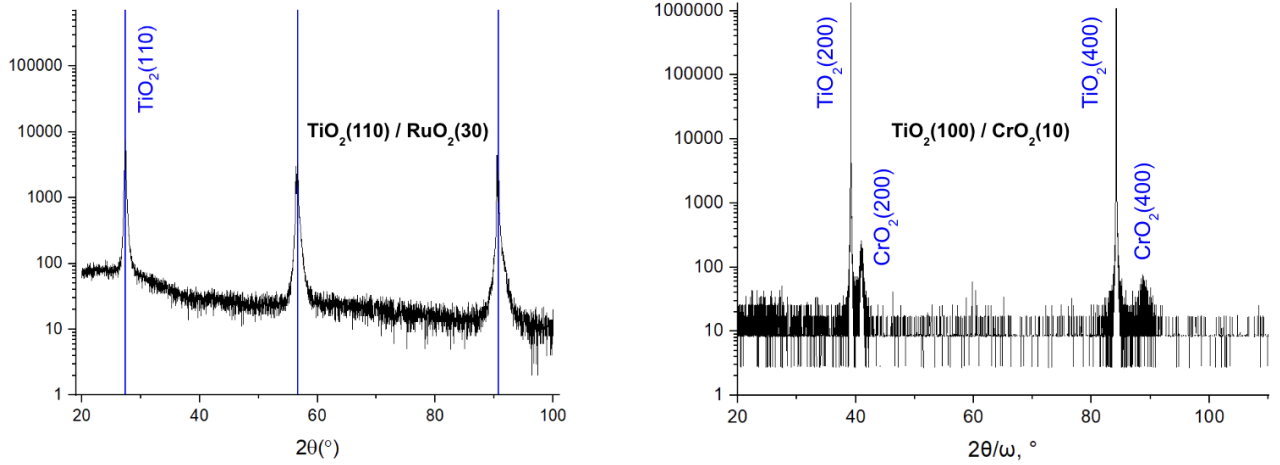


Figure 53: (Left) XRD scans of 30 nm RuO₂ film sputtered on double-side polished (DSP) (110) TiO₂ substrate. (Right) 10 nm CrO₂ film deposited via CVD on single-side polished (SSP) (100) TiO₂. Clear peaks indicate good epitaxial film quality.

by our collaborators at the University of Delaware. They confirm high-quality epitaxial growth of the RuO₂ films with XRD.

Once received, the CrO₂ film is deposited on top via CVD. To ensure that the interfacial exchange bias effect can be resolved with the PPMS, the thickness of CrO₂ was also chosen to be on the order to 10 nm. This is a key design consideration for observing a measurable and tunable EB in such bilayer systems. The EB field arises from interfacial exchange coupling, and only a small fraction of spins at the interface contribute to H_{EB} . The bulk magnetic behavior of the layers can dominate and dilute the interfacial effects if the layers are too thick. Thinner RuO₂ films supports single-domain-like AFM ordering that can couple more coherently to CrO₂. The EB field roughly scales as:

$$H_{EB} \propto \frac{J_{int}}{M_s t_{FM}} \quad (46)$$

where t_{FM} is the thickness of the FM. Following this, a thinner CrO₂ layer leads to a larger shift because the interfacial coupling energy is distributed over a smaller magnetic volume. A layer too thin, however, may lose the necessary long-range FM ordering. Strain coupling also promotes stronger, more temperature-dependent exchange coupling. Since both materials are rutile oxides of similar lattice constants with a slight mismatch of about 2%, thinner films will have more pronounced strain than thicker ones. Finally, the blocking temperature goes as

$$T_B \propto K_{AFM} t_{AFM} \quad (47)$$

where K_{AFM} is the anisotropy energy. It follows from this relation that if RuO₂ is too thin, T_B approaches T_N , and if too thin, T_B drops rapidly due to unstable AFM ordering.

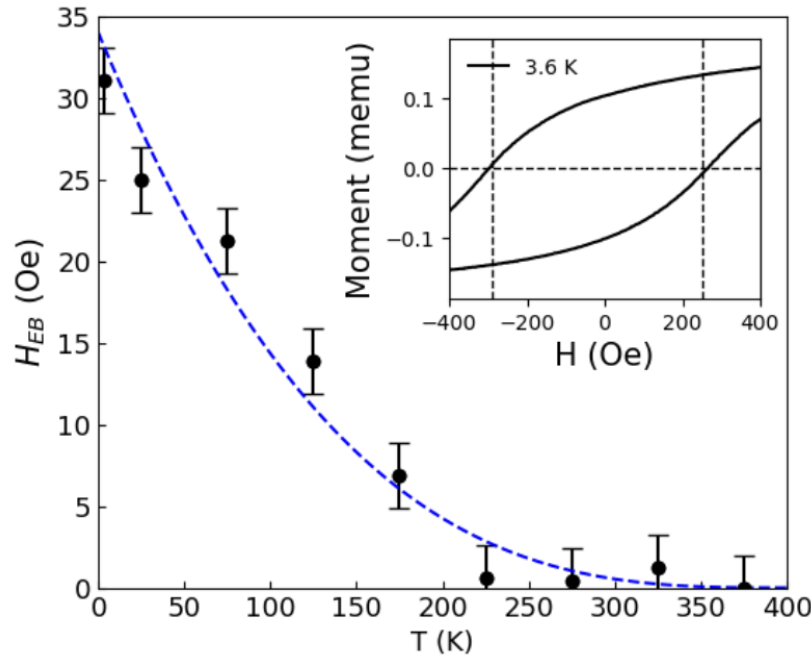


Figure 54: Temperature-dependent exchange bias in (111) $\text{RuO}_2(10\text{nm})/\text{CrO}_2(10\text{nm})$ bilayer. Inset illustrates the largest observed bias at 3.6 K.

Temperature-Dependent Exchange Bias

In the $\text{RuO}_2/\text{CrO}_2$ bilayer system, the temperature-dependent EB provides a sensitive probe of the magnetic ordering in RuO_2 and its coupling to the half-metallic CrO_2 layer. Experimentally, a non-negligible, temperature-dependent H_{EB} is observed. The maximum EB is approximately 30 Oe, with a blocking temperature of $T_B \approx 250$ K, which is consistent with the prediction that $T_B < T_N$. This points to the presence of robust spin correlations or weak ferromagnetic canting in the RuO_2 layer.

A power-law decay exponent of $m = 3$ is observed, which is highly unusual and a very significant result for a FM/AFM coupled system. Such a high exponent indicates a rapid suppression of interfacial exchange coupling with increasing temperature, consistent with strong thermal fluctuations in the interfacial spin structure. This behavior suggests that the RuO_2 layer exhibits short-range or frustrated AFM order that becomes unstable as the thermal energy approaches the anisotropy energy of the AFM domains.

The large m value further implies that the EB originates from a nonuniform interfacial spin configuration, likely influenced by finite-size effects and reduced dimensionality in the thin RuO_2 layer. Similar temperature dependencies have been reported in systems like $\text{LaSrMnO}_3/\text{SrMnO}_3$ [87], $\text{LaCaMnO}_3/\text{CaMnO}_3$ [88], and

The size of the error bars in this plot reflects the resolution of the PPMS.

The exchange bias field was measured after an oscillating 3 Tesla degaussing procedure to ensure that no trapped remnant fields from the PPMS chamber contributed to this effect.

NiMn/CoFeB [89]. These studies report spin-glass-like freezing at the bilayer interfaces, with exchange bias emerging below the interfacial freezing temperature and well below T_N . The magnitude of the exchange bias is further influenced by the training effect and by interfacial frustration and strain. These findings support the interpretation that the RuO₂/CrO₂ interface hosts a complex exchange environment rather than an ideal AFM/FM coupling.

As the temperature increases, these correlations diminish, leading to the disappearance of the bias field. The sharpness of this transition and the overall magnitude of H_{EB} thus reveal crucial information about the interfacial exchange strength, the degree of spin compensation in RuO₂, and the thermal stability of the exchange coupling. In this bilayer, the extracted $m = 3$ exponent suggests a strongly coupled interfacial exchange regime, where the AFM RuO₂ spins exhibit rigid pinning of the CrO₂ FM layer. This behavior typically arises from enhanced interfacial anisotropy or spin canting, leading to a more abrupt decay of the exchange bias field with temperature compared to systems dominated by weaker coupling (e.g. $m < 2$). Below is a summary of model systems, their power-law exponents, and the typical underlying mechanisms:

FM/AFM Bilayer	Power-law Exponent	Possible Mechanisms
Co/CoO	0.5-1	Weak interfacial coupling Uniaxial AFM domain behavior
NiFe/FeMn	1-2	Moderate spin frustration Thermally activated reversal
Fe/IrMn	2-3	Strong interfacial exchange Anisotropy coupling
RuO ₂ /CrO ₂ (this work)	3	Rigid exchange coupling Interfacial spin pinning AFM spin canting

Table 4: Model systems exhibiting temperature-dependent exchange bias [1-3].

Trends in Coercive Field

In addition to analyzing H_{EB} , the coercive field also exhibits a pronounced temperature dependence. The decay of H_c is more gradual and diminishes only at high temperatures, close to the T_C of CrO₂. The values of H_c can be extracted with much higher precision than those of H_{EB} due to a significantly larger magnitude of the effect. The H_c often shows the opposite trend of H_{EB} , peaking near T_B as thermal energy facilitates domain-wall pinning and reversal asymmetry.

In exchange-biased heterostructures like RuO₂/CrO₂, the coercive field H_c reflects the strength of pinning and the energy barrier for

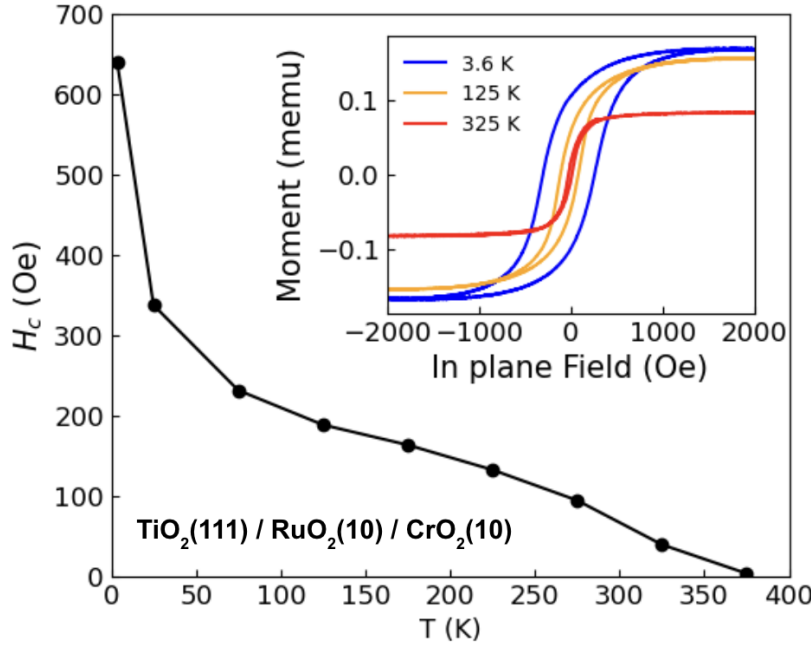


Figure 55: Temperature-dependent coercive field of (111)RuO₂/CrO₂ bilayer. Inset compares MH loops at three different temperatures to show variation in H_c .

magnetization reversal. Its temperature dependence typically differs from that of H_{EB} because:

- Different origins: H_{EB} arises from unidirectional exchange anisotropy at the FM/AFM interface whereas H_c arises from domain wall pinning and magnetocrystalline anisotropy of the FM layer, which are both temperature-dependent.
- Persistence of interfacial pinning: Even after H_{EB} vanishes (when interfacial exchange becomes thermally randomized near the T_B), local AFM order or uncompensated spins can still pin domain walls. This leads to finite coercivity up to the T_C of CrO₂, even though H_{EB} becomes negligible.
- H_c follows a similar power-law decay:

$$H_c(T) \propto \left(1 - \frac{T}{T_C}\right)^n \quad (48)$$

where n depends on the anisotropy and domain behavior of the system. The slower decay compared to $H_{EB}(T)$ reflects that magnetocrystalline anisotropy and pinning energies decrease more gradually than interfacial exchange coupling.

- The large H_c at 2 K indicates strong interfacial roughness, spin disorder, or exchange frustration at the RuO₂/CrO₂ interface.

The coercive field of the RuO₂/CrO₂ bilayer exhibits a markedly different temperature dependence from the exchange bias field. While H_{EB} decays rapidly and vanishes near the blocking temperature of RuO₂, H_c remains large (reaching approximately 650 Oe at 2 K) and decreases gradually with increasing temperature, ultimately approaching zero near the Curie temperature of CrO₂. This distinct behavior arises from their different physical origins.

H_{EB} reflects the interfacial unidirectional exchange anisotropy, which is sensitive to thermal fluctuations in the AFM, whereas H_c is governed primarily by magnetocrystalline anisotropy and domain wall pinning within the FM. Even after the interfacial EB is thermally randomized, residual uncompensated RuO₂ spins and interfacial roughness continue to impede magnetization reversal, leading to a finite coercivity up to T_C . The high low-temperature coercivity therefore reflects strong interfacial coupling and spin frustration at the RuO₂/CrO₂ interface, consistent with rigid exchange pinning inferred from the $m = 3$ dependence.

Thermal Evolution of Magnetization

The most notable behavior of the magnetic ordering at this interface can be captured by zero-field-cooled (ZFC) and field-cooled (FC) measurements. These magnetization measurements are used to reveal temperature-dependent magnetic behavior including magnetic hysteresis, irreversibility, divergence, superparamagnetic blocking, and spin-glass-like behavior.

ZFC measurements are collected by cooling the sample down from 400 K (carefully chosen to be above the ordering temperature(s) of the system: T_N and T_C), down to 3 K in zero field. The magnetization of the sample is then measured as it is heated under a constant external applied field $H_{ext} = 5000$ Oe. Conversely, FC measurements involve cooling the sample down from above the ordering temperature in the presence of the same applied external field. The characteristic features of these measurements and resulting inconsistencies yields very important information about how the magnetic ordering of RuO₂ is influenced by CrO₂ at the interface.

Typically, the measurement order is as follows: ZFC → FC. This ensures that we accurately capture the spin behavior while also minimizing the time required for repeated cycles.

Firstly, the FC and ZFC data in the high-temperature regime are indistinguishable. This overlap indicates that the system exhibits thermally-reversible magnetic behavior. It signifies that thermal energy is sufficient to overcome any anisotropy barriers or interfacial spin pinning such that the magnetization is independent of the sample's thermal or magnetic history. In this state, the RuO₂ layer behaves as a paramagnetic or weakly antiferromagnetic system, and

no exchange coupling to the CrO_2 layer is observed. The absence of bifurcation at high temperatures therefore suggests that the measurement was performed above the interfacial spin-freezing or blocking temperature, above which the RuO_2 spins remain thermally disordered and do not contribute to exchange bias.

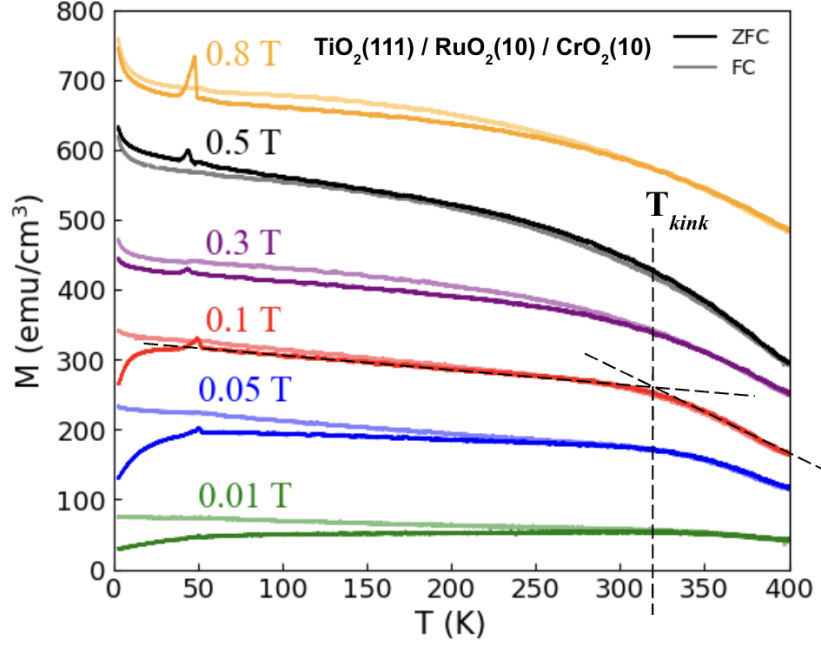


Figure 56: Temperature-dependent FC and ZFC measurements of same $\text{RuO}_2/\text{CrO}_2$ bilayer. Scans were collected at various H_{ext} . Lighter-tone lines represent FC and darker-tone lines represent ZFC data.

In addition to the overlap of the ZFC and FC curves at high temperatures, a distinct kink (T_{kink}) is observed in this regime, separating two linear regions with different temperature dependencies (see dashed lines). Closer analysis of the kink feature reveals a strong field dependence. The position of this kink shifts systematically with the applied external magnetic field strength. As the measurement field increases, the temperature at which the kink occurs decreases approximately linearly. This linear field-dependent behavior suggests that the kink corresponds to a magnetic transition or crossover that is sensitive to the applied field strength, such as the suppression of magnetic correlations or the gradual onset of short-range order within the RuO_2 layer. The linear decrease of the kink temperature with field indicates a Zeeman-like influence on the transition, consistent with a magnetic energy scale that competes with thermal fluctuations. This feature may therefore reflect a field-tunable magnetic crossover in the RuO_2 layer or at the $\text{RuO}_2/\text{CrO}_2$ interface, preceding the onset of exchange coupling at lower temperatures.

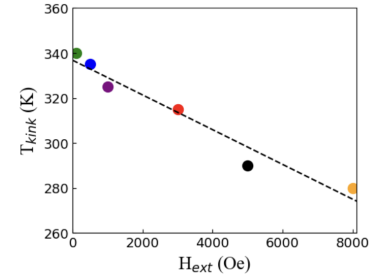


Figure 57: Kink temperature behavior as a function of applied external field. Dashed line to illustrate linear fit.

Magnetic Irreversibility

Upon cooling, a divergence between the ZFC and FC curves emerges as interfacial spins begin to freeze, marking the onset of exchange bias coupling in the bilayer. At low temperatures, a distinct bifurcation between the ZFC and FC magnetization curves is observed, indicating the onset of magnetic irreversibility associated with interfacial spin freezing or exchange coupling. For small applied magnetic fields, the ZFC magnetization decreases upon cooling while the FC curve remains relatively flat, suggesting that spins at the RuO₂/CrO₂ interface become partially frozen in random orientations in the absence of sufficient field alignment. This behavior is characteristic of systems with pinned or frustrated spins, where thermal relaxation is hindered below the blocking temperature.

Interestingly as the external applied field strength is increased beyond approximately 0.3 T, the ZFC magnetization instead increases at low temperatures, and the FC curve follows this upward trend. This upward trend indicates that the previously frustrated or pinned interfacial spins are now more free to align with the larger external field, thereby increasing the net magnetization. The crossover in the low-temperature ZFC data reflects the field-induced alignment of interfacial spins and the progressive suppression of the frozen spin component. The high-field regime therefore corresponds to a transition from a spin-frozen to a more uniformly-aligned interfacial state, consistent with the field dependence expected for an exchange-biased AFM/FM bilayer.

Anomalous Features

Interestingly, a sharp peak appears exclusively in the ZFC magnetization data at approximately 50 K for all applied field strengths, while the FC curves remain smooth through this temperature. The presence of this anomaly only in the ZFC curve indicates that it originates from a thermally-activated spin reconfiguration that depends on the sample's magnetic history. In the ZFC protocol, the system is cooled in zero field, leading to a frozen, randomly-oriented configuration of interfacial or surface spins. Upon warming in the presence of a small measurement field, these frustrated spins gradually gain enough thermal energy to overcome local anisotropy barriers and align with the applied field, resulting in a sudden increase in magnetization. Once this alignment occurs, further heating leads to thermal demagnetization, producing the characteristic peak shape in the ZFC curve. In contrast, during FC measurements the spins are already aligned while cooling under an external field, so this relaxation process does not occur and no corresponding peak is observed.

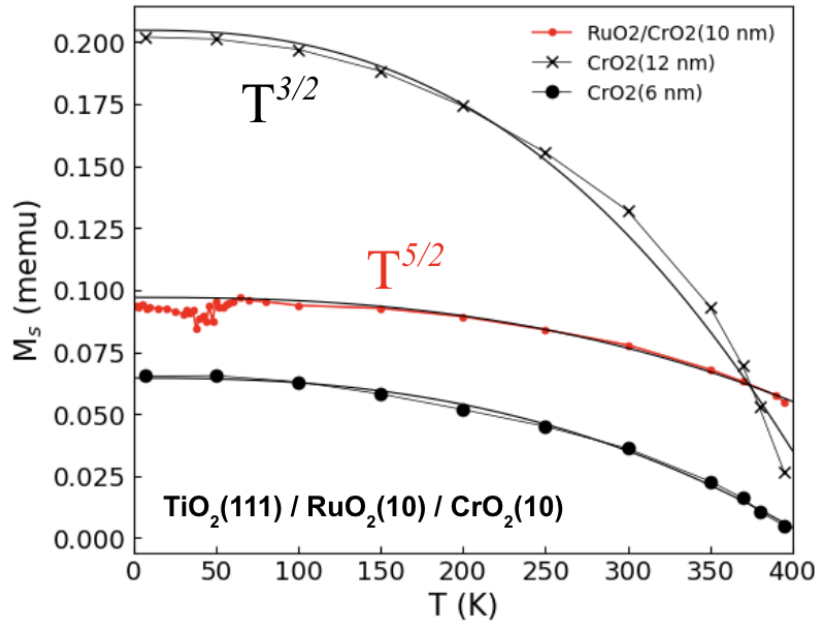


Figure 58: Temperature-dependent saturation magnetization measurement of RuO₂/CrO₂ bilayer. For comparison, single-layer CrO₂ films of comparable thicknesses also plotted, showing expected Bloch's law exponent.

Similarly to the magnetic characterization carried out on the CrO₂ films in the previous chapter, the temperature-dependent saturation magnetization (M_s) of the RuO₂/CrO₂ bilayer sample was collected and plotted relative to select CrO₂ films of varying thicknesses. This temperature dependence reveals a marked deviation from conventional Bloch's law behavior in the bilayer. While CrO₂ films exhibit a $T^{3/2}$ dependence consistent with thermally-excited spin waves in a 3D FM as shown in the previous chapter, the bilayer follows a $T^{5/2}$ trend. The steeper temperature dependence indicates a modification of the magnon spectrum due to interfacial exchange coupling and enhanced spin-orbit scattering introduced by the RuO₂ layer. Such effects suppress low-energy magnon excitations and introduce higher-order temperature terms in the magnetization decay.

The same interfacial coupling likely gives rise to the anomaly observed near 50 K, where partially frustrated RuO₂ spins reorient and transiently modify the effective magnetization of the bilayer. The temperature of this ZFC peak (≈ 50 K) corresponds closely to the location of the dip observed in the temperature dependence of the saturation magnetization M_s , suggesting a common origin. This feature likely marks a spin reorientation or spin-freezing transition within the RuO₂/CrO₂ bilayer, arising from competition between the exchange coupling at the interface and the intrinsic anisotropy of the CrO₂ layer. At this temperature, interfacial RuO₂ spins that

are weakly coupled to CrO₂ may undergo partial ordering or a spin-glass-like freezing, transiently enhancing the net magnetization before becoming thermally disordered at higher temperatures. The observation of this anomaly in both ZFC and $M_s(T)$ data supports the interpretation of a localized, field-sensitive interfacial transition that modifies the effective magnetization of the bilayer around 50 K.

Additional Physical Interpretations

Role of Spin-Orbit Coupling

RuO₂ possesses intrinsically strong SOC arising from its 4d electron character and partially filled t_{2g} orbitals. The SOC constant for Ru is an order of magnitude larger than that for typical 3d FMs [90], leading to significant spin-orbit-driven band splitting and Berry curvature effects.

Additionally, the rutile crystal field resulting from tetragonal symmetry leads to splitting of the Ru 4d states. SOC can strongly mix the spin and orbital degrees of freedom of these degenerate states near the Fermi level, leading to band splitting and anisotropic transport properties confirmed previously in both ARPES and ab initio studies [90, 91].

While direct studies of the RuO₂/CrO₂ interface have not yet been published, analogous oxide heterostructure work shows that broken inversion symmetry at heavy-element oxide interfaces can enable large Rashba-type SOC [92]. The rutile structure also lacks inversion symmetry along certain directions at the surface or interface. While bulk RuO₂ is centrosymmetric, interfacing it with another oxide like CrO₂ can break that symmetry, enabling Rashba-type SOC at the interface. So, even if bulk RuO₂ has moderate SOC, the interfacial SOC becomes enhanced due to the combination of heavy 4d element, broken inversion symmetry, and hybridization with 3d Cr orbitals.

Magnetic Proximity and the Induced Moment

Although RuO₂ is nominally antiferromagnetic or paramagnetic, proximity to ferromagnetic CrO₂ could induce a small ferromagnetic polarization in the RuO₂ layer near the interface. This may explain the field-dependent kink and exchange bias, both as signatures of a magnetically-polarized interfacial RuO₂ layer that gradually decouples with temperature. This idea connects to proximity magnetism which is currently a hot topic in the field of oxide spintronics.

Field-tunable Interfacial Exchange Regime

The linear decrease of the kink temperature with applied field could be interpreted as a Zeeman-driven suppression of interfacial spin correlations, implying that the interface behaves as a tunable magnetic subsystem. This could suggest that $\text{RuO}_2/\text{CrO}_2$ serves as a model system for field-controlled exchange bias, an idea worth exploring for various spintronic applications.

Magnetic Relaxation and Training Effects

Repeated hysteresis data after successive field cycles could help us better understand and comment on training effects. One such example is the gradual reduction of exchange bias with cycling to support the interfacial spin freezing interpretation. Future work could examine this aspect of the measurement routine.

Key Findings

The strong SOC of RuO_2 , when interfaced with the HMF CrO_2 , creates a platform where exchange interactions and relativistic effects coexist at the atomic scale. The combination of a heavy 4d AFM with a nearly 100% spin-polarized FM breaks inversion symmetry and gives rise to pronounced interfacial spin-orbit fields. These can modify the interfacial anisotropy and even induce weak spin canting or pinned uncompensated spins at the interface. These are all consistent with the exchange bias, spin freezing, and temperature-dependent irreversibility observed experimentally. The non-monotonic features in magnetization and the deviation from conventional Bloch-like behavior further suggest that SOC modifies spin dynamics and magnon spectra in these coupled system.

Looking Ahead

Beyond explaining these specific magnetic anomalies, the $\text{RuO}_2/\text{CrO}_2$ bilayer highlights a broader design principle for oxide spintronics: using 4d and 5d transition-metal oxides as SOC-active layers to tune interfacial exchange, magnetic anisotropy, and spin transport in ferromagnetic heterostructures. Because RuO_2 is metallic, chemically stable, and epitaxially compatible with CrO_2 , such bilayers could serve as a model system for exploring spin-orbit torques, topological spin textures, or spin Hall effects in fully oxide-based devices. This work thus establishes $\text{RuO}_2/\text{CrO}_2$ as a versatile and previously unexplored material platform for controlling interfacial magnetism through spin-orbit coupling.

Chapter 5 | Extrinsic Charge–Spin Conversion in Ru–Pt Alloys

IN THE broader context of spintronic devices, the ability to convert charge currents into spin currents efficiently is as critical as generating and transmitting spin polarization. Materials exhibiting a large SHE play a central role in this process, enabling SOT mechanisms that can manipulate magnetization without the need for charge currents to flow through a FM, as in conventional STT devices. By providing a robust and controllable source of spin-polarized currents, SHE materials form the final stage of the spin current “pipeline,” complementing the intrinsic spin generation in HMFs and the spin transmission across AFM/FM interfaces studied in the preceding chapters.

Among candidate SHE materials, Ru–Pt alloys are particularly attractive. Their strong SOC, structural stability across a wide compositional range, low electrical resistivity, and high crystalline order promote them ideal platforms for exploring the factors that control spin-charge conversion efficiency. In this chapter, the detection of the SHA in Ru–Pt alloys is carried out using three complementary experimental techniques: magneto-optic Kerr effect (MOKE) microscopy, spin-torque ferromagnetic resonance (ST-FMR), and time-domain terahertz (THz) emission spectroscopy (TDTS). The results discussed in this chapter have been submitted for publication [V. S. Bheemarasetty et al. "Structure-Driven Trends of the Spin Hall Effect in $\text{Ru}_x\text{Pt}_{1-x}$ Thin Films" *manuscript submitted 2025*]. This multimodal approach provides both static and dynamic probes of the spin currents generated in Ru–Pt, establishing a foundation for understanding how composition, structure, and SOC jointly influence spin-charge interconversion.

By situating Ru–Pt in the context of the broader spintronic workflow from generation (CrO_2) to transmission ($\text{RuO}_2/\text{CrO}_2$ interfaces) to conversion (Ru–Pt alloys), this study emphasizes the material’s role as a tunable, high-performance platform for next-generation SOT devices.

MOTIVATION

The search for materials with high charge-to-spin conversion efficiency remains one of the key challenges in spintronics. This efficiency represents the ability of a material to transform an applied charge current into a spin current, a process that underpins the operation of SOT devices, SHE-based architectures, and numerous other spintronic applications. Materials exhibiting a large conversion efficiency are particularly desirable because they enable reduced power consumption, faster switching dynamics, and improved de-

vice functionality, all of which are critical for the advancement of next-generation memory and logic technologies.

Several heavy metals including β -W [93] and Ta [94] have been extensively investigated due to their relatively large SHAs, which can reach values exceeding 0.33. These materials, however, are hindered by very high resistivities. For instance, β -W has a resistivity of approximately $263 \mu\Omega \text{ cm}$ which is considerably greater than those of typical conductive materials. Elevated resistivity introduces significant Joule heating during current flow, leading to energy dissipation and compromised thermal stability. These can be particularly problematic for nanoscale spintronic devices. In addition, high resistivity restricts the maximum current density that can be driven through the material and reduces the efficiency of spin-current injection into adjacent magnetic layers. This poses obstacles to scalability and long-term device reliability.

The strong correlation between resistivity and SHA in these systems often arises from intrinsic mechanisms of the SHE. In such cases, the SHA typically increases with resistivity, as materials with stronger Berry curvature in their electronic band structures naturally exhibit larger conversion efficiencies [95].

Beyond intrinsic mechanisms, the SHE can also arise from extrinsic processes, which are driven by spin-dependent scattering from impurities or structural defects. Two dominant scattering mechanisms have been identified: skew scattering [96] and side-jump scattering [97]. Skew scattering has been experimentally reported in several alloy systems including CuIr, CuBi, and CuPb, with measured SHAs of approximately 0.02, 0.24, and 0.13, respectively [95, 98, 99]. These examples point to alloy engineering as a promising strategy for enhancing SHA by introducing controlled impurity concentrations, thereby leveraging extrinsic contributions to the SHE. This helps to optimize the balance between spin-charge conversion efficiency and conductivity.

The resistivity of a sample can be engineered by carefully selecting alloy constituents. AuTa alloys, for example, can achieve a SHA of 0.5 while maintaining a moderate resistivity of around $85 \mu\Omega \text{ cm}$, making them attractive for device applications. Inspired by these results, a similar design strategy is adopted here: selecting a host metal with desirable properties and incorporating heavy-element impurities to enhance the SHA. Since the strength of the SOC scales with the atomic number as Z^4 , heavier elements are particularly favorable for maximizing the SHE.

The design of effective spin Hall materials requires consideration of SOC strength as well as structural stability, phase formation, and ease of fabrication. Platinum (Pt), a 5d transition metal, is well known for its strong SOC and large intrinsic SHA, making it an excellent candidate for engineering high-performance spintronic materials. Ruthenium (Ru), a 4d transition metal, is widely used in spintronic devices due to its role in interlayer exchange coupling, and offers structural compatibility with Pt. Specifically, Pt crystallizes in a face-centered cubic (FCC) lattice, which is highly compatible with the hexagonal close-packed (HCP) structure of Ru, enabling stable alloy formation. When combined, the Ru-Pt binary system offers a promising platform to investigate tunable electronic properties and the potential for enhanced performance through extrinsic scattering mechanisms. Pt provides strong SOC and an FCC crystal structure, while Ru contributes structural compatibility, reduced resistivity, and stability across a wide composition range. This allows for direct investigation of how crystal structure and impurity concentration impact spin-charge conversion.

In this chapter, the Ru-Pt binary alloy is systematically explored as a model platform for probing the interplay between intrinsic and extrinsic spin-charge conversion mechanisms. Its tunable composition and structural phase evolution offer control over both electronic coherence and scattering environments, while maintaining excellent chemical stability and compatibility with FM interfaces.

Method of Co-sputtering

Ru-Pt alloy thin films are deposited on silicon substrates using high-vacuum magnetron sputtering. The sputtering was performed at a dc power of 30 W with a background argon pressure of 4 mTorr and a base chamber pressure of 3×10^{-8} Torr. A series of $\text{Ru}_x\text{Pt}_{1-x}$ alloys from $x = 0$ to 100 were fabricated via the co-sputtering technique from elemental Ru and Pt targets, allowing precise control of film composition. Co-sputtering here refers to the geometric configuration of a complete, primary target (i.e. Ru) with pieces of a secondary target (i.e. Pt) placed on top. This arrangement effectively forms a binary target whose composition can be carefully tuned by varying the geometry of both materials. The nominal stoichiometry of bulk samples was estimated from the relative geometry of the binary target using area calculations. The compositions of each element were further verified to high precision using energy dispersive X-ray spectroscopy (EDS) in an SEM. The bulk film thicknesses were determined using X-ray reflectivity (XRR), which revealed a monotonic decrease in deposition rate with increasing Ru content from 1.214

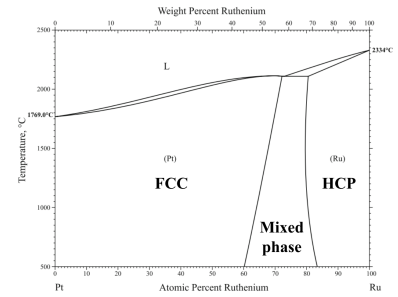


Figure 59: Binary phase diagram of the Ru-Pt system. Solid phases labeled with stable-state crystal structures [100].

$\text{\AA}/\text{s}$ at $x = 38$ to $0.689 \text{ \AA}/\text{s}$ at $x = 100$. This is consistent with the relative sputtering rate yields of these elements: 16 Pt atoms are ejected for 10 Ar ions, while only 13 Ru are ejected for the same ion count. As such, a higher Pt concentration in the alloy is expected to yield a higher sputtering rate

Nominal (Ru:Pt)	Ru (x)	Pt (1-x)	Rate ($\text{\AA}/\text{s}$)
0:100	0	100	1.03
25:75	38.0 ± 1.0	62.0 ± 1.0	1.21
45:55	49.5 ± 1.3	50.5 ± 1.3	1.19
55:45	58.7 ± 0.9	41.3 ± 0.9	1.04
70:30	64.7 ± 0.7	35.3 ± 0.7	1.03
80:20	78.4 ± 0.4	21.6 ± 0.4	0.92
90:10	86.5 ± 0.5	13.5 ± 0.5	0.92
100:0	100	0	0.69

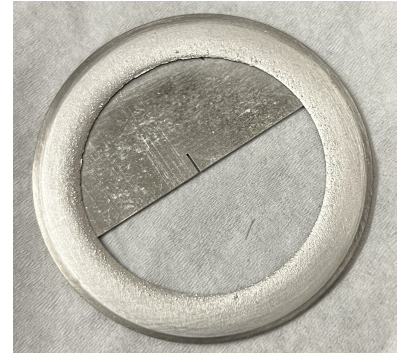


Figure 60: Sample co-sputtering target geometry for Ru-Pt alloy. This shows Pt target geometry for the 45:55 nominal stoichiometry.

Crystal Structure Analysis

The as-grown crystalline structure of the $\text{Ru}_x\text{Pt}_{1-x}$ films was analyzed using XRD. The pure Pt film ($x = 0$) exhibits an FCC structure dominated by the (111) diffraction peak, which was indexed using the ICDD Powder Diffraction File. Conversely, the pure Ru film ($x = 100$) displayed an HCP structure with a prominent (002) peak. As the stoichiometry is systematically tuned across the alloy series, a continuous structural transition from FCC to HCP is observed. This is consistent with increasing the concentration and is evident by the primary diffraction peak shifting from the FCC (111) position towards the HCP (002) 2θ position with increasing Ru content.

This trend is also confirmed using the Visualization for Electronic and Structural Analysis (VESTA) simulation software package. This is a 3D visualization program that can simulate diffraction patterns from a given structural model or crystal morphology. Beginning with the FCC Pt unit cell, Ru impurity atoms were gradually introduced, first as interstitial impurity atoms and then as substitutional atoms in the lattice. By incrementally changing the lattice constant of the Pt FCC unit cell, the experimental XRD results are reproduced to very good precision.

The gradual peak shift reflects a progressive reduction in the lattice constant as Pt atoms are increasingly substituted by smaller Ru atoms. Importantly, the presence of sharp and well-defined diffraction peaks confirms that the films are polycrystalline with clear grain

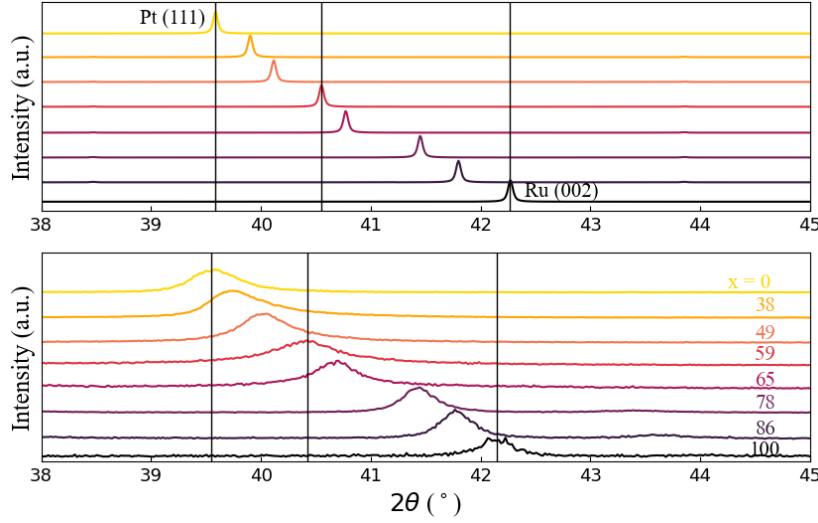


Figure 61: VESTA simulation results (top) confirm experimental XRD scans of Ru-Pt alloys shifting from Pt FCC (111) peak to Ru HCP (002) peak (bottom). Vertical lines are guide to the eye.

structure, even though they were deposited by magnetron sputtering without any post-deposition annealing. This poses as a major advantage to the Ru-Pt alloy system, since no post-processing procedures are required to attain robust polycrystalline structure.

The lattice constants of the $\text{Ru}_x\text{Pt}_{1-x}$ alloys were determined using the 2θ positions of these diffraction peaks and Bragg's law:

$$n\lambda = 2d_{hkl} \sin \theta, \quad (49)$$

where n is the diffraction order taken to be 1, λ is the characteristic Cu-K α X-ray wavelength, and d_{hkl} is the interplanar spacing of the (hkl) plane.

For the FCC structure, the cubic lattice constant a_{FCC} is determined using

$$a_{\text{FCC}} = d_{hkl} \sqrt{h^2 + k^2 + l^2}. \quad (50)$$

For the HCP structure, the unique lattice constants a_{HCP} and c_{HCP} were calculated using the relation:

$$\frac{1}{d_{hkl}^2} = \frac{4(h^2 + hk + k^2)}{3a_{\text{HCP}}^2} + \frac{l^2}{c_{\text{HCP}}^2}. \quad (51)$$

Conveniently, the interplanar spacing of the FCC (111) planes is equivalent to that between the HCP (002) planes, allowing a direct comparison of this parameter. As calculated, the measured d-spacing (d_{hkl}) decreases smoothly as the alloy composition transitions from FCC-dominated Pt-rich films to HCP-dominated Ru-rich films. This continuous trend is consistent with the Ru-Pt binary phase diagram

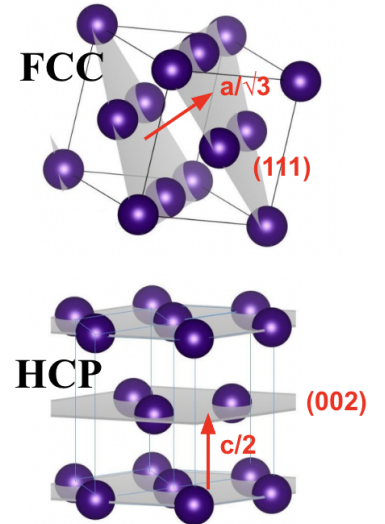


Figure 62: Comparison of interplanar spacing in FCC and HCP planes with corresponding lattice constants denoted.

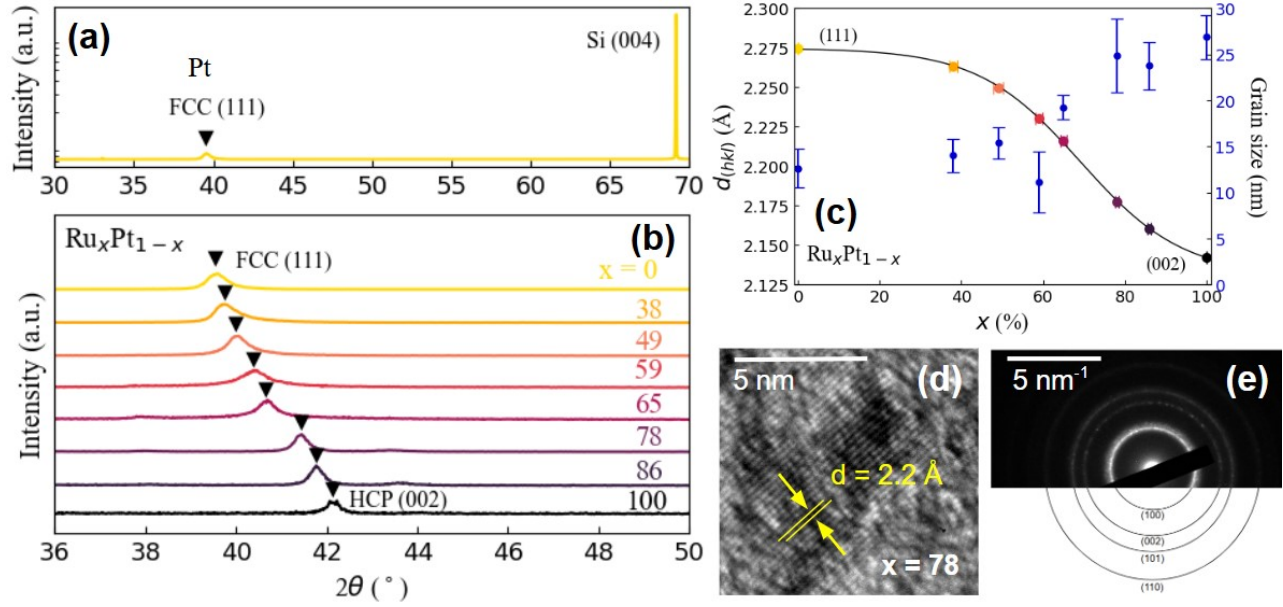


Figure 63: Characterization of as-grown Ru-Pt alloys. (a) $\theta - 2\theta$ scan for Pt sample indicating FCC (111) peak and Si substrate. (b) $\theta - 2\theta$ scans of alloys with increasing Ru content. (c) Extracted lattice spacing d_{hkl} and corresponding grain size (blue data) from full-width at half-maximum of XRD peaks in (b). (d) STEM micrograph of $x = 78$ sputtered film. (e) Diffraction pattern of sample in (d) with HCP planes identified.

and reflects the progressive lattice compression caused by substituting smaller Ru atoms into the Pt lattice. The systematic constriction of the lattice constant confirms that alloying drives a well-behaved structural evolution across the entire composition range.

TEM analysis of a sputtered $x = 78$ alloy is also presented, which independently confirms the lattice spacing values obtained from the previous calculations. The corresponding selected-area electron diffraction (SAED) pattern exhibits high-intensity diffraction rings pointing to a polycrystalline sample. Careful image analysis indicates these rings are indeed aligned along the specific crystallographic planes that are characteristic of the HCP-dominant crystal structure of the $x = 78$ composition.

To evaluate the average grain size of the $\text{Ru}_x\text{Pt}_{1-x}$ polycrystalline films, the Scherrer equation is used:

$$g_s = \frac{k\lambda}{\beta_{FWHM} \cos \theta}, \quad (52)$$

where g_s is the average grain size, k is the shape factor (taken as 0.89 for approximately spherical grains), λ is the characteristic X-ray wavelength, θ is the Bragg angle, and β_{FWHM} is the full-width at half-maximum of the dominant diffraction peak, expressed in radians. The average grain sizes fall within the range of approximately 11 nm to 27 nm, indicating that the films are composed of nanocrystalline grains. The grains tend to be smaller for Pt-rich films, approaching 10 nm in size, and grow to almost three times that size for Ru-rich films.

This behavior can be understood in terms of nucleation and growth dynamics during film deposition, adatom mobility, and alloying effects.

Grain growth in thin films is fundamentally driven by the minimization of total energy, including surface and grain boundary contributions. Ru, with stronger metallic bonding compared to Pt, tends to form fewer nucleation sites during deposition. The lower nucleation density allows individual grains to grow larger before impinging on neighboring grains, resulting in a coarser microstructure. In contrast, platinum-rich films favor a higher nucleation rate due to lower surface energy and more uniform wetting of the substrate, producing a larger number of smaller grains.

Adatom mobility further contributes to this trend. Ru adatoms possess higher diffusion lengths under typical deposition conditions, facilitating growth of existing grains rather than the formation of new nuclei. Conversely, Pt adatoms, which nucleate more readily and experience solute drag at grain boundaries, are restricted in lateral motion, limiting grain coarsening. In alloyed films, increasing Pt content can further impede Ru grain growth due to lattice strain and solute effects, reinforcing the formation of finer grains.

Electronic Transport Properties

As before, the $\text{Ru}_x\text{Pt}_{1-x}$ films are patterned into $20 \times 55 \mu\text{m}^2$ Hall-bar devices using photolithography followed by physical ion milling. The longitudinal resistivity (ρ_{xx}) of the patterned films was measured as a function of temperature using a PPMS.

Temperature-dependent ρ_{xx} indicates clear metallic transport behavior, characterized by a gradual increase in resistivity with temperature over the range from 2 K to 300 K. The RRR values, defined here as $\rho_{xx}(300 \text{ K})/\rho_{xx}(2 \text{ K})$, are found to be relatively low (≤ 1.3) for all alloys. This indicates the presence of strong impurity and alloy scattering in this compositional range. In contrast, typical high-purity metals have very large RRR values reaching 1000 due to minimal scattering.

Despite the enhanced scattering, the measured resistivity for all alloys in this series remains below $70 \mu\Omega \text{ cm}$. This is significantly lower than other high-resistivity alloy systems such as AuTa and TaW which are commonly noted for their large SHAs, promoting $\text{Ru}_x\text{Pt}_{1-x}$ alloys for spintronic device applications with minimized Joule heating effects and reduced energy dissipation. When plotted as a function of composition, the ρ_{xx} data at 2 K exhibits a clear

parabolic dependence, characteristic of alloy scattering. This smooth, systematic behavior confirms that the observed resistivity trends are consistent with what is expected and that no anomalous scattering mechanisms dominate within this composition range.

An important connection can now be identified between the evolution of the lattice structure, the resistivity, and the spin Hall performance: FCC alloys, marked by low Ru content ($x < 50\%$), have less scattering due to higher symmetry. This results in lower ρ_{xx} .

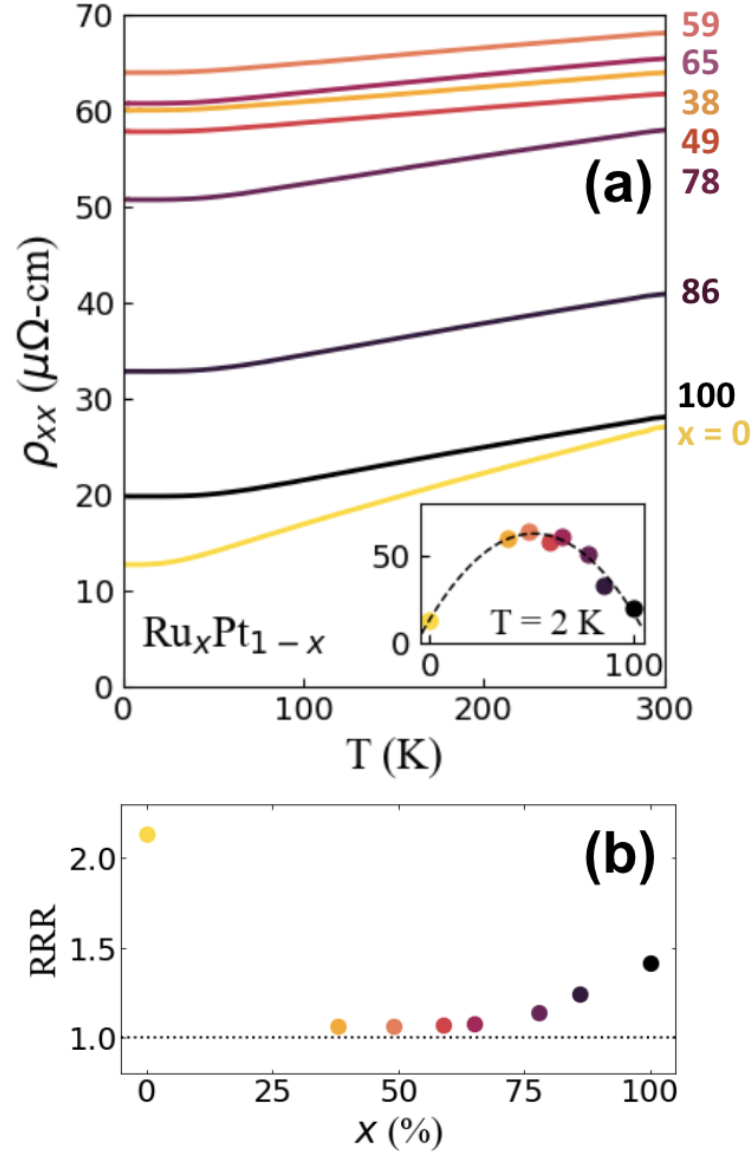


Figure 64: (a) Longitudinal resistivity as a function of temperature for various alloy compositions. Inset: Resistivities at 2 K plotted as a function of Ru composition ($x\%$). (b) Evolution of residual resistivity ratio (RRR) with $x\%$.

The combination of FCC structure and a low Ru impurity content favors skew scattering-dominated contributions to the SHE, enabling larger SHAs without compromising on the conductivity of the alloy.

As Ru content increases leading to HCP-dominant alloys, the lattice becomes increasingly more compressed with higher impurities, introducing additional disorder and scattering centers. This increases ρ_{xx} . HCP alloys with higher disorder and smaller lattice constants are more susceptible to side-jump and intrinsic mechanisms, which typically require higher resistivities to achieve comparable SHA values. This connection between crystal structure and electronic properties points to the significance of crystallography and alloy design in optimizing charge-to-spin conversion efficiency for spintronic applications.

Spin Hall Angle Characterization

Bilayer structures of $\text{Ru}_x\text{Pt}_{1-x}(5\text{nm})/\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}(10\text{nm})$ were fabricated to quantify the charge-conversion efficiency of the non-magnetic HM Ru-Pt layers. Here, $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}$ is the FM layer which serves as a spin detector. When a charge current flows through the HM, the SHE generates a transverse spin current. This spin current diffuses into the adjacent FM, where it can exert a torque on the magnetization or produce a measurable voltage. Without the FM layer, there is no mechanism to convert the spin accumulation into an observable signal, making it impossible to quantify the SHA.

Three independent techniques were utilized to directly probe the SHE in these $\text{Ru}_x\text{Pt}_{1-x}$ alloys: spin-torque magneto-optic Kerr effect (ST-MOKE) microscopy, spin-torque ferromagnetic resonance (ST-FMR), and time-domain terahertz (THz) emission spectroscopy (TDTS). The results of these measurements provide insightful connections between the observed spin-charge conversion efficiency, alloy composition, crystal structure, and scattering mechanisms.

Spin-Torque MOKE

The SHA of the $\text{Ru}_x\text{Pt}_{1-x}$ alloys was determined firstly using the spin-transfer MOKE (ST-MOKE) microscopy technique. This technique is used to probe magnetization dynamics in ferromagnetic materials. In this method, a spin-polarized current or spin current exerts a torque on the magnetization of a ferromagnet, inducing precession or switching. The resulting changes in magnetization are detected optically via the Kerr effect, where the polarization of reflected light rotates proportionally to the magnetization component along the measurement axis, enabling time-resolved observation of spin-transfer-induced dynamics. The setup is similar to the polar-MOKE setup discussed previously, with a few modifications to allow for rapid detection of spin dynamics.

A pulsed laser source with a wavelength of 780 nm was used with a focused spot size of approximately $19.6 \mu\text{m}^2$ [101]. During the measurement, a sinusoidal AC current with a peak amplitude of 20 mA at 1.333 kHz was applied along the x -axis of the sample. MOKE voltages are measured as the laser position is held fixed and the sample is moved along the y -direction. The MOKE signal was then used to probe the effective fields associated with the damping-like and the field-like torques.

To extract the SHA, the self-calibration method was employed [102, 103]. In this approach, the measured MOKE signal contains contributions from both the SOT and the Oersted fields, allowing us to simultaneously quantify their respective effects. The resulting signal can be expressed as:

$$\Delta V_{\text{Oersted}}(x, y) = \zeta \frac{1}{2\pi r^2} \oint \int dy' dx' \epsilon \frac{h_{\text{Oersted}}(y')}{H_{\text{eff}} + M_{\text{eff}}} \quad (53)$$

and

$$\Delta V_{\text{SOT}}(x, y) = \zeta \frac{1}{2\pi r^2} \oint \int dy' dx' \epsilon \frac{h_{\text{SOT}}(y')}{H_{\text{ext}} + M_{\text{eff}}}, \quad (54)$$

where r is the radius of the laser spot,

$$\epsilon = e^{-\frac{((x-x')^2 + (y-y')^2)}{r^2}} \quad (55)$$

and ζ describes the sensitivity of the MOKE response defined as $\zeta = \frac{V_{\text{MOKE}}}{\Delta m_z}$. The finite size of the laser spot necessitates that the Kerr rotation be calculated as an integral of the local magnetization reorientation, weighted by the Gaussian function of the spatial distribution of the laser. The magnitude of the SOT-induced reorientation is then calculated as [104]:

$$\frac{h_{\text{SOT}}}{I_c/2\pi w} = \frac{\Delta V_{\text{SOT}}(x, y)/\epsilon}{\Delta V_{\text{offset}}(x, z)/\oint \int dy' dx' \ln(\frac{w-y'}{y'})\epsilon}. \quad (56)$$

with $w = 50 \mu\text{m}$ being the width of the device strip. The SHA of the $\text{Ru}_x\text{Pt}_{1-x}$ alloy using MOKE, Θ_{MOKE} , can then be calculated by:

$$\Theta_{\text{MOKE}} = \frac{2e}{\hbar} \frac{h_{\text{SOT}}}{j_{\text{Ru-Pt}}} \mu_0 M_s d_{\text{CFB}}, \quad (57)$$

where e is the elementary charge, $\hbar$ is the reduced Planck's constant, $j_{\text{Ru-Pt}}$ is the current density in the $\text{Ru}_x\text{Pt}_{1-x}$ layer, μ_0 is the vacuum permeability, M_s and d_{CFB} are the saturation magnetization and thickness of the CoFeB layer, respectively. CoFeB has a resistivity of about $100 \mu\Omega \text{ cm}$. By considering the thickness of $\text{Ru}_x\text{Pt}_{1-x}$ and CoFeB through the parallel resistor model, the current density in the $\text{Ru}_x\text{Pt}_{1-x}$ layer at different compositions can be estimated reasonably accurately.

ST-MOKE measurements were performed by our collaborators Xinhao Wang and John Q. Xiao at the University of Delaware.

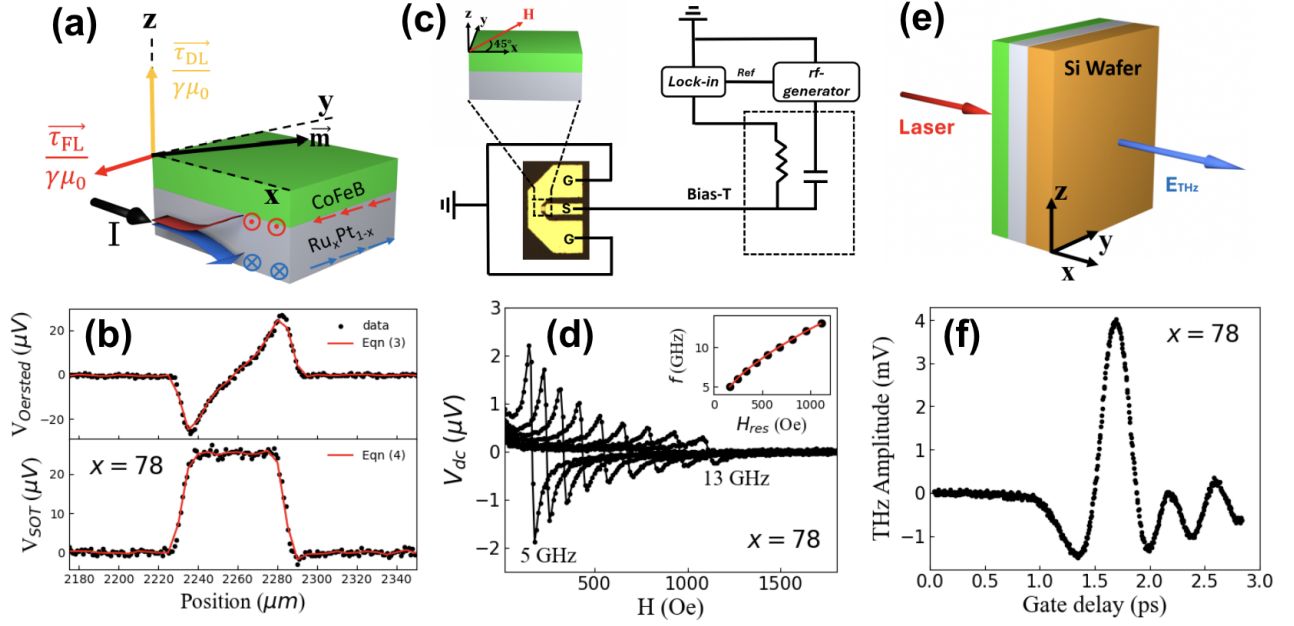


Figure 65: (a) Schematic of SOT in CoFeB from spin Hall effect in Ru-Pt used for MOKE measurement. (b) Determination of the magnitude of SOT using self-calibration. Magnitude of the SOT is determined by fitting to the out-of-plane Oersted field (top) and SOT (bottom). (c) Schematic of the ST-FMR measurement setup and device structure with rf excitation and lock-in detection. (d) Frequency-dependent ST-FMR spectra showing resonance shift with external magnetic field. Inset: Kittel fit. (e) Schematic of THz spectroscopy setup. (f) THz emission of $x=78$ alloy.

Spin-Torque FMR

To independently confirm the SHA obtained from the ST-MOKE measurements, the spin-torque ferromagnetic resonance (ST-FMR) technique is used. This technique measure SOTs and magnetization dynamics in ferromagnetic/heavy-metal heterostructures. An rf current applied to the device generates a spin current via the SHE in the heavy metal, which exerts a torque on the ferromagnet and drives magnetization precession. The resulting oscillations of the magnetization produce a rectified voltage across the device due to the anisotropic magnetoresistance, allowing quantitative measurement of the spin-torque efficiency and magnetic properties.

The Ru_xPt_{1-x}(5nm)/Co₄₀Fe₄₀B₂₀(10nm) bilayers are now patterned into coplanar waveguide (CPW) devices with the desired G-S-G probe configuration detailed previously. An rf current (I_{rf}) with an input power of +10 dBm was applied along the x -axis of the patterned device. This rf current induces SOTs in the CoFeB layer due to the spin current generated within the Ru_xPt_{1-x} layer. During the measurement, the external magnetic field was fixed at an in-plane angle of 45° to maximize the mixing signal. The resulting rectified voltage V_{mix} arises from the dynamic interaction between the rf current, magnetization precession, and SOTs present. The measured voltage spectrum is fitted using a combination of symmetric Lorentzian and

antisymmetric dispersive components [105] and expressed as:

$$V_{dc} = c \left[V_{sym} \frac{\Delta H^2}{\Delta H^2 + (H_{ext} - H_{res})^2} + V_{asym} \frac{\Delta H(H_{ext} - H_{res})}{\Delta H^2 + (H_{ext} - H_{res})^2} \right]. \quad (58)$$

Here, c is an arbitrary constant used as a scaling factor, ΔH is the half linewidth of the Lorentzian, H_{res} is the resonance field, and H_{ext} is the applied external magnetic field, respectively.

By this notation, V_{sym} and V_{asym} are proportional to the in-plane torque $\tau_{||}$ and the out-of-plane torque $\tau_{\perp}$, respectively. Analogously, these correspond to the out-of-plane effective field and the in-plane effective field, respectively. In this case it is reasonable to assume that $\tau_{\perp}$ is dominated by the torque originating from the Oersted field [94, 105]. The SHA of the Ru-Pt HM can thus be expressed as:

$$\begin{aligned} \Theta_{FMR} &= \frac{\tau_{||}}{\tau_{\perp}} \frac{e\mu_0 M_s}{\hbar} d_{CFB} d_{Ru-Pt} \\ &= \frac{V_{sym}}{V_{asym}} \frac{e\mu_0 M_s}{\hbar} d_{CFB} d_{Ru-Pt} \sqrt{1 + \frac{M_{eff}}{H_R}}. \end{aligned} \quad (59)$$

Sweeping the rf signal frequency enables the extraction of M_{eff} by fitting the resonant frequency f_{res} to the resonant field H_{res} using the Kittel formula [106]:

$$f_{res} = \frac{\gamma}{2\pi} \sqrt{H_{res}(H_{res} + \mu_0 M_{eff})}. \quad (60)$$

THz Emission Spectroscopy

As a third independent verification of the SHE in Ru_xPt_{1-x} , time-domain terahertz emission spectroscopy (TETS) was utilized. THz spectroscopy uses short pulses of electromagnetic radiation in the THz frequency range to probe the dynamical properties of materials. A THz pulse is typically generated and directed onto the sample, where it interacts with charge carriers, spins, or lattice vibrations. By measuring the transmitted or reflected THz pulse in the time domain and performing a Fourier transform, one can extract frequency-dependent material properties such as conductivity, dielectric response, or spin dynamics.

In this experiment, 150 mW femtosecond laser pulses ~ 40 fs in duration with a 800 nm center wavelength, 10 kHz repetition rate, a fluence of 2.23 mJ cm^{-2} , and a spot size of roughly 1 mm were directed onto the sample. In the context of THz spectroscopy, the fluence of a

pulse refers to the energy per unit area delivered to the sample. It is an important parameter because the sample's response depends on fluence, with higher values potentially inducing nonlinear effects or heating. Electro-optic sampling was used to detect the emitted THz wave with a 1 mm-thick (110) ZnTe crystal. The single-crystal ZnTe is used as a nonlinear optical crystal to both generate THz pulses via optical rectification and detect them through electro-optic sampling. This enables time-resolved measurement of the THz electric field. A static magnetic field of 1.4 kOe, generated by a pair of permanent magnets, maintained the magnetization direction of the FM layer during measurements. The total electric field of the THz radiation is then extracted in order to calculate the SHA.

When the laser excites the CoFeB layer, photo-excited spin-up and spin-down electrons are driven above the Fermi level and diffuse into the $\text{Ru}_x\text{Pt}_{1-x}$ underlayer, creating a spin current. Within the HM layer, this spin current is converted into a transverse charge current through the ISHE [107], which emits a broadband THz pulse (also referred to as THz wave radiation). The magnetization direction of CoFeB determined the polarization of the injected spin current. As such, the sign of the THz electric field reverses when the magnetization of the CoFeB is flipped by the external field, confirming the spin-dependent origin of the signal. A finite THz response is detected across the full Ru-Pt composition range. The emitted amplitude reflects not only the SHA but also contributions from the HM resistivity and the optical absorption efficiency of the FM layer, consistent with prior studies [108–110].

To quantify the relative SHA from THz spectroscopy, Θ_{THz} , of each alloy, the analysis in [111] is implemented. The THz signals of each alloy cannot be determined absolutely but rather as a proportion relative to that of pure Pt. This is due to various technical limitations of the setup used. In order to calculate the absolute SHA of Pt, the detector response function and photo-to-spin current conversion efficiency, $B(\omega)$, must be known, as shown below:

$$S_{\text{THz}}(\omega) = AB(\omega)\lambda_r \tanh\left(\frac{t_{\text{HM}}}{\lambda_r}\right)\theta_{\text{SH}}Z(\omega). \quad (61)$$

In this equation, $S_{\text{THz}}(\omega)$ is the emitted THz signal amplitude measured in volts. A is the pump-fluence absorbance which represents the fraction of the pump pulse energy absorbed by the sample. In optically-pumped THz-probe experiments, only the absorbed portion of the pump can excite carriers, spins, or lattice vibrations, which then contribute to THz emission. Higher absorption generally leads to stronger THz signals. $B(\omega)$ is the phonon spin-conversion efficiency which quantifies how efficiently energy from the pump-

THz emission spectroscopy measurements were performed by our collaborator Shreya Shrestha under the supervision of advisors Matthias Benjamin Jungfleisch and Lars Gundlach at the University of Delaware.

induced lattice vibrations (i.e. phonons) is converted into a spin current in the material. In some spintronic THz experiments, lattice excitations can couple to the spin system so $B(\omega)$ measures the fraction of this energy that contributes to spin dynamics.

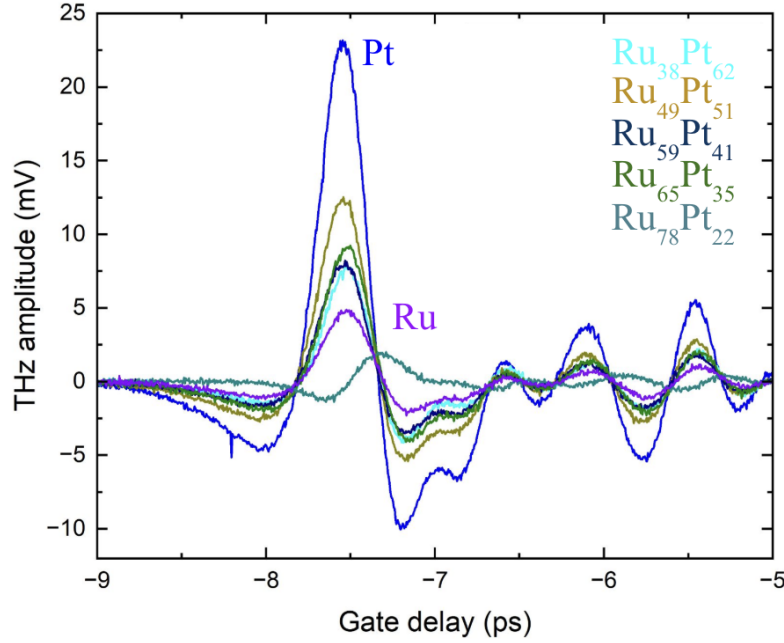


Figure 66: Summary of THz emission signals of Ru-Pt alloys relative to Pt signal. Compositions labeled.

λ_r is the spin-current relaxation length representing the characteristic distance over which a spin current decays in a material due to spin scattering or relaxation. In a heavy-metal/ferromagnet heterostructure, it determines how far the spin current can propagate before losing polarization, which directly affects the amplitude of the THz emission. Finally, $Z(\omega)$ is the effective impedance of the spintronic heterostructure to the emitted THz radiation. It determines how efficiently the spin-to-charge conversion (or spin-current-induced THz emission) couples to free space. A mismatch in impedance can reduce the observed THz signal.

Given the limitations of the setup, these parameters cannot be calculated in a straightforward manner for these samples. As an alternative approach, we extract the SHA from amplitude of the Ru-Pt alloys THz signals relative to that of pure Pt. We do so by assuming that the detector response function will be the same for all the samples, as all the samples are measured in the same system under identical conditions. We also assume and that the spin-to-phonon conversion efficiency is the same for every sample:

Note: The spin-current relaxation length λ_r describes the distance over which an ultrafast, pump-induced spin current decays before it relaxes or is converted into a measurable signal. This is different from the spin diffusion length λ_{sf} , which characterizes the steady-state decay of spin polarization in a material.

$$\frac{\Theta_{\text{THz}}}{\theta_{\text{Pt}}} = \frac{S_{\text{THz}}(\omega)}{AB(\omega)\lambda_r \tanh(\frac{d_{\text{Ru-Pt}}}{2\lambda_s})Z(\omega)}, \quad (62)$$

Here, θ_{Pt} is the reference SHA of pure Pt. From this, the SHA calculation can be simplified from

$$\begin{aligned} \frac{\Theta_{\text{THz}}^{\text{Ru-Pt}}}{\theta_{\text{THz}}^{\text{Pt}}} &= \frac{S_{\text{THz}}^{\text{Ru-Pt}}(\omega)}{S_{\text{THz}}^{\text{Pt}}(\omega)} \times \frac{A^{\text{Pt}}}{A^{\text{Ru-Pt}}} \times \frac{B^{\text{Pt}}(\omega)}{B^{\text{Ru-Pt}}(\omega)} \\ &\times \frac{\lambda_r^{\text{Pt}}}{\lambda_r^{\text{Ru-Pt}}} \times \frac{\tanh(\frac{t_{\text{Pt}}}{\lambda_r^{\text{Pt}}})}{\tanh(\frac{t_{\text{Ru-Pt}}}{\lambda_r^{\text{Ru-Pt}}})} \times \frac{Z^{\text{Pt}}(\omega)}{Z^{\text{Ru-Pt}}(\omega)} \end{aligned} \quad (63)$$

to

$$\frac{\Theta_{\text{THz}}^{\text{Ru-Pt}}}{\theta_{\text{THz}}^{\text{Pt}}} = \frac{S_{\text{THz}}^{\text{Ru-Pt}}(\omega)}{S_{\text{THz}}^{\text{Pt}}(\omega)} \times \frac{A^{\text{Pt}}}{A^{\text{Ru-Pt}}} \times \frac{\lambda_r^{\text{Pt}}}{\lambda_r^{\text{Ru-Pt}}} \times \frac{\tanh(\frac{t_{\text{Pt}}}{\lambda_r^{\text{Pt}}})}{\tanh(\frac{t_{\text{Ru-Pt}}}{\lambda_r^{\text{Ru-Pt}}})} \times \frac{Z^{\text{Pt}}(\omega)}{Z^{\text{Ru-Pt}}(\omega)}. \quad (64)$$

Following the approximation made by [111],

$$\frac{\lambda_{\text{MF}}}{\sigma_{xx}} = 0.3 \times 10^{-15} \Omega m^2, \quad (65)$$

with λ_{MF} denoting the electron mean free path which λ_r approaches as $\omega \rightarrow \infty$, and σ_{xx} is the electrical conductivity of the HM. Using the room-temperature ρ_{xx} data previously collected, we can calculate the corresponding σ_{xx} with which we further extract the λ_{MF} for each composition.

Contextual Analysis of Ru-Pt Spin Hall Angles

The SHA values derived from ST-MOKE peak at $x = 38$ with a value of 0.117, which is 36% higher than the 0.086 measured for pure Pt under the same conditions. ST-FMR confirms the trend with an even larger SHA of 0.158 at the same composition. The composition-dependent SHA exhibits a skewed parabolic trend with a maximum skewed towards Pt-rich compositions. This is consistent with previous studies on Pt-based alloys where maxima were observed for Pt concentrations between 60 – 90% [112]. This parabolic behavior can be understood in terms of the scattering mechanisms. Pt-rich alloys exhibit strong SOC and low disorder favor skew scattering, which enhances the SHA without significantly increasing resistivity. As Ru content is increased, the lattice transitions towards an HCP structure and additional disorder introduces side-jump scattering contributions which scale significantly with resistivity and reduce the SHA at higher Ru concentrations despite continued alloying.

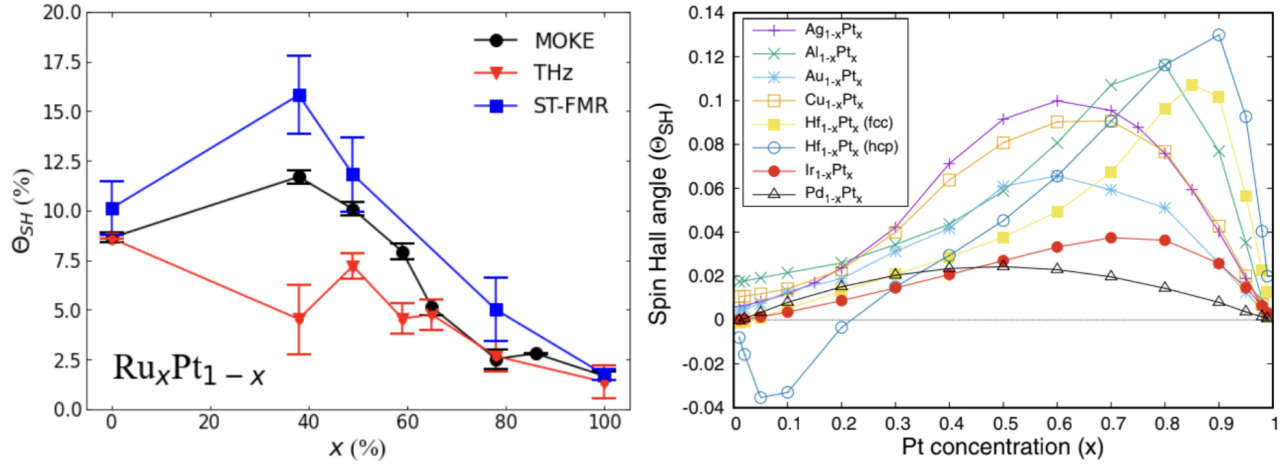


Figure 67: (Left) Summary of SHA data measured for each Ru–Pt alloy composition using three independent techniques. (Right) Comparison of trends in SHA as a function of Pt concentration for various Pt-based alloys [112].

For comparison, the best-performing alloy composition in this study ($Ru_{38}Pt_{62}$) is placed in the context of other well-studied HM systems with regards to resistivity and SHA. The Ru–Pt binary alloy poses as a promising candidate with a competitive SHA and a relatively low room-temperature resistivity compared to leading heavy metals such as β -W and Ta or topological insulators, highlighting its potential as a competitive spin-current source. These material properties are advantageous for energy-efficient spintronic devices compared to higher-resistivity systems. This underscores the advantages of FC-rich, low-impurity Ru–Pt alloys where structural order, moderate resistivity, and skew scattering combine to optimize spin-charge conversion efficiency.

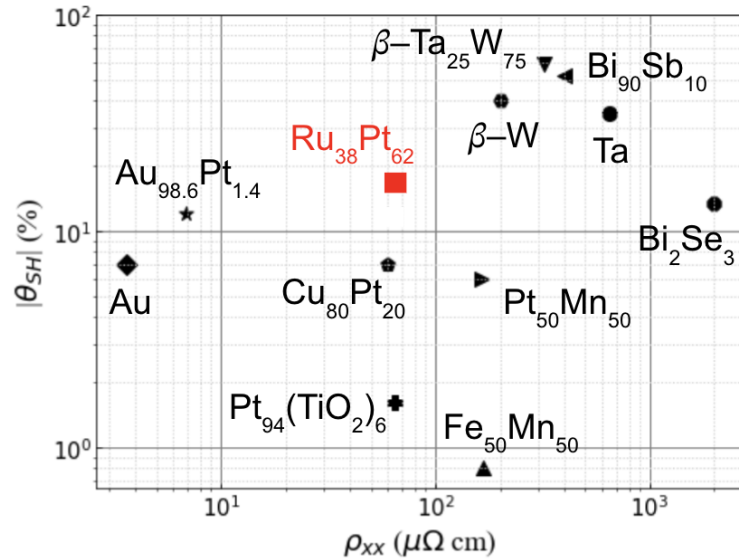


Figure 68: Comparison of maximum SHA from this work ($Ru_{38}Pt_{62}$) to other elements and alloys of interest.

Probing the Scattering Mechanisms

To further understand the contributions to the notable SHA observed in the Ru-Pt alloy system, the SHA is plotted as a function of the alloy resistivity at room temperature, ρ_{xx} . Both the intrinsic mechanism and the side-jump scattering contribution exhibit a linear dependence between SHA and ρ_{xx} . This linear dependence suggests that side-jump is the main contribution to the extrinsic SHA.

To further prove this claim, we employ following equation to separate the intrinsic and extrinsic components of the spin Hall resistivity:

$$-\rho_{SH} = \sigma_{SH}^{int} \rho_{xx}^2 - \rho_{SH}^{imp} \quad (66)$$

where ρ_{SH} is the total spin Hall resistivity of a given $\text{Ru}_x\text{Pt}_{1-x}$ alloy extracted from

$$\theta_{SH} = -\frac{\rho_{SH}}{\rho_{xx}}. \quad (67)$$

Here, σ_{SH}^{int} is the intrinsic contribution of Ru to the spin Hall resistivity, and ρ_{SH}^{imp} is the extrinsic spin Hall resistivity introduced by Pt. Since the phonon contribution to the total spin Hall resistivity is known to be negligible, it is omitted from our analysis. However, the intrinsic contributions of the spin Hall conductivity, σ_{SH}^{int} , is retained because pure Ru exhibits a finite spin Hall angle even though it is roughly an order of magnitude smaller than that observed in $\text{Ru}_{38}\text{Pt}_{62}$.

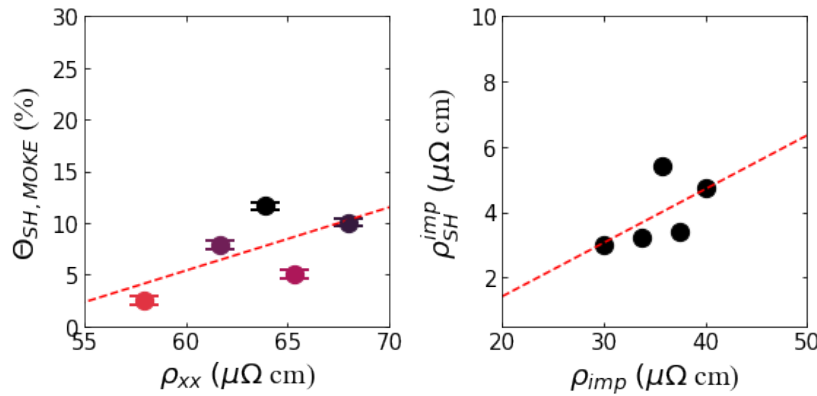


Figure 69: (a) MOKE SHA values plotted as a function alloy resistivity over the alloy region. (b) Extracted values of ρ_{SH}^{imp} versus ρ_{imp} . Linear fits in red.

To quantify this intrinsic term, we examine the limiting case of $x = 100$, where the impurity-related spin Hall resistivity ρ_{SH}^{imp} vanishes and the total resistivity of the alloy reduces to that of pure Ru ($x = 0$). Under this condition, we can rewrite the previous expression as:

$$\sigma_{\text{SH}}^{\text{int}} = -\frac{\rho_{\text{SH}}}{\rho_{\text{Ru}}^2} = \frac{\theta_{\text{SH, Ru}}}{\rho_{\text{Ru}}}, \quad (68)$$

where $\theta_{\text{SH, Ru}}$ and ρ_{Ru} are the SHA and resistivity of pure Ru, respectively. By substituting these relations for $\sigma_{\text{SH}}^{\text{int}}$ and θ_{SH} , the expression simplifies to

$$-\rho_{\text{SH}}^{\text{imp}} = \rho_{xx}\theta_{\text{SH}} - \left(\frac{\theta_{\text{SH, Ru}}}{\rho_{\text{Ru}}}\right)\rho_{xx}^2. \quad (69)$$

We plot $|\rho_{\text{SH}}^{\text{imp}}|$ as a function of ρ_{imp} , which is defined as $\rho_{\text{imp}} = \rho_{xx} - \rho_{\text{Ru}}$. The curve can then be fitted with

$$|\rho_{\text{SH}}^{\text{imp}}| = \sigma_{\text{SH}}^{\text{SS}}\rho_{\text{imp}} + \sigma_{\text{SH}}^{\text{SJ}}\rho_{\text{imp}}^2, \quad (70)$$

where $\sigma_{\text{SH}}^{\text{SS}}$ represent the contribution from skew scattering and $\sigma_{\text{SH}}^{\text{SJ}}$ represent the contribution from side-jump. These terms represent the two mechanisms of the extrinsic SHE. From the fitting, we determine the magnitudes of these two contributions: $\sigma_{\text{SH}}^{\text{SS}} = 0.052$ and $\sigma_{\text{SH}}^{\text{SJ}} = 0.002\mu\Omega^{-1}\text{cm}^{-1}$. We find that for $\rho_{\text{imp}} = 26\Omega\text{cm}$, the contributions from skew scattering and side jump to the extrinsic SHE are comparable. For $\rho_{\text{imp}} > 26\Omega\text{cm}$, the side-jump contribution is larger. Considering that the resistivity of the alloys are all above $40\Omega\text{cm}$, we can conclude that side-jump is the dominant scattering mechanism in the Ru-Pt alloy samples studied here.

Reliability of Measurement Techniques

While the ST-FMR results exhibit the same overall trend as the ST-MOKE SHAs across the entire Ru-Pt composition range, the absolute values obtained from ST-FMR are systematically larger. This discrepancy can be attributed to the presence of artifact contributions in the ST-FMR measurements that are absent in the MOKE technique.

Artifacts in ST-FMR have been widely studied and are known to arise from several competing mechanisms [104, 113–120]. When the FM layer is driven into resonance, additional spin currents can be generated within the FM itself through processes such as spin pumping (SP) and the longitudinal spin Seebeck effect (LSSE). These spin currents flow from the FM layer into the HM layer, where they are converted into a detectable dc voltage via the ISHE. Furthermore, resonant microwave excitation leads to localized heating within the bilayer, creating an out-of-plane thermal gradient. This gradient can induce additional thermoelectric voltages through the ordinary Nernst effect (ONE) or the anomalous Nernst effect (ANE).

Many of these artifact-related voltages contribute a symmetric Lorentzian component to the ST-FMR signal, typically oriented perpendicular to the magnetization axis. If sufficiently strong, these contributions can mask or distort the extraction of the damping-like torque (τ_{DL}), which plays a central role in determining the SHA from ST-FMR analysis.

Another challenge arises from the angular dependence of the signals. Both genuine spin-torque rectification voltages and spurious signals from SP and thermoelectric effects share the same functional dependence under an in-plane magnetic field, following a characteristic $\sin(2\theta)$ behavior, where θ is the angle between the applied field and the current direction [121–123]. This similarity in angular dependence makes it particularly challenging to disentangle artifact-induced contributions from the intrinsic ST-FMR signal in quantitative SOT studies such as this one.

Similarly to the ST-FMR signal contributions, the framework used by THz spectroscopy to calculate the relative SHAs considers that the spin-to-charge conversion is only taking place in the HM layer due to the SOC. However, the process may be a convolution of the conversion in the FM and FM–HM interface as well, which has not been taken into consideration. For the initial change in the signal from $x = 0\%$ to 38% , the FM–HM interface change may also be contributing to the off-trend in THz emission amplitudes. Literature has also predicted that the THz emission from the FM–HM interface may be primarily due to the charge pumping in the FM layer itself [124, 125]. Contributions from FM–HM interface charge pumping can reduce the apparent SHA and may explain the consistently lower SHA values obtained by THz emission spectroscopy [111, 126]

Key Findings

A systematic mapping of SHA versus composition in Ru–Pt alloys using complementary techniques (ST-MOKE, ST-FMR, THz) has not previously been reported. The observation of a maximum SHA of 0.158 in the $\text{Ru}_{38}\text{Pt}_{62}$ alloy composition with resistivity below $70 \mu\Omega \text{ cm}$ is novel and of clear relevance for device applications. Connecting this enhancement to the structural evolution from FCC to HCP represents a noteworthy structure-property correlation with implications for alloy design in spintronics.

From a practical perspective, these large SHA values are achieved in as-deposited sputtered films across the full alloy composition range, eliminating the need for complex growth protocols or post-deposition annealing. These findings highlight Ru–Pt alloys as a scalable and tunable platform for SOT device applications, where

phase evolution itself serves as an effective control parameter for optimizing spin transport. While the broader concept of impurity tuning to enhance SHA is well established in alloys such as CuBi, AuTa, and PtIr, Ru–Pt occupies a distinct niche: it bridges the gap between 4d and 5d metals and provides a rare binary example where structural phase evolution itself acts as a tuning parameter.

Looking Ahead

Looking forward, several key directions emerge. (1) Integrating Ru–Pt with ferromagnetic and antiferromagnetic oxide layers could enable hybrid heterostructures that combine efficient spin-charge conversion with tunable interfacial exchange coupling, extending the concept of all-oxide spintronic platforms. (2) Probing the temperature and frequency dependence of the spin Hall response using ultrafast and high-frequency probes may reveal new insights into dynamic SOTs and magnon-spin current interactions. (3) The compositional flexibility of the Ru–Pt system opens the door to exploring ternary extensions such as Ru–PtIr or Ru–PtOs, where additional spin-orbit channels or Berry curvature hot spots may further enhance spin conversion efficiency.

Beyond immediate materials optimization, this work points to a broader design principle: crystal symmetry and electronic topology can be engineered to tailor the flow of spin and charge. The Ru–Pt study thus represents not only the culmination of the spin current pipeline from generation in CrO_2 to transmission in $\text{RuO}_2/\text{CrO}_2$ and conversion in Ru–Pt, but also a blueprint for discovering and controlling spin-orbit phenomena across a wide range of correlated and metallic systems.

Conclusion

REVISITING THE MOTIVATION

At the most fundamental level, this dissertation aims to understand how spin currents (i.e. collective flows of angular momentum) can be generated, transmitted, and converted across materials with distinct magnetic and electronic symmetries. As the field of spintronics moves beyond conventional ferromagnet/heavy-metal stacks toward all-oxide and strongly correlated systems, the interfacial and spin-orbit-driven mechanisms underlying spin transport become increasingly rich and complex. My work explores this frontier through three carefully chosen systems: CrO_2 , $\text{RuO}_2/\text{CrO}_2$, and Ru-Pt alloys. Together, these candidate systems map out the full life cycle of a spin current from its intrinsic generation in a half-metal, to its transmission across a spin-orbit-active interface, and finally to its extrinsic conversion into charge.

While each material system has its own microscopic mechanisms, unifying themes include spin-orbit coupling (SOC) and broken symmetry at interfaces which dictate the efficiency and characteristics of the resulting spin transport. SOC not only links spin and momentum, but also acts as the bridge between magnetic order and electronic topology, allowing purely electronic structure effects to manifest as measurable magnetic phenomena. This dissertation demonstrates how tuning SOC through composition, crystal symmetry, and dimensionality can control the fundamental processes that underlie spintronic function.

A Unified Picture of Spin Current Evolution

In epitaxial CrO_2 , the story begins with the intrinsic generation of spin-polarized currents. As a half-metal with nearly complete spin polarization at the Fermi level, CrO_2 provides a unique platform where the spin degree of freedom is not an additional variable but a defining feature of conduction itself. The observation of quantum oscillations and coherent electronic transport reveals that the

spin-polarized carriers in CrO_2 maintain phase coherence over long distances, enabling a regime where quantum transport and spin transport are inseparable. This establishes CrO_2 as a model for understanding intrinsic spin current generation in systems where magnetism and electronic structure are strongly intertwined.

At the next stage, the $\text{RuO}_2/\text{CrO}_2$ bilayer shifts the focus from generation to transmission. Here, the interface between an antiferromagnet (RuO_2) and a ferromagnet (CrO_2) serves as a natural system for probing how spin information is transferred across dissimilar magnetic orders. The interplay between interfacial exchange, spin reorientation, and SOC reveals a complex picture. Even in the absence of long-range order coupling, the interface can mediate effective exchange bias and modify the spin-wave spectrum of the underlying ferromagnet. These findings extend our understanding of spin transmission beyond the classical exchange bias paradigm, showing that spin communication across oxide interfaces can be driven as much by spin-orbit interaction and anisotropy as by conventional exchange coupling.

Finally, in Ru–Pt alloys, the story culminates in the conversion of spin current into charge current. By systematically mapping the spin Hall angle across the compositional and structural phase space, this work demonstrates how SOC and crystal symmetry jointly determine the efficiency of spin-charge interconversion. The discovery that structural phase evolution, rather than impurity scattering alone, can tune the spin Hall response establishes a new design scheme. Here, crystal structure itself can act as a continuous control parameter for SOC-driven effects. In this way, Ru–Pt provides both a mechanistic insight and a materials blueprint for engineering efficient spin-orbit torque platforms.

Taken together, these three studies reveal a coherent narrative of spin transport: CrO_2 generates spin-polarized currents, $\text{RuO}_2/\text{CrO}_2$ transmits them across a complex interfacial barrier, and Ru–Pt converts them into charge signals. This progression reflects the fundamental processes underlying all spintronic devices, yet explored here in materials that highlight the physics of each stage with clarity using novel techniques.

Broader Physical Insights

The broader significance of this work lies in demonstrating that spin transport is not a single material property but an emergent, multiscale phenomenon arising from the coupling between electronic structure, magnetic order, and interfacial symmetry. Several key in-

sights emerge:

- **Interfacial spin coupling as a SOC-mediated process:** In oxide heterostructures, SOC can serve as an effective channel for spin exchange even in the absence of direct orbital overlap. This reframes the notion of exchange bias as merely a static pinning effect, but rather a dynamic reflection of interfacial spin-orbit entanglement.
- **Quantum coherence in half-metals:** The persistence of quantum oscillations in CrO_2 suggests that fully spin-polarized systems can sustain coherent transport typically associated with nonmagnetic conductors. This bridges the conceptual gap between quantum materials physics and spintronics, showing that half-metallic ferromagnets can host topological or Berry-phase-driven transport phenomena traditionally reserved for nonmagnetic topological systems.
- **Structural control of spin-orbit effects:** The Ru–Pt study demonstrates that manipulating crystal symmetry (i.e. transitioning from cubic to hexagonal coordination) provides a powerful means of tuning SOC strength and spin Hall response. This suggests that lattice symmetry can be an equally powerful lever as chemical composition in governing spin-charge coupling.

Collectively, these insights deepen our understanding of how SOC mediates spin current behavior across different energy and length scales. They move the field toward a unified description of spin transport grounded in symmetry, topology, and collective excitations rather than purely phenomenological parameters.

Extending the Principles to Other Systems

The principles developed here extend naturally to a wide class of materials beyond those studied in this work. In oxide heterostructures, the sensitivity of spin transmission to interfacial symmetry and anisotropy opens the door to designing engineered spin-orbit-active interfaces using perovskite oxides, antiferromagnets, or topological insulators. Similarly, the demonstration of spin-coherent transport in CrO_2 suggests that other half-metallic systems such as Fe_3O_4 or double perovskites could host quantum oscillations or topological spin textures under appropriate strain or dimensional confinement.

In metallic systems, the structural tunability observed in Ru–Pt points toward a new class of compositionally flexible spin-orbit materials, where the interplay of disorder, phase transitions, and lattice symmetry can be harnessed to tailor spin-orbit torque efficiency. This

approach could guide the design of future device platforms where crystalline order and spin current conversion are co-optimized.

At a conceptual level, the unifying idea is that spin transport can be coherently engineered by controlling SOC and symmetry across interfaces. This principle transcends specific material chemistries and applies equally to oxides, half metals, and complex alloys, providing a road map for integrating spin functionality into diverse technological contexts from memory and logic devices to quantum sensing.

Outlook and Future Directions

Looking ahead, several avenues emerge from this work. On the experimental front, coupling the CrO_2 and $\text{RuO}_2/\text{CrO}_2$ systems with direct spin transport probes, such as spin Seebeck or spin pumping measurements, could quantitatively establish the efficiency of spin transmission and the role of interfacial SOC in spin angular momentum dissipation. Time-resolved or angle-resolved spectroscopy could further elucidate how interfacial magnetic states evolve dynamically under applied current or field.

For Ru–Pt and related alloys, extending the phase-space mapping to include epitaxial strain, substrate effects, and temperature-dependent transport could uncover new regimes of spin Hall enhancement, potentially approaching those of topological materials but in simpler metallic hosts. On the theoretical side, *ab initio* modeling of $\text{RuO}_2/\text{CrO}_2$ and Ru–Pt could provide microscopic insight into the SOC-induced spin textures and Fermi surface anisotropies responsible for the observed phenomena.

In the broader context of spintronics, the conceptual framework advanced here points toward a materials-by-design approach where SOC strength, lattice symmetry, and interfacial structure are jointly optimized to control the full spin current pathway. The insights gained may inform the design of next-generation spin-orbit torque memory elements, oxide-based spin transistors, and hybrid systems that couple spin transport with superconductivity or ferroelectricity. Furthermore, the demonstration of quantum-coherent and spin-polarized transport in CrO_2 suggests intriguing possibilities for integrating half-metals into quantum information or neuromorphic architectures, where spin coherence and dissipation-less control are essential.

Closing Remarks

In summary, this dissertation presents a cohesive exploration of how spin-orbit coupling and interfacial symmetry govern the flow of spin

information across complex materials. From the intrinsic quantum transport in CrO_2 to the interfacial exchange in $\text{RuO}_2/\text{CrO}_2$ and the spin Hall conversion in Ru–Pt, the findings together construct a unified picture of spin current evolution. Beyond the novel individual discoveries, the overarching insight is that spin transport can be coherently designed and tuned, not merely observed, by leveraging SOC and symmetry as active control parameters.

This principle bridges the gap between fundamental spin physics and practical spintronic design. It shows that the same underlying interactions responsible for magnetism, anisotropy, and quantum coherence can be orchestrated to realize scalable, energy-efficient devices. In doing so, the work not only deepens our understanding of spin transport mechanisms but also provides a conceptual foundation for the next generation of materials and technologies built upon the spin degree of freedom.

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