



Trinity College Dublin
Coláiste na Tríonóide, Baile Átha Cliath
The University of Dublin

SCHOOL OF PHYSICS

QUANTUM TRANSPORT IN ANTIFERROMAGNETIC SPINTRONICS

WILLY ROBERT LABORDE

SUPERVISED BY DR. MARIA STAMENOVA & PROF. STEFANO
SANVITO

APRIL 2026

SUBMITTED IN PARTIAL FULFILMENT OF THE REQUIREMENTS FOR THE DEGREE OF

DOCTOR IN PHILOSOPHY - SCHOOL OF PHYSICS

Nothing in life is to be feared, it is only to be understood.
Now is the time to understand more, so that we may fear less.
Marie Curie

This thesis is the culmination of years of effort, and it would not have been possible without the guidance, support, and encouragement of many individuals to whom I am deeply grateful. First and foremost, I wish to express my most sincere gratitude to my supervisor, Professor Stefano Sanvito, for his mentorship, unwavering support, and scientific insight throughout this PhD journey. His guidance gave me the direction to navigate complex problems while allowing me the freedom to explore my own ideas. I am also deeply thankful to my co-supervisor, Doctor Maria Stamenova, for her insightful perspectives, constant encouragement and help on a daily basis on this entire work. I extend my thanks to all members of the Computational Spintronics research group and all the Physics Section of Trinity College Dublin. The stimulating discussions, collaborative spirit, and daily camaraderie created an environment where this work could flourish. I am grateful for the countless moments of shared knowledge and mutual support. The computationally intensive nature of this research would have been impossible without the provision of computational facilities and support by Research IT of Trinity College Dublin and the Irish Centre for High-End Computing (ICHEC). Some of the results of this thesis were derived on Kay Cluster part of ICHEC. Some calculations were performed on the Kelvin and Lanczos cluster maintained by the Trinity Centre for High Performance Computing. Kelvin cluster was funded through grants from the Higher Education Authority, through its PRTL program. Many thanks to the diligent assistance of their technical staff. This work was made possible by the generous financial support from Science Foundation

of Ireland (SFI) under grant number 15821. To my friends, both within and outside of academia, thank you for the much-needed perspective, laughter and encouragement that kept me grounded. Finally, my deepest and most heartfelt thanks go to my family. To my parents, and finally to my wife, Adeline, for your unconditional love, endless patience, and unwavering belief in me.

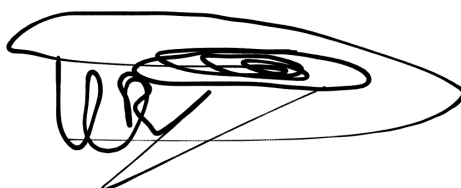
Dublin, April 21, 2026

W. R. L.



Declaration

I declare that this thesis has not been submitted as an exercise for a degree at this or any other university and it is entirely my own work. I agree to deposit this thesis in the University's open access institutional repository or allow the Library to do so on my behalf, subject to Irish Copyright Legislation and Trinity College Library conditions of use and acknowledgement. I consent to the examiner retaining a copy of the thesis beyond the examining period, should they so wish (EU GDPR May 2018).





Summary

This thesis presents a first-principles theoretical investigation into the electronic, magnetic, and quantum transport properties of altermagnets, a recently discovered third class of collinear magnetic materials. It aims to bridge the gap between fundamental theory and device application focusing on the prototypical yet controversial altermagnet, Ruthenium Dioxide (RuO_2). Indeed, RuO_2 shows potential for use in high-performance spintronic devices.

The research presented in this manuscript is motivated by the limitations of conventional spintronics and the promise of antiferromagnetic materials which offer robustness, zero stray fields, and ultrafast dynamics. Altermagnets, a theoretically predicted third phase of magnetism, uniquely combine these advantages with the large, non-relativistic spin-splitting of electronic bands characteristic of ferromagnets. They can offer a novel platform for device engineering. This work confronts the central controversy surrounding RuO_2 , where compelling theoretical predictions and experimental reports of altermagnetism are challenged by high-precision measurements that find no evidence of intrinsic magnetic order.

The methods employed are rooted in state-of-the-art computational physics. The electronic and magnetic ground states of materials are determined using Density Functional Theory. To overcome the known failures of standard exchange-correlation functionals for this class of materials, advanced techniques are implemented, including the parametric DFT+ U method and, most centrally, the parameter-free Atomic Self-Interaction Correction method. Quantum transport properties are simulated using the Non-Equilibrium Green's Function (NEGF) formalism as implemented in the Smeago1 TCD in-house code, which is coupled to the SIESTA DFT package. This allows for a microscopic, atomic-level under-

standing of current flow in nanostructured devices under both zero and finite bias voltages. Furthermore, a systematic materials screening methodology, based on a set of physically-grounded "golden rules" is developed to identify experimentally viable heterostructures for realisable altermagnetic tunnel junctions.

The major findings of this thesis are threefold. First, our computational investigation into the RuO₂ ground state confirms that standard DFT functionals fail to stabilize a magnetic moment, consistent with experiments on pristine crystals. We demonstrate that the altermagnetic phase, with its characteristic *d*-wave spin-splitting, can be robustly stabilized by applying a physically-motivated, selective self-interaction correction to the Ruthenium atoms. This establishes a reliable and justified computational model for the material, providing a crucial theoretical benchmark amidst the experimental controversy.

Second, using this model, we designed and simulated an altermagnetic tunnel junction (ALMTJ) based on a RuO₂ / TiO₂ / RuO₂ heterostructure. Our zero-bias transport calculations predict a giant tunnelling magnetoresistance (TMR) of approximately 427% for a four-monolayer TiO₂ barrier. This result validates the core concept of altermagnetic spintronics: that a large, technologically relevant magnetoresistance can be achieved in a device with zero net magnetization.

Third, and most significantly, this thesis presents the first investigation of this ALMTJ under finite bias conditions. The calculations reveal a novel, non-equilibrium phenomenon: a giant inversion of the TMR. The TMR is shown to decay from its large positive value at zero bias, passing through zero and becoming strongly negative at a critical voltage of approximately 0.35 V. Through a detailed analysis of the momentum-resolved transmission channels, we identify the microscopic origin of this effect as a bias-induced spin-symmetry filtering mechanism. The applied voltage acts as a spin-selective switch, activating a dormant, highly conductive transport channel in the antiparallel magnetic state, causing its conductance to surpass that of the parallel state.

In conclusion, this thesis provides a comprehensive theoretical framework for understanding and designing altermagnetic spintronic devices. It clarifies the computational conditions required to model the prototypical altermagnet RuO₂, predicts the high performance of a RuO₂ / TiO₂ / RuO₂ device, and discovers a fundamental, bias-driven TMR inversion. These findings offer critical insights for the ongoing experimental efforts and lay the groundwork for a new class of non-volatile, high-speed, and field-robust memory technologies.



Abstract

Antiferromagnetic spintronics represents a promising evolution beyond conventional ferromagnet-based technologies. By utilizing materials with zero net magnetization, antiferromagnets offer intrinsic advantages such as robustness against magnetic perturbations, the absence of stray fields for ultra-dense memory integration, and terahertz-scale dynamics for ultrafast operation. Despite this potential, their compensated magnetic order has been very difficult to study with simple electrical probes, posing significant challenges for reading and writing information. Altermagnetism, a third theoretically predicted magnetic phase, presents a new frontier for spintronics by combining the large, non-relativistic spin-splitting of ferromagnets with the vanishing net magnetization of antiferromagnets. This thesis explores the fundamental physics and device potential of this new magnetic class, centred on its prototypical yet highly controversial material candidate, Ruthenium Dioxide (RuO_2). The research directly confronts the primary obstacle in the field: the ongoing debate over whether RuO_2 possesses an intrinsic magnetic ground state. This work navigates the landscape of conflicting experimental reports by providing a complete computational overview and establishing a theoretical benchmark for the performance of an ideal altermagnetic RuO_2 device. The thesis delivers three primary contributions. First, it presents a comprehensive computational characterization of RuO_2 , demonstrating that a parameter-free approach using the Atomic Self-Interaction Correction (ASIC) method provides a physically sound model for its hypothesized altermagnetic phase. Second, it establishes a general "golden rule" framework for the systematic design of novel, experimentally viable altermagnetic tunnel junctions (ALMTJs). This design recipe, based on screening for metallic character, high Néel temperature, and structural compatibility, successfully identifies promising new heterostructures, such as $\text{CrSb} / \text{NiTe}$, demonstrating its utility beyond a single material. The third and central contribution is the first-ever

theoretical investigation of finite-bias quantum transport in a prototypical RuO₂ / TiO₂ / RuO₂ junction, performed using a combination of Density Functional Theory and Non-Equilibrium Green's Function (NEGF) formalism. These first-principles simulations predict a giant tunnelling magnetoresistance (TMR) of approximately 427% at zero bias. Critically, the calculations reveal novel non-equilibrium physics, discovering a remarkable inversion of the TMR from large positive to large negative values at a finite bias of around 0.35 V. This effect is traced to a bias-induced spin-symmetry filtering mechanism. By delivering a robust computational model for RuO₂, a tangible framework for future materials discovery, and the discovery of novel transport phenomena, this thesis provides a critical theoretical foundation for the advancement of altermagnetic spintronics.



Contents

Declaration	v
Summary	vi
Abstract	i
I Introduction	1
0.1 The Dawn of Spintronics: From Giant Magnetoresistance to Magnetic Tunnel Junctions	3
0.2 The Next Frontier: Antiferromagnetic Spintronics	4
0.3 Altermagnetism: A Third Fundamental Magnetic Phase	5
0.4 RuO ₂ : The Prototypical Altermagnet Candidate	6
0.5 The Central Problem and Thesis Motivation	7
0.6 Thesis Outline	8
II Spintronics General Concepts	10
1 From Magnetic Symmetries to the Altermagnetic Phase	11
1.1 The Promise and Paradox of Antiferromagnetic Spintronics	11
1.2 A New Foundation: The Theory of Spin Symmetry Groups	16
1.3 Re-classification: The Three Non-Relativistic Magnetic Phases	17
1.4 Beyond Collinear Order: The Emergence of Anti-Altermagnetism	19
1.5 Signatures and Consequences of the Altermagnetic Phase	23
1.5.1 Momentum-Dependent Spin Splitting	23
1.5.2 Alternating Spin Polarization in Real and Momentum Space	24

1.5.3	Anisotropic Magneto-transport and Time-Reversal Symmetry Breaking	24
1.5.4	"Wave" Symmetry of the Spin Splitting	25
1.6	Theoretical Limitations and Open Questions	26
1.6.1	The Challenge of a Unique Order Parameter	26
1.6.2	The Non-Relativistic Idealization versus Real Materials	26
1.6.3	Microscopic Origins and Strong Correlation	27
1.6.4	Collective Excitations: The Quest for the Altermagnon	28
1.6.5	The Question of Generality and Material Realizations	28
1.7	Conclusion and Bridge to Device Physics	28
2	Computational Methodology	30
2.1	Fundamentals of Density Functional Theory (DFT)	31
2.2	Spin-Polarized Density Functional Theory	34
2.3	Beyond Standard DFT: Treating Correlation and Self-Interaction	35
2.3.1	The DFT+ U Method: A Correction for On-Site Correlation	36
2.3.2	ASIC: A Parameter-Free Correction for Self-Interaction Error	37
2.3.3	Comparison with Alternative Advanced Functionals	39
2.4	Simulating Open Quantum Systems: The NEGF-DFT Formalism	40
2.5	Convergence and Numerical Precision	44
2.5.1	Ground-State DFT Convergence	44
2.5.2	Quantum Transport Convergence	44
2.5.3	Sensitivity to Correlation Parameters	45
2.5.4	Chapter Conclusion	46
III	Altermagnetism and Spintronics Application	47
3	Ruthenium Dioxide RuO_2: A Material at the Crossroads of Magnetism	48
3.1	Fundamental Properties of Ruthenium Dioxide	49
3.2	The Magnetic Controversy: A Tale of Conflicting Evidence	50
3.2.1	The Traditional View: A Pauli Paramagnet	50
3.2.2	A Paradigm Shift: The Altermagnetic Picture	51
3.2.3	The Refutation: Evidence for a Non-Magnetic Ground State	52
3.2.4	Reconciling the Discrepancies: The Role of Stoichiometry, Strain, and Defects	54
3.3	Relevance for Spintronics under the Altermagnetic Hypothesis	58

3.4	Computational Investigation of the RuO ₂ Ground State	62
3.4.1	Computational Methodology	62
3.4.2	Magnetic Instability in Standard DFT: The Effect of Strain	65
3.4.3	A Survey of Computational Approaches in the Literature	69
3.4.4	Inducing Magnetism: Electron Correlation and Self-Interaction	74
3.5	Hall Effects as a Probe of the RuO ₂ Ground State	79
3.5.1	The Anomalous Hall Conductivity: A Signature Dependent on Magnetic Order	80
3.5.2	Computational Challenges: The Sensitivity of Wannier Interpolation	82
3.5.3	Methodological Validation: The Spin Hall Conductivity	86
4	Design and Quantum Transport in Altermagnetic Tunnel Junctions	90
4.1	The Promise of Altermagnetic Tunnelling	91
4.2	A "Golden Rule" Framework for Antiferromagnetic Tunnel Junction Design	92
4.2.1	The Golden Rules for AFMTJ / ALMTJ Design	93
4.3	Candidate Materials and Heterostructure Design	96
4.3.1	The RuO ₂ / TiO ₂ / RuO ₂ System: A Prototypical Case Study	96
4.3.2	The CrSb/ NiTe System: A High-Temperature Candidate	98
4.3.3	The Case of SST-4 CrSe: Challenges in Insulator Matching	98
4.4	The RuO ₂ / TiO ₂ / RuO ₂ Model System	99
4.4.1	Device Geometry and Magnetic States	99
4.4.2	Computational Methodology for Quantum Transport	101
4.5	Zero-Bias Transport Properties and Tunnelling Magnetoresistance	103
4.5.1	Giant Tunnelling Magnetoresistance verification in the ASIC Model	103
4.5.2	Effect of Barrier Thickness and the Failure of LDA	105
4.6	Quantum Transport under Finite Bias	106
4.6.1	Theoretical Framework for Finite-Bias Transport	107
4.6.2	Transmission Spectra under Finite Bias	107
4.6.3	Current-Voltage Characteristics and Giant TMR Inversion	108
4.7	Microscopic Origin of the Bias-dependent TMR	110
4.7.1	$\mathbf{k}_{\perp}$ -resolved Transmission Analysis at Zero and Finite Bias	111
4.7.2	Bias-Induced Symmetry Filtering	114
4.8	Limitations: The Pristine Interface Assumption	115
4.9	Limitations: The Non-Relativistic Approximation and Spin-Orbit Coupling	117

IV Outlook, Future work and Conclusion	119
5 Outlook and Conclusion	120
5.1 Summary of Key Findings	120
5.2 Outlook and Future Research Directions	122
5.2.1 Experimental Directions	122
5.2.2 Theoretical Directions	123
5.2.3 Long-Term Vision	124
A List of 2D and 3D Altermagnet Candidates	127
B Table of Spin-splitting Types 4 & 5 Magnetic Space Groups	135

Introduction **Part I**



Introduction

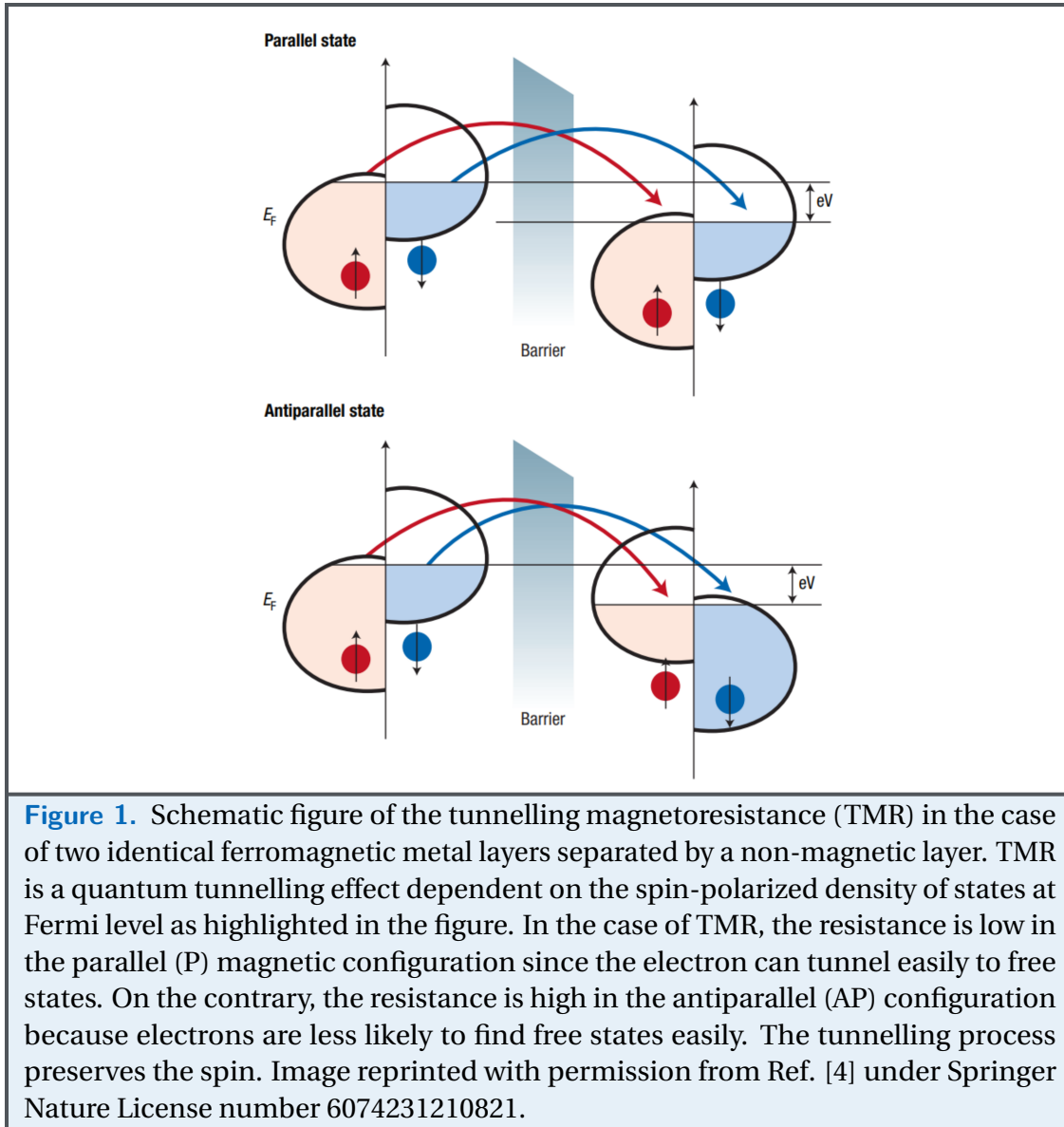
The relentless progress of modern information technology, governed for decades by the empirical trend of Moore's Law, is encountering fundamental physical limits in device scaling, speed, and energy efficiency. Conventional electronics, which is predicated on the manipulation and storage of the electron's charge, faces significant challenges related to power dissipation and quantum tunnelling at the nanoscale. In this context, spintronics or spin-electronics, has emerged as an innovative paradigm shift by exploiting an additional intrinsic property of the electron: its spin [1]. By exploiting the spin degree of freedom, spintronics offers pathways to novel devices that are non-volatile, faster, and more energy-efficient than their charge-based counterparts.

This introductory chapter provides the motivation and context for the research presented in this thesis. First, the introduction begins by tracing the origins of spintronics from the discovery of giant magneto-resistance (GMR). Then, it explores the evolution of the field towards antiferromagnetic (AFM) materials, which offer unique advantages over conventional ferromagnets (FM). Subsequently, it introduces the recent theoretical breakthrough of altermagnetism (ALM), a third fundamental magnetic phase that promises to unlock unprecedented spintronic phenomena. This leads to the central material of this work, Ruthenium Dioxide (RuO_2), a key altermagnet proof-of-concept material, and the scientific controversy surrounding its magnetic nature. Finally, the following sections outline the central objectives and structure of this thesis, which aims to navigate this complex landscape through first-principles computational modelling.

— 0.1 The Dawn of Spintronics: From Giant Magnetoresistance to Magnetic Tunnel Junctions —

The field of spintronics was born from the discovery of giant magneto-resistance in 1988, an achievement recognized with the Nobel Prize in Physics awarded to Albert Fert and Peter Grünberg [2, 3]. GMR occurs in metallic multilayer structures, typically composed of two ferromagnetic (FM) layers separated by a thin, non-magnetic metallic spacer (e.g., Fe/Cr/Fe). The electrical resistance of the structure was found to depend dramatically on the relative orientation of the magnetisations of the two FM layers. When the magnetisations of the ferromagnetic layers are parallel (P state), electrons with spin parallel to the magnetisation experience low scattering and traverse the device with low resistance. Conversely, when the magnetisations are antiparallel (AP state), electrons of both spin orientations experience strong scattering in one of the FM layers, resulting in a significantly higher overall resistance. This ability to control an electrical current with a magnetic configuration was the first major demonstration of active spin control and rapidly led to the development of highly sensitive read heads for magnetic hard-disk drives, revolutionizing the data storage industry [4]. A parallel and equally significant development was the realization of tunnelling magneto-resistance (TMR). First proposed by Jullière in 1975 [5], the effect was only harnessed effectively decades later with the advent of high-quality magnetic tunnel junctions (MTJs). An MTJ consists of two ferromagnetic electrodes separated by an insulating barrier. In this case, transport is governed by spin-conserving quantum tunnelling. The TMR arises from the difference in the electronic density of states (DOS) for spin-up and spin-down electrons at the Fermi level in the FM electrodes. In the P configuration, electrons from the majority-spin band of the first electrode can tunnel into the empty majority-spin states of the second, leading to a high conductance. In the AP configuration, the majority-spin electrons of the first electrode face the minority-spin band of the second, which has a mismatching DOS at the Fermi level. Tunnelling is thus strongly suppressed, leading to a state of high resistance. The development of MTJs with crystalline magnesium oxide (MgO) barriers led to TMR ratios of several hundred percent at room temperature, paving the way for non-volatile magnetic random-access memory (MRAM)

[6, 7].



0.2 The Next Frontier: Antiferromagnetic Spintronics

While spintronics based on ferromagnets has been remarkably successful, these materials possess inherent limitations, including the generation of stray magnetic fields that interfere with neighbouring devices and a vulnerability to external magnetic perturbations. Antiferromagnetic (AFM) materials, in which magnetic moments on neighbouring atoms align in an antiparallel fashion, naturally overcome these issues. Their zero net magnetisation makes them robust against external fields and eliminates stray fields, allowing for denser device integration. Furthermore, their intrinsic dynamics are orders of magnitude faster

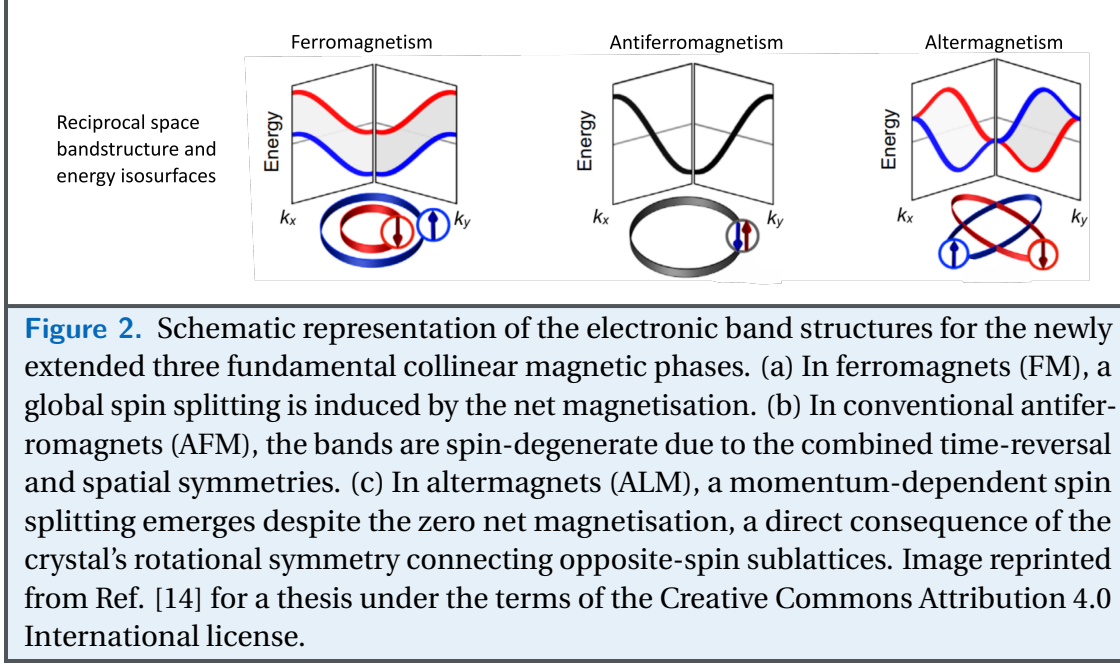
(in the THz range) than those of ferromagnets (GHz range) [8, 9]. Namely, the dynamics is driven by stronger exchange interactions between sublattices as opposed to relativistic effects due to spin-orbit coupling (magnetic anisotropy) in FMs.

For many years, the same properties that make AFMs attractive also made them difficult to use; the absence of a net moment meant they were notoriously difficult to encode information into magnetic domains. However, recent breakthroughs have demonstrated that AFM order can be manipulated and detected electrically, primarily through spin-orbit torques and novel magnetoresistive effects [10]. Seminal work showed that an AFM can even act as the active component in a tunnel junction, producing a spin-valve-like effect [11]. This has opened up the vibrant field of AFM spintronics which seeks to exploit the unique properties of antiferromagnets for next-generation spintronic technologies.

— 0.3 Altermagnetism: A Third Fundamental Magnetic Phase —

For over half a century, the landscape of collinear magnetism was described by a simple dichotomy: ferromagnetism and antiferromagnetism. This classification, rooted in the framework of Shubnikov magnetic space groups, seemed exhaustive. However, a recent theoretical breakthrough has unveiled what has been seen as a third fundamental magnetic phase: altermagnetism [12, 13, 14]. So-called altermagnets are materials like conventional AFMs, have a collinear, compensated magnetic order and thus a vanishing net magnetisation. The crucial difference lies in the crystal symmetry that connects the opposite-spin sublattices. In conventional AFMs, this symmetry operation is either a lattice translation or an inversion. In altermagnets, the opposite-spin sublattices are connected by a crystal rotation (e.g., a 90-degree rotation). Crucially, non-magnetic atoms are essential in the lattice for such symmetry. The crystallographic and the spin space are decoupled construction on the group theory discarding the spin-orbit coupling (SOC) possibility. This seemingly subtle distinction in symmetry has a profound consequence: it allows for a large, momentum-dependent spin splitting of the electronic bands, even in the absence of net magnetisation or relativistic SOC. The resulting band spin splitting along specific directions in altermagnets is hence a non-relativistic effect originating from the crystal field, and can be orders of magnitude larger than typical SOC-induced splittings. This phenomenon was systematically classified using a more general spin-group theory, which

decouples spin and crystal space symmetries, leading to a new materials classification scheme compared to existing magnetic space group derivation [15, 16]. Altermagnetism thus potentially combines the most sought-after properties of both ferromagnets (large spin polarization) and antiferromagnets (zero stray fields, ultrafast dynamics), making it an exceptionally promising application for spintronics.



0.4 RuO₂ : The Prototypical Altermagnet

Candidate

Concurrently with the theoretical prediction, the metallic rutile-structured oxide RuO₂ was identified as the canonical material candidate for this new magnetic phase [12]. Its crystal structure possesses the requisite symmetries to host a large, non-relativistic spin splitting with a characteristic "d-wave" symmetry in momentum space. This identification sparked a flurry of theoretical, computational and experimental activity. Theoretically, RuO₂ was predicted to exhibit giant magneto-transport phenomena, including an extremely efficient electrical spin-splitter effect [17] and a TMR effect exceeding 500% in an altermagnetic tunnel junction (ALMTJ) based on the RuO₂ / TiO₂ / RuO₂ structure [18]. These predictions were quickly followed by experimental reports of altermagnetic signatures in RuO₂ thin films, such as a large anomalous Hall effect [19], current-induced spin-splitter torques capable of switching an adjacent ferromagnet [20, 21]. Even the observation of time-reversal symmetry breaking in its band structure via magnetic circular dichroism in photoemission

was reported [22].

— **0.5 The Central Problem and Thesis Motivation**

Despite the wave of exciting results positioning RuO₂ as the flagship altermagnet, the fundamental premise of intrinsic and stable magnetic order in the material has recently come under serious questioning. The initial report of itinerant antiferromagnetism from [23] is now directly challenged by a series of high-precision experimental studies using techniques such as muon spin rotation (μ SR) and quantum oscillations, which find no evidence of static magnetic order in high-quality bulk single crystals [24, 25, 26]. These experimental findings are supported by theoretical work suggesting that the magnetic ground state in RuO₂ is extremely fragile and may be suppressed by quantum fluctuations or be highly sensitive to defects and strain [27]. Importantly, however, a growing body of theoretical evidence suggests that this fragility cuts both ways: epitaxial strain imposed by a suitable substrate can conditionally stabilize the altermagnetic phase in thin films even when it is absent in the bulk [28], implying that the experimental picture depends critically on growth conditions rather than on a fixed intrinsic material property.

This controversy creates a critical knowledge gap and it revealed itself progressively as the central motivation for this thesis. It is now imperative to establish a clear and comprehensive theoretical picture to guide future experimental efforts. This research addresses this challenge by pursuing three primary objectives:

1. To conduct a thorough computational characterization of RuO₂, providing definitive theoretical predictions for its electronic and magnetic properties.
2. To perform the first theoretical investigation of quantum transport in an altermagnetic tunnel junction, specifically the RuO₂ / TiO₂ / RuO₂ system under finite bias conditions along with the TMR prediction.
3. To establish a general "design recipe" for constructing experimentally viable altermagnetic or antiferromagnetic tunnel junctions, bridging the gap between theoretical prediction and practical realization.

To tackle this rich spintronic physics, first-principles simulations are an indispensable

tool. By employing density functional theory (DFT) with advanced corrections for electron correlation (DFT+U) and self-interaction correction (ASIC), in conjunction with the non-equilibrium Green's function (NEGF) formalism for quantum transport, this work provides a comprehensive, microscopic understanding of the governing phenomena. Ultimately, this research aims to clarify the potential of RuO₂ and lay a foundational framework for the design of next-generation spintronic devices based on the novel physics of altermagnetism.

— 0.6 Thesis Outline —

The remainder of this thesis is structured as follows:

- **Chapter 2: The Physics of Altermagnetism** provides a dedicated review of the concept of altermagnetism, covering its symmetry-based origins, its key physical signatures, and its distinction from conventional ferromagnetism and antiferromagnetism.
- **Chapter 3: Computational Methodology** details the theoretical and computational framework used throughout this work, including Density Functional Theory, the non-equilibrium Green's function method for quantum transport, and advanced corrections such as DFT+*U* and ASIC.
- **Chapter 4: The Case of RuO₂** presents our core results on the material itself. It begins with an extensive literature review of the ongoing magnetism controversy before detailing our own first-principles characterization of its electronic and magnetic properties.
- **Chapter 5: Quantum Transport in an Altermagnetic Tunnel Junction** focuses on the device application, presenting the first theoretical analysis of the RuO₂ / TiO₂ / RuO₂ under zero and finite bias. We evaluate its potential for TMR, STT, and spin-filtering effects.
- **Chapter 6: Conclusion and Outlook** summarizes the main findings of this research, discusses their implications for the field, and provides an outlook on promising future research directions in altermagnetic spintronics.

Together, these contributions aim to establish a rigorous theoretical foundation for the

emerging field of altermagnetic spintronics.

Spintronics General Concepts **Part II**

1 From Magnetic Symmetries to the Altermagnetic Phase

The exploration of spintronics has, for most of its history, been dominated by the physics of ferromagnets. However, the fundamental limitations of ferromagnetic materials have pushed the scientific community to explore new classes of magnetic order. This chapter delves into the theoretical foundations of one such frontier: the physics of compensated magnets, which have zero net magnetization. The goal is to provide a rigorous theoretical foundation for altermagnetism, recently proclaimed as the third fundamental class of magnetism. The narrative begins by exploring the promise and the central paradox of antiferromagnetic spintronics, viewed through the lens of traditional magnetic group theory. Then, it introduces the novel proposition of non-relativistic spin-group theory that ultimately resolved this paradox. This new framework not only provides a more fundamental understanding of conventional antiferromagnetism but also leads directly to the discovery of altermagnetism. Altermagnetism predicts unique physical signatures and properties.

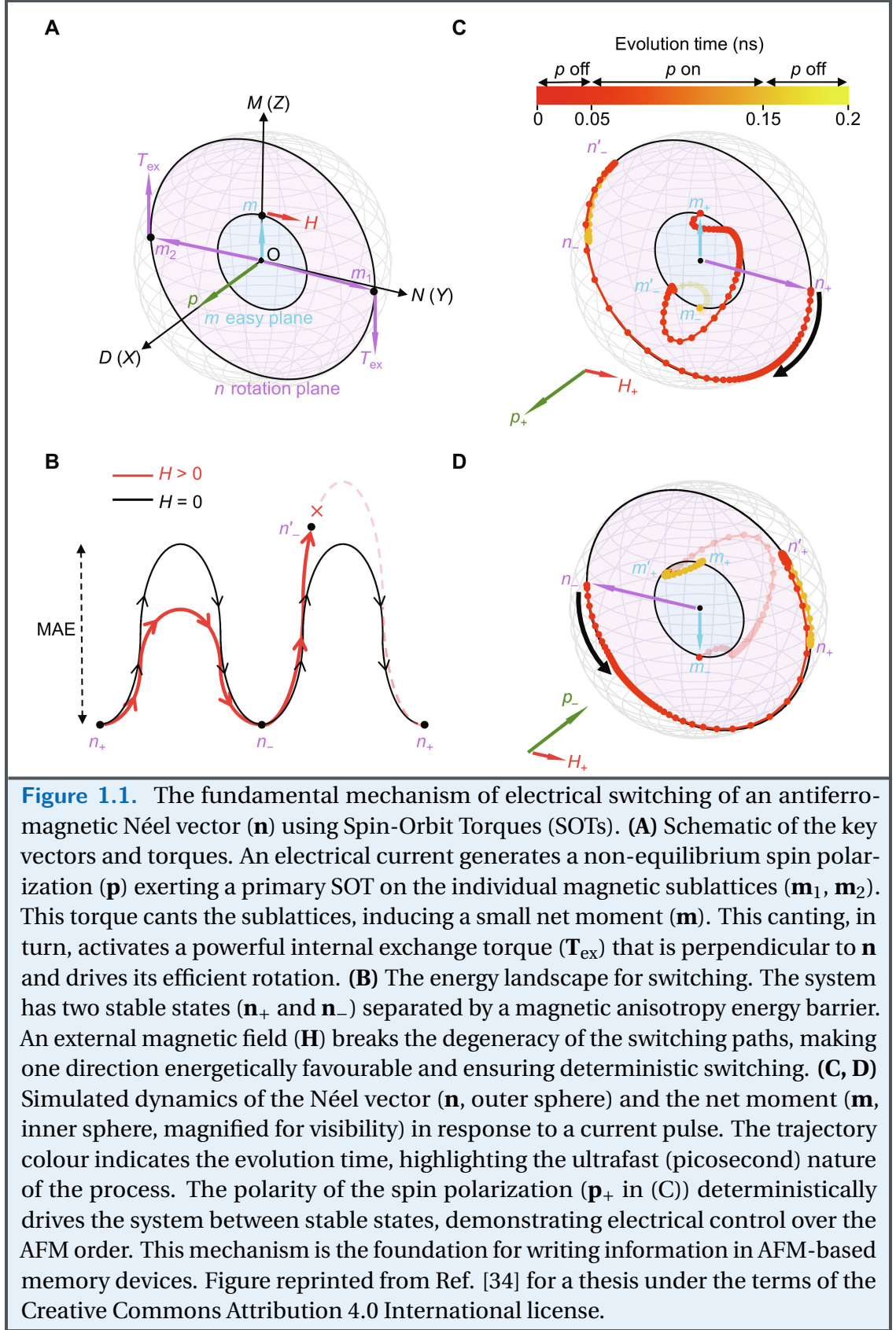
— 1.1 The Promise and Paradox of Antiferromagnetic Spintronics —

Antiferromagnetic materials have magnetic moments on neighbouring atoms aligned in an antiparallel fashion. This class of materials have long been regarded as the "sleeping giant" of magnetism. For decades, they were considered scientifically interesting but technologically inert due to the absence of a net magnetic moment, which made their magnetic

state difficult to manipulate or detect with conventional magnetic fields. However, a series of experimental and theoretical breakthroughs has awakened this giant, giving rise to the vibrant field of antiferromagnetic spintronics [8, 9]. The promise of AFM spintronics is rooted in the inherent advantages that antiferromagnets hold over their ferromagnetic counterparts. Firstly, their zero net magnetization means they do not generate stray magnetic fields, making them robust against external magnetic perturbations and allowing for ultra-dense integration of memory elements without cross-talk. Secondly, the exchange interactions that govern their magnetic order are typically much stronger than the magnetocrystalline anisotropy. Consequently, their intrinsic dynamics are orders of magnitude faster (in the terahertz range), compared to the gigahertz range of ferromagnets [29]. These properties position antiferromagnets as ideal candidates for future memory and logic devices that are simultaneously faster, smaller and more robust.

The central challenge has been developing methods for the electrical writing and reading of the antiferromagnetic order parameter, specifically the Néel vector. A key breakthrough occurred with the demonstration of Néel vector switching via spin-orbit torques (SOTs) in metallic antiferromagnets like CuMnAs and Mn₂Au [30, 31]. In these materials, an applied electrical current generates a non-equilibrium spin accumulation and spin current via the bulk or interfacial spin Hall effect. This, in turn, exerts a torque on the individual magnetic sublattices, enabling the coherent reorientation of the Néel vector. On the readout side, a variety of magnetoresistive effects have been exploited. The anisotropic magnetoresistance (AMR), where the resistance depends on the angle between the current and the Néel vector, provides a direct electrical signature of the antiferromagnetic state. More recently, the spin Hall magnetoresistance (SMR) has emerged as a powerful probe, sensitive to the Néel vector orientation at the interface between an AFM and a heavy metal layer [32, 33]. Even tunnelling magnetoresistance has been realized in junctions employing

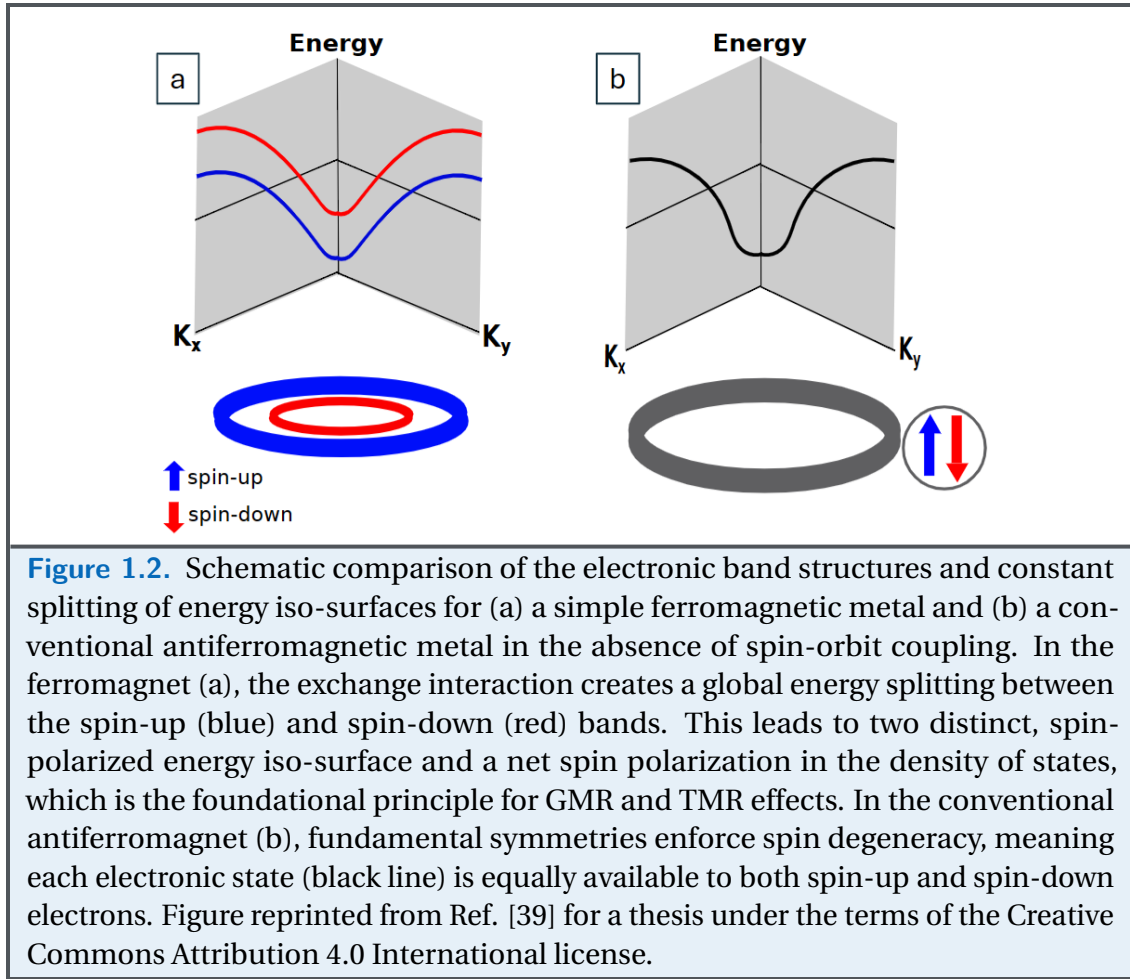
antiferromagnets as active components, demonstrating a spin-valve-like effect [11].



Despite these remarkable experimental advancements, a deep theoretical paradox remained at the heart of antiferromagnetic spintronics. The very phenomena that make

ferromagnets so useful for spintronics such as giant magnetoresistance are fundamentally tied to their spin-split electronic band structure. It was widely believed that the band structure of a conventional antiferromagnet must be spin-degenerate. This conclusion originates directly from their fundamental symmetries. In a simple antiferromagnet, the two opposite-spin sublattices are connected by a symmetry operation that includes time-reversal (θ) combined with a spatial operation, such as a lattice translation (T_τ) or inversion (T_i). This combined symmetry (e.g., $T_\tau\theta$) acts as an effective time-reversal operator for a given spin channel. According to Kramers' theorem, this guarantees that for every state with momentum $\mathbf{k}$ and energy E , there must be another state with the same momentum and energy but opposite spin. Therefore, in the absence of relativistic spin-orbit coupling (SOC), the bands must be fully spin-degenerate: $\epsilon_\uparrow(\mathbf{k}) = \epsilon_\downarrow(\mathbf{k})$, $\uparrow$ and $\downarrow$ denote spin-up and spin-down states, respectively. This spin degeneracy has profound consequences: it implies that a conventional antiferromagnet, in the absence of relativistic effects, should not be able to produce or sustain a spin-polarized current. Any spintronic phenomena would therefore have to rely on the smaller, relativistic SOC as their sole mechanism. Consequently, this created a significant tension within the field, as a growing body of experimental work demonstrated that antiferromagnets could be remarkably active spintronic components. For instance, experiments showed that pure spin currents could be efficiently transmitted across several micrometers in antiferromagnetic insulators like hematite ($\alpha\text{-Fe}_2\text{O}_3$) via the spin Seebeck effect [35]. Similarly, spin pumping experiments demonstrated that antiferromagnetic insulators such as NiO and MnF₂ could effectively conduct spin angular momentum pumped from an adjacent ferromagnet [36, 37]. Furthermore, the orientation of the Néel vector could be detected electrically through spin Hall magnetoresistance (SMR) in heterostructures like Pt/NiO, providing a direct electrical readout mechanism for the antiferromagnetic state [38]. These results presented a paradox. While the prevailing explanation attributed these effects to SOC, the high efficiency and long-distance transport observed in some of these experiments seemed disproportionately large for a purely relativistic phenomenon. This created an explanatory gap, suggesting that the foundational theoretical model of a fully spin-degenerate antiferromagnet, while correct in its symmetry arguments, might be an incomplete picture for describing all

antiferromagnetic materials.



The traditional framework for describing these symmetries is the theory of Magnetic Space Groups (MSGs) or Shubnikov groups. This powerful but inherently relativistic formalism rigidly couples spin and spatial operations. The culmination of this approach has been the development of the Spin-Splitting Type (SST) classification [15, 16]. The SST classes framework systematically categorised all 1651 MSGs based on their symmetry-allowed spin-splitting characteristics. It successfully identifies nonetheless a key class of materials, designated SST-4, which includes compensated magnets where a momentum-dependent spin splitting is allowed. However, because the MSG framework inextricably links spin, orbital, and lattice degrees of freedom, it could not cleanly separate the potentially large spin splitting arising from the non-relativistic crystal field from the smaller splitting induced by spin-orbit coupling. This limitation highlights the need for a more fundamental, non-relativistic theory to fully resolve the paradox and explain the full potential of compensated magnets in spintronics.

1.2 A New Foundation: The Theory of Spin Symmetry Groups

To resolve these limitations of the Magnetic Space Group (MSG) framework, a more specialised symmetry theory is required. Namely, one capable of describing magnetic crystals in a purely non-relativistic limit. A natural description comes with the realization that the rigid coupling of spin and spatial operations, inherent to the MSG formalism, could be lifted. This has led to the development of a novel theory of spin symmetry groups, a framework that treats the symmetries of the crystal lattice and the spin space as independent entities [14]. This decoupling provides the necessary theoretical freedom to specify and distinguish between different types of compensated magnetic order and unlocks new possibilities of their electronic properties. The central innovation of this new framework is the departure from a single, coupled symmetry operator to a composite operator formed from the direct product of the crystal space group, G_c , and the spin space group, G_s . The full symmetry group of the system is thus given by $G = G_c \times G_s$. An element $g \in G$ is a pair (g_c, g_s) , often written in the notation $[g_s || g_c]$, where $g_c \in G_c$ acts only on the spatial and orbital degrees of freedom, and $g_s \in G_s$ acts only on the spin degree of freedom.

The power of this formalism lies in its ability to describe the symmetries of a magnetic crystal in a non-relativistic limit. For any collinear magnet, there exists a non-trivial spin-only group, r_s , which contains symmetries that act only on the spin degrees of freedom. This group is given by $r_s = C_\infty + \bar{C}_2 C_\infty$, and it has two profound and universal consequences for the electronic band structure, $\epsilon(s, \mathbf{k})$:

1. The continuous rotation symmetry C_∞ about the collinear spin axis implies that the spin projection along this axis is conserved. Therefore, spin remains a good quantum number, and the electronic structure separates into non-mixing spin-up and spin-down channels.
2. The symmetry $\bar{C}_2$ corresponds to a 180° rotation of the spins about an axis perpendicular to the collinear axis. In the spin-group formalism, this operation is accompanied by the action of time-reversal, T , in the real (momentum) space. The resulting symmetry $[\bar{C}_2 || T]$ must leave the band structure invariant. Since $[\bar{C}_2 || T]\epsilon(s, \mathbf{k}) = \epsilon(s, -\mathbf{k})$,

it follows that for all collinear magnets:

$$\epsilon(s, \mathbf{k}) = \epsilon(s, -\mathbf{k}) \quad (1)$$

This is a remarkable result: the electronic bands of all non-relativistic collinear magnets are symmetric with respect to momentum-space inversion, regardless of whether the crystal lattice itself possesses inversion symmetry.

While these properties are universal, the distinction between the fundamental magnetic phases arises from the non-trivial spin groups. These are constructed by combining specific spin-space transformations with the crystallographic symmetries of the real space. For the purpose of classifying the spin-momentum locking of the band structure, it is sufficient to consider the 11 crystallographic Laue groups (the point groups that include inversion) for the real-space part. The resulting constructed groups are known as the non-trivial spin Laue groups and their structure dictates the magnetic phase of the material.

— 1.3 Re-classification: The Three Non-Relativistic Magnetic Phases —

The spin-group formalism reveals that there are precisely three distinct types of nontrivial spin Laue groups for collinear magnets. These are constructed by combining a group of spin-space transformations with one of the 11 crystallographic Laue groups, G , using the method of isomorphic coset decompositions. The resulting classification depends critically on the nature of the symmetry operation that connects the two opposite-spin sublattices. This leads to a rigorous re-classification of collinear magnetism into three fundamental phases, as illustrated schematically in Fig. 1.3.

- **Type I: Ferromagnetism.** The first type of nontrivial spin Laue group is given by:

$$\mathcal{R}_s^I = [E||G] \quad (2)$$

Here, E is the identity in spin space. In this phase, there is no symmetry operation that connects the opposite-spin sublattices; they are independent entities or there is only one spin lattice. Consequently, the group structure imposes no condition that

would enforce spin degeneracy at any point in the Brillouin zone. The bands are globally spin-split, $\epsilon_{\uparrow}(\mathbf{k}) \neq \epsilon_{\downarrow}(\mathbf{k})$, resulting in a net magnetization. This type describes conventional ferromagnets and ferrimagnets. There are 11 such groups, one for each crystallographic Laue group G .

- **Type II: Conventional Antiferromagnetism.** The second type of spin Laue group includes an additional generator involving the spin rotation $\bar{C}_2$:

$$\mathcal{R}_s^{\text{II}} = [E||G] + [C_2||G] \quad (3)$$

The key feature here is the presence of a symmetry operation of the form $[C_2||G]$ that connects opposite-spin sublattices, where the real-space operation $R \in G$ must be a **translation** or **inversion**. Since these operations leave the momentum vector $\mathbf{k}$ invariant (or map it to an equivalent point via a reciprocal lattice vector), the symmetry requirement $[C_2||R]\epsilon(s, \mathbf{k}) = \epsilon(-s, \mathbf{k}) = \epsilon(s, \mathbf{k})$ must hold. This enforces spin degeneracy for all $\mathbf{k}$ -vectors in the Brillouin zone. This provides the rigorous, non-relativistic definition of a conventional antiferromagnet, whose bands are spin-degenerate and time-reversal symmetric (in the absence of spin-orbit coupling). There are also 11 such groups.

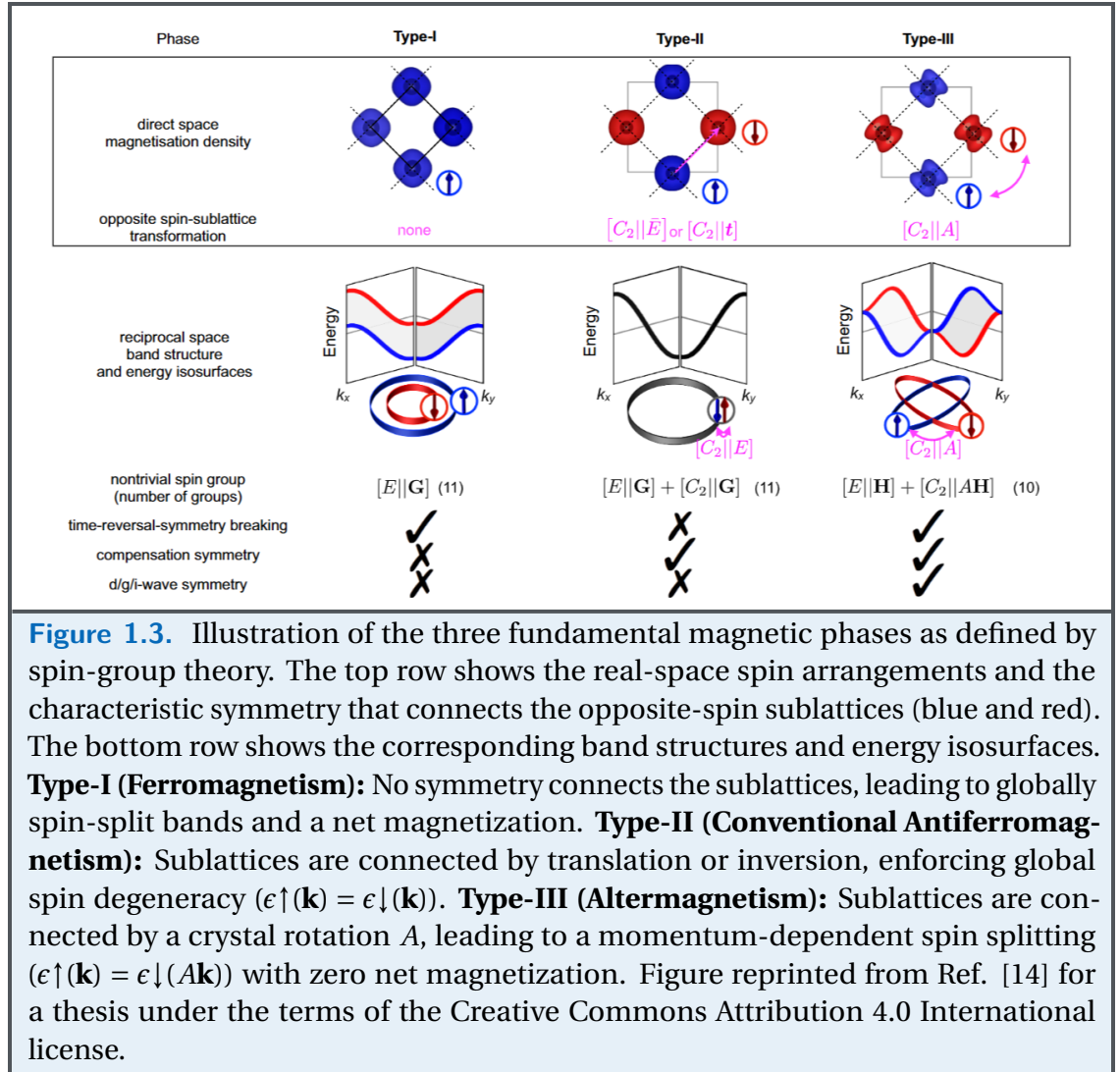
- **Type III: Altermagnetism.** The third and final type of spin Laue group has a more complex structure, based on a sublattice coset decomposition of the Laue group G . The group G is decomposed as $G = H + (G - H)$, where H is a halving subgroup containing all real-space transformations that map atoms to the *same* sublattice, and $(G - H)$ is its coset, containing all transformations that map atoms to the *opposite* sublattice. The nontrivial spin Laue group is then given by:

$$\mathcal{R}_s^{\text{III}} = [E||H] + [C_2||(G - H)] \quad (4)$$

The crucial feature of this phase is that the operations in the coset $(G - H)$ must be real-space **rotations**, and not translations or inversions. Let A be a representative rotation from this coset. The symmetry operation connecting opposite spins is therefore $[C_2||AH]$. Its action on the band structure implies the relation:

$$\epsilon(s, \mathbf{k}) = \epsilon(-s, A\mathbf{k}), \quad A\mathbf{k} \neq \mathbf{k} \quad (5)$$

Since A can only be a real-space proper or improper rotation, in general $AH\mathbf{k} \neq \mathbf{k}$. This means that spin degeneracy is lifted, $\epsilon(s, \mathbf{k}) \neq \epsilon(-s, \mathbf{k})$. However, unlike in a ferromagnet, the spin splitting is not global; it has a specific momentum dependence dictated by the rotation A . Furthermore, because the spin-up and spin-down states are related by a symmetry transformation, their energy isosurfaces must be equally populated, guaranteeing zero net magnetization. This combination of a momentum-dependent spin splitting with a compensated magnetic order is the unique signature of altermagnetism.



1.4 Beyond Collinear Order: The Emergence of Anti-Altermagnetism

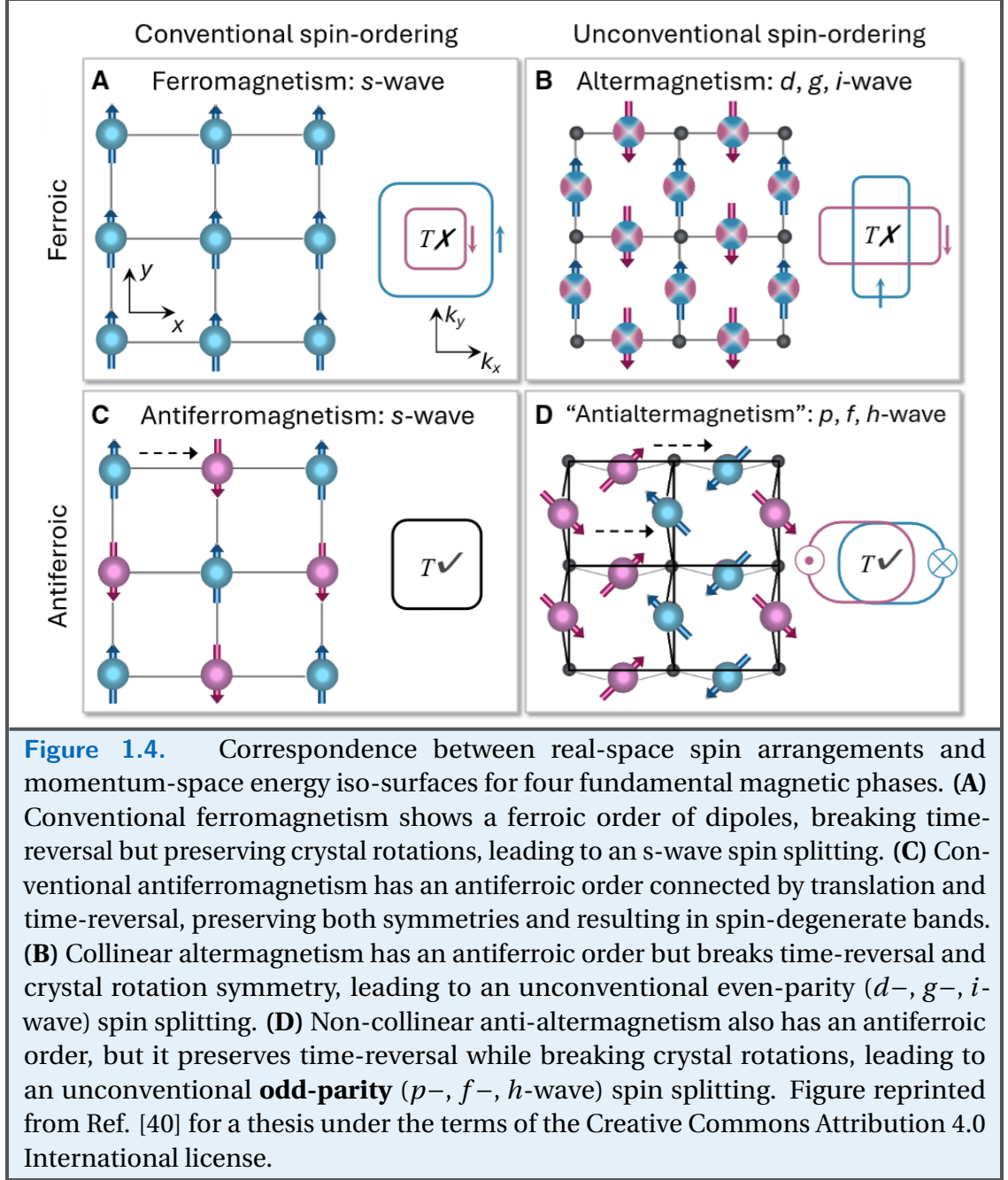
The spin group theory described in the preceding Sections 1.2 and 1.3 provides a complete and exhaustive classification for all non-relativistic collinear magnetic phases. However,

the rich landscape of magnetism also includes non-collinear spin arrangements, which can arise from phenomena such as frustrated exchange interactions or the Dzyaloshinskii-Moriya interaction. Very recently, a new class of unconventional magnetism has been identified within these non-collinear systems, which shares intriguing analogies with altermagnetism but is fundamentally distinct in its symmetry. Termed anti-altermagnetism, this phase applies to specific non-collinear, compensated spin structures that exhibit their own unique form of momentum-dependent spin splitting [40]. To understand this new phase, it is crucial to examine its defining symmetries and contrast them with those of altermagnetism and conventional antiferromagnetism as illustrated schematically in Fig. 1.4.

- **Real-Space Spin Structure:** Unlike the three collinear phases, anti-altermagnets have a non-collinear coplanar spin texture. Despite this, they maintain a compensated (antiferroic) order of local atomic dipoles, resulting in zero net magnetization.
- **Fundamental Symmetries:** The defining symmetry of an anti-altermagnet is the combination of time-reversal and a spatial operation such as translation. This is the same symmetry that defines a conventional Type-II antiferromagnet and which is explicitly broken in a Type-III altermagnet. Consequently, anti-altermagnets, like conventional antiferromagnets, are time-reversal symmetric. However, they simultaneously break spatial inversion (parity) symmetry.
- **Momentum-Space Consequences:** This unique combination of symmetries (preserving time-reversal while breaking parity) has profound consequences for the band structure. The overall time-reversal symmetry of the system leads to an electronic structure that is also time-reversal invariant. However, the broken parity and non-collinear nature of the magnetism allow for a momentum-dependent spin splitting. This splitting has an odd-parity character in momentum space, corresponding to p -, f -, or h -wave symmetries. This stands in stark contrast to the even-parity (d -, g -, or i -wave) spin splitting that characterizes altermagnetism.

In essence, anti-altermagnetism represents a parallel path to achieving unconventional spin textures in momentum space. While altermagnetism achieves this via a non-relativistic, time-reversal-breaking mechanism in collinear systems, anti-altermagnetism does so via a time-reversal-preserving, parity-breaking mechanism in non-collinear systems. The discovery of this phase further expands the landscape of magnetism, demonstrating that

the interplay of crystal symmetry and spin arrangement can generate a diverse family of materials with novel spintronic potential. While a full exploration of anti-altermagnetism is beyond the scope of this thesis, its existence underscores the power of the symmetry-based approach. The remainder of this work will focus on the unique physical phenomena arising from the even-parity, time-reversal-breaking Type-III altermagnetic phase.



A crucial question arises when bridging the idealized, non-relativistic theory of altermagnetism with real materials: how does the framework accommodate effects like the Dzyaloshinskii-Moriya interaction (DMI), which is driven by spin-orbit coupling and inherently induces non-collinearity? The answer lies in recognizing the hierarchy of en-

ergy scales and understanding that the spin-group classification describes the dominant, underlying symmetry of the non-relativistic Hamiltonian, upon which relativistic effects act as a perturbation. The altermagnetic phase is defined in a non-relativistic limit for perfectly collinear spin structures. The exchange interaction and crystal field potential, which together establish this primary order, are typically the largest energy scales in the system, often on the order of electron-volts (eV). The spin-splitting they produce is a direct consequence of these strong, non-relativistic forces. In contrast, the Dzyaloshinskii-Moriya interaction is a relativistic effect scaling with the strength of the spin-orbit coupling. It arises in crystals lacking an inversion centre between the interacting magnetic ions and manifests as an energy term of the form $E_{DMI} \propto \mathbf{D} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$, where $\mathbf{D}$ is the DMI vector, $\mathbf{S}_{i,j}$ the spin operator at position i, j . This interaction favours a canted, or non-collinear, arrangement of spins, and its energy scale is typically orders of magnitude smaller (on the order of milli-electron-volts, meV) than the primary exchange interactions [9]. Therefore, the DMI does not compete with the formation of the altermagnetic state; rather, it acts upon the already established altermagnetic order. The situation is analogous to the well-known case of canted antiferromagnetism in materials like hematite ($\alpha\text{-Fe}_2\text{O}_3$). The primary, dominant order is antiferromagnetic. The weaker DMI then introduces a slight canting of the sublattices, resulting in a weak ferromagnetic moment. We do not cease to call the material an antiferromagnet; we recognize it as a "canted antiferromagnet" where the underlying AFM physics still governs its properties [8]. The same logic applies directly to altermagnetism. If a material's symmetry allows for DMI, the interaction will induce a small canting of the spins away from perfect collinearity. The material becomes a "canted altermagnet." However, as long as the primary non-relativistic spin splitting ($\sim$ eV) is substantially larger than the energy scales of the SOC-driven perturbations ($\sim$ meV), the defining characteristics of altermagnetism will persist:

- The band structure will still be dominated by a large, even-parity, momentum-dependent spin splitting.
- The unique transport phenomena, such as the crystal anomalous Hall effect and spin-splitter torques, will remain the primary response functions, albeit slightly modified by the relativistic corrections.

In summary, the spin-group classification for altermagnetism refers to the symmetry of the dominant, non-relativistic part of the Hamiltonian. The presence of SOC-driven

DMI and the resulting non-collinearity are real-world perturbations that can coexist with the altermagnetic phase. They modify the ideal state but do not destroy it provided the hierarchy of energy scales is respected. Thus, a material can be correctly classified as an altermagnet based on its non-relativistic properties even while exhibiting secondary effects like weak ferromagnetism due to DMI-induced canting [14].

— 1.5 Signatures and Consequences of the Altermagnetic Phase —

The unique symmetry of altermagnets where opposite-spin sublattices are connected by a crystal rotation, gives rise to a host of remarkable physical phenomena that are forbidden in conventional antiferromagnets. These signatures are not subtle, relativistic corrections; they are large, non-relativistic effects that fundamentally alter the electronic, optical, and transport properties of the material. This section details these profound consequences, which serve as the primary experimental fingerprints for identifying and exploiting the altermagnetic phase.

1.5.1 Momentum-Dependent Spin Splitting

The primary and most fundamental consequence of altermagnetic symmetry is the emergence of a large, momentum-dependent spin splitting of the electronic bands, as dictated by Eq. (5). Unlike the global spin splitting in ferromagnets driven by the net exchange field, or the typically smaller splitting in non-centrosymmetric materials driven by spin-orbit coupling, the splitting in altermagnets is a non-relativistic effect rooted in the crystal field potential [14]. This mechanism can generate energy splitting on the order of hundreds of meV and, in some cases, exceeding 1 eV [17, 18]. This large magnitude is a direct consequence of its non-relativistic origin, making it comparable to the exchange splitting in common ferromagnets. To place these magnitudes in context: the thermal energy at room temperature is $k_B T \approx 25$ meV at $T = 300$ K, meaning the altermagnetic spin-valley splitting typically exceeds the thermal scale by a factor of 10–40. This hierarchy is crucial for practical applications: it ensures that the spin-polarized valleys remain robust and well-defined at operating temperature, and that spin-dependent transport effects are not washed out by thermal fluctuations. The energy scale of the spin splitting is also large compared to the bias voltages required for device operation; for instance, the TMR inversion

voltage predicted for the RuO_2 / TiO_2 / RuO_2 system ($V_{inv} \approx 0.35 \text{ V} \equiv 350 \text{ meV}$, Chapter 4) falls well within the spin-valley gap, confirming that the underlying altermagnetic order is not destabilized by the applied electric field. This giant spin splitting was first identified through ab-initio calculations in materials like RuO_2 and MnTe [15, 41], and has since been predicted in a wide range of compounds [42], see Chapters A and B. The key feature is that the sign of the spin splitting, $\Delta\epsilon(\mathbf{k}) = \epsilon_{\uparrow}(\mathbf{k}) - \epsilon_{\downarrow}(\mathbf{k})$, alternates in momentum space according to the rotational symmetry A . This alternating nature ensures that while the bands are locally spin-polarized, the total density of states remains spin-degenerate, thus preserving the zero net magnetization of the compensated magnetic state. This fundamental difference between the three magnetic phases is illustrated schematically in Fig. 1.3.

1.5.2 Alternating Spin Polarization in Real and Momentum Space

The momentum-space spin splitting has a direct and visually intuitive counterpart in both the real-space crystal structure and the momentum-space Fermi surface. In real space, it corresponds to an alternating spin polarization of the local electronic density around the magnetic atoms. This is distinct from a simple antiparallel alignment of atomic moments; it reflects the anisotropic shape of the local spin density itself, which is rotated from one magnetic sublattice to the next [14] due to the presence of non-magnetic atoms. In momentum space, the spin splitting transforms the Fermi surface into a rich texture of alternating spin polarization. Regions of the Brillouin zone, often referred to as "spin-momentum-locked valleys", become highly spin-polarized. For a given energy, the Fermi surface is composed of distinct sheets, each with a definite spin character, ensuring that the total volume enclosed by the spin-up and spin-down Fermi surfaces is identical. This alternating spin texture in the Brillouin zone is the key to understanding the unique transport properties of altermagnets. This direct link between the real-space rotational symmetry and the resulting momentum-space spin texture for an archetypal altermagnet is shown in Fig. 1.4.

1.5.3 Anisotropic Magneto-transport and Time-Reversal Symmetry Breaking

The breaking of time-reversal symmetry by the spin-split bands, even with zero net magnetization, allows for a range of magneto-transport phenomena previously thought to be exclusive to ferromagnets. The most prominent of these is the Anomalous Hall Effect typical for ferromagnets where a transverse voltage is generated in response to a longitudinal current. In altermagnets, the AHE arises from the non-zero Berry curvature integrated over

the occupied spin-split bands [43]. This "crystal Hall effect" was predicted theoretically and subsequently observed experimentally in RuO₂ thin films, providing one of the first key experimental proofs of its altermagnetic nature [19]. Beyond the AHE, the unique spin textures enable novel spin-current phenomena and spin-orbit torques (SOTs). It has been shown that passing a current through an altermagnet can generate unconventional spin currents, including out-of-plane polarized spins, which are highly efficient for switching adjacent ferromagnetic layers [17]. This "spin-splitter torque" has been experimentally confirmed in RuO₂ demonstrating its potential for creating field-free switching mechanisms in SOT-MRAM devices [20, 21, 44]. Furthermore, the altermagnetic spin splitting is predicted to give rise to giant and tunnelling magnetoresistance effects in suitably designed heterostructures [45], a topic that forms the core motivation of this thesis.

1.5.4 "Wave" Symmetry of the Spin Splitting

The specific momentum-space pattern of the spin splitting is determined by the rotational symmetry A that connects the opposite-spin sublattices. In an analogy to the theory of unconventional superconductivity, this pattern is often described by a "wave" symmetry nomenclature (e.g., d-wave, g-wave, i-wave) that reflects its angular momentum character [14].

- ***d*-wave:** Arises from a four-fold rotational symmetry ($A = C_{4z}$). The spin splitting function, $\Delta\epsilon(\mathbf{k})$, has four nodal planes and changes sign four times upon a rotation in a plane perpendicular to the rotation axis, analogous to a d_{xy} or $d_{x^2-y^2}$ orbital. RuO₂ is the canonical example of a *d*-wave altermagnet [12, 41].
- ***g*-wave:** Arises from a six-fold rotational symmetry ($A = C_{6z}$). The splitting function has six sign changes, leading to a more complex, star-shaped spin texture in the Brillouin zone. Materials like MnTe and CrSb are predicted to be *g*-wave altermagnets [12].
- ***i*-wave:** Arises from higher-order symmetries, leading to even more complex spin textures with eight or more sign changes.

This classification is crucial as it directly determines which k-space "valleys" are spin-polarized and thus dictates the symmetry of any resulting transport or optical phenomena, providing a predictive link between crystal structure and device functionality.

1.6 Theoretical Limitations and Open Questions

While the theory of spin symmetry groups provides a powerful and elegant framework for deriving and understanding altermagnetism, it is essential to recognize its current limitations and the open questions it brings to the forefront of condensed matter physics. These challenges do not diminish the theory's impact; rather, they define the active frontiers of research and highlight the areas where further theoretical and experimental work is most needed. This section provides a critical overview of these theoretical limitations.

1.6.1 The Challenge of a Unique Order Parameter

A significant theoretical challenge for altermagnetism is the absence of a simple, unique order parameter that distinguishes it from conventional antiferromagnetism. In the ferromagnetic case, the magnetization, $\mathbf{M}$, defines a ferromagnet. Both conventional antiferromagnets and altermagnets are described by a Néel vector, $\mathbf{N}$ (or staggered magnetization), which specifies the antiparallel orientation of the magnetic sublattices. However, the Néel vector itself is blind to the defining characteristic of altermagnetism: the crystal symmetry that connects the opposite-spin sublattices. For instance, two materials could share an identical G -type antiferromagnetic structure, described by the same Néel vector. Yet, one could be a conventional antiferromagnet with spin-degenerate bands (like NiO), while the other is an altermagnet with massive spin splitting (like RuO₂). The Néel vector, $\mathbf{N}$, fails to capture this fundamental distinction. While the full magnetic or spin space group provides a complete symmetry description, a more practical, vector-like order parameter that encapsulates the "rotational" nature of the magnetic order is currently lacking. This complicates the development of phenomenological models, such as Landau theory for altermagnetic phase transitions, and makes experimental interpretation more nuanced than for ferromagnets.

1.6.2 The Non-Relativistic Idealization versus Real Materials

The power of the spin-group theory lies in its non-relativistic formulation, which successfully isolates the crystal-field-driven spin splitting from relativistic effects. However, all real materials possess some degree of spin-orbit coupling, an inherently relativistic phenomenon. The presence of spin-orbit coupling complicates the ideal altermagnetic

picture in several ways [14]:

- **Spin Canting and Weak Ferromagnetism:** In certain crystal symmetries, spin-orbit coupling can permit a Dzyaloshinskii-Moriya interaction (DMI), which can cause a slight canting of the sublattice spins. This results in a small net magnetization (weak ferromagnetism), technically breaking the "perfectly compensated" condition of the ideal altermagnetic state.
- **Mixing of Spin Channels:** In the pure non-relativistic limit, the spin-up and spin-down channels are perfectly decoupled. Spin-orbit coupling mixes these states, meaning spin projection along a single axis is no longer a perfectly conserved quantum number. This can reduce the degree of spin polarization in the k-space valleys.
- **Superposition of Splitting Mechanisms:** The final electronic band structure in a real material is a superposition of the large, even-parity altermagnetic splitting and the typically smaller, odd-parity spin-orbit coupling-induced splitting (e.g., Rashba or Dresselhaus effects). While the altermagnetic component is expected to dominate in materials with light elements, experimentally and theoretically disentangling these two contributions remains a key challenge.

Thus, the "pure" altermagnetic phase is an idealization. Real materials will always exhibit a hybrid of non-relativistic and relativistic phenomena, and understanding this interplay is crucial for accurate material characterization and device modelling.

1.6.3 Microscopic Origins and Strong Correlation

The spin-group framework is a symmetry-based theory. It is exceptionally powerful at classifying the possible ground states and describing their electronic properties. However, it does not by itself explain the microscopic origin of the magnetic ordering, especially in systems where strong electron-electron correlations are important. The theory does not intrinsically predict why a material should choose an altermagnetic ground state over a conventional antiferromagnetic or non-magnetic one. For instance, in the case of RuO_2 , one prominent theory suggests that the magnetic order arises from a Fermi surface instability known as a "d-wave Pomeranchuk instability," a mechanism driven by strong electron-electron interactions [41]. The spin-group formalism can classify the resulting state as altermagnetic, but it does not provide the tools to model the interaction-driven instability itself. The predictive power of the theory is therefore for the properties of a

given phase, not necessarily for the energetic stability of that phase, which often requires many-body theoretical approaches.

1.6.4 Collective Excitations: The Quest for the Altermagnon

The initial focus of altermagnetism theory has been on the static electronic ground state (i.e., the band structure). The corresponding collective excitations, such as magnons, are much less understood. In conventional magnets, magnon modes are well-defined: ferromagnets have one circularly polarized mode, while antiferromagnets typically have two degenerate, linearly polarized modes at zero magnetic field. The unique spin-split band structure of altermagnets must lead to equally unique magnon dispersions. It is expected that the magnon branches in an altermagnet will also exhibit a momentum-dependent splitting, lifting the degeneracy found in conventional antiferromagnets. This would have profound implications for THz spintronics, calorimetry, and other dynamic experiments. While this is an active area of research, with recent theoretical work beginning to explore the properties of these "altermagnons" [46], a complete and experimentally verified theory of altermagnetic spin dynamics is still in its early stages.

1.6.5 The Question of Generality and Material Realizations

While spin-group theory predicts ten possible non-trivial spin Laue groups for altermagnetism, suggesting the phase is abundant in principle [13], the number of experimentally verified, "clean" examples remains small. The very controversy surrounding RuO_2 , the canonical example, highlights the profound experimental challenge. As recent studies suggest, the stability of the altermagnetic phase can be extremely sensitive to stoichiometry, strain, defects, and quantum fluctuations potentially making it a fragile state that exists only under very specific conditions [47, 48]. The true "generality" of the phase in real-world materials is therefore still an open experimental question that requires careful, multifaceted verification for each new candidate material.

— 1.7 Conclusion and Bridge to Device Physics —

This chapter has outlined a path from the foundational open questions of conventional antiferromagnetism to the formal derivation and critical evaluation of a third fundamental

magnetic phase. By moving beyond the traditional, relativistic framework of Magnetic Space Groups and employing the more specialised theory of non-relativistic spin symmetry groups, we have established a rigorous foundation for altermagnetism. The central discovery is that altermagnetism is not merely an intermediate case, but a distinct phase of collinear magnetism, as fundamental as ferromagnetism and conventional antiferromagnetism. It describes a remarkable class of materials that combines the most desirable properties of its counterparts: the zero net magnetization, robustness against external fields, and ultrafast THz dynamics of antiferromagnets are merged with the large, non-relativistic spin splitting of the electronic bands characteristic of ferromagnets.

At the same time, we have critically examined the frontiers of this emerging field, acknowledging the present challenges that define its research landscape. The absence of a simple, unique order parameter beyond the Néel vector, the idealization of the non-relativistic limit in the face of real-world spin-orbit coupling, and the open questions surrounding material stability and collective "altermagnon" dynamics are all crucial areas of ongoing investigation. However, these limitations do not diminish the profound potential implications of altermagnetism for spintronics; rather, they underscore the urgent need for robust theoretical benchmarks. The promise of a large, non-relativistic spin splitting provides a powerful new mechanism for generating and manipulating spin currents without a net magnetic moment, an ideal combination for device applications. This property is precisely the ingredient required to engineer novel spintronic devices with superior performance. It is this potential that forms the central motivation for the remainder of this thesis. The giant, momentum-dependent spin splitting, whose origins were detailed in this chapter, is the core theoretical mechanism expected to generate giant tunnelling magnetoresistance effects. The symmetry principles outlined here are not merely an academic classification. They are the predictive tools that will guide our search for new materials and provide the physical intuition for understanding the quantum transport phenomena at the heart of this work, even as the broader field continues to evolve and address these foundational questions.

2 Computational Methodology

The preceding chapter introduced and described altermagnetism as a potential third fundamental magnetic phase, distinct from both ferromagnetism and conventional antiferromagnetism. We have seen that this discovery, rooted in a more fundamental understanding of spin symmetry, presents a remarkable opportunity for spintronics. It offers a class of materials that merges the most desirable traits of its counterparts: the large, non-relativistic spin splitting of ferromagnets with the zero net magnetization and ultrafast dynamics of antiferromagnets. This unique combination provides, in principle, an ideal platform for engineering novel spintronic devices with superior performance. However, we also acknowledged the frontiers of this emerging field. The ideal non-relativistic picture is complicated by real-world spin-orbit coupling, and the very stability of the altermagnetic phase in candidate materials like RuO_2 is the subject of intense scientific debate. These challenges do not diminish the potential of altermagnetism; rather, they highlight an urgent need for robust, predictive, and quantitative theoretical benchmarks. To bridge the gap between theoretical promise and practical realization, we must turn to first-principles computational modelling. It is the essential tool required to investigate the subtle balance of energies that determines a material's magnetic ground state and to predict the performance of a device before it is built.

This chapter details the first-principles computational framework that forms the bedrock of this thesis. The most important goal is to justify our choice of methods by linking them directly to the physical problems at hand. We will begin with the fundamentals of Density Functional Theory (DFT), the workhorse of modern materials science. Then,

we address its limitations and introduce the essential corrections, namely the DFT+ U and Atomic Self-Interaction Correction (ASIC) methods. These methods are critical for accurately describing the electron correlation effects hypothesized to drive magnetism in Ruthenium dioxide RuO_2 (Chapter 3). Finally, to probe the device applications, we will outline the Non-Equilibrium Green's Function (NEGF) formalism, which when combined with our corrected DFT model, allows us to simulate quantum transport and provide the theoretical predictions for the performance of novel altermagnetic tunnel junctions design (Chapter 4).

— 2.1 Fundamentals of Density Functional Theory (DFT) —

The primary challenge in condensed matter theory is to solve the quantum mechanical equations governing the behaviour of interacting electrons and nuclei. For a system of N electrons, this is described by the many-body time-independent Schrödinger equation:

$$\hat{H}\Psi(\mathbf{r}_1, \dots, \mathbf{r}_N) = E\Psi(\mathbf{r}_1, \dots, \mathbf{r}_N) \quad (1)$$

where Ψ is the many-body electronic wavefunction and $\hat{H}$ is the Hamiltonian operator. Within the Born-Oppenheimer approximation [49], where the nuclei are treated as fixed classical particles, the electronic Hamiltonian can be written:

$$\hat{H} = \hat{T} + \hat{V}_{\text{ext}} + \hat{W} = -\sum_{i=1}^N \frac{\hbar^2}{2m_e} \nabla_i^2 + \sum_{i=1}^N V_{\text{ext}}(\mathbf{r}_i) + \sum_{i<j}^N \frac{e^2}{4\pi\epsilon_0|\mathbf{r}_i - \mathbf{r}_j|} \quad (2)$$

Here, $\hat{T}$ is the kinetic energy of the electrons, $\hat{V}_{\text{ext}}$ is the external potential created by the fixed atomic nuclei, and $\hat{W}$ represents the complex and computationally prohibitive electron-electron interaction term. The variables m_e and e are the electron mass and charge, respectively. Solving this equation directly is intractable for all but the simplest systems due to the high dimensionality and the coupling of all electronic coordinates in the interaction term.

Density Functional Theory provides an elegant and powerful alternative. Instead of the complex many-body wavefunction, DFT recasts the entire problem in terms of a much

simpler quantity: the electronic ground-state density, $\rho_0(\mathbf{r})$, which is a function of only three spatial coordinates.

$$\rho_0(\mathbf{r}) = N \int \dots \int |\Psi(\mathbf{r}, \mathbf{r}_2, \dots, \mathbf{r}_N)|^2 d\mathbf{r}_2 \dots d\mathbf{r}_N \quad (3)$$

The formal justification for this transformative simplification rests on two foundational theorems proven by Hohenberg and Kohn [50].

- **The First Hohenberg-Kohn Theorem** states that there exists a one-to-one correspondence between the external potential $V_{\text{ext}}(\mathbf{r})$ and the ground-state electron density $\rho(\mathbf{r})$. This is a profound statement: since V_{ext} defines the Hamiltonian, it implies that the ground-state density uniquely determines all properties of the system, including the total energy, which can therefore be written as a functional of the density: $E_0 = E[\rho_0]$.
- **The Second Hohenberg-Kohn Theorem** establishes a variational principle for the energy functional. It states that for any trial density $\rho(\mathbf{r})$ (such that $\rho(\mathbf{r}) \geq 0$ and $\int \rho(\mathbf{r}) d\mathbf{r} = N$), the energy calculated from the functional $E[\rho]$ will be greater than or equal to the true ground-state energy, E_0 . The minimum value is achieved only when the trial density is the true ground-state density, $\rho_0(\mathbf{r})$.

While the HK theorems provide an exact reformulation of the many-body problem, they do not prescribe the explicit form of the universal energy functional $E[\rho]$. The breakthrough of Kohn and Sham was to propose a practical method for obtaining the ground-state density [51]. Their approach replaces the intractable interacting system with a fictitious, auxiliary system of non-interacting electrons that, by construction, has the exact same ground-state density $\rho_0(\mathbf{r})$ as the real, interacting system. The wavefunctions of this auxiliary system, $\psi_i(\mathbf{r})$, are determined by a set of single-particle Schrödinger-like equations known as the Kohn-Sham (KS) equations:

$$\left[-\frac{\hbar^2}{2m_e} \nabla^2 + V_{\text{eff}}(\mathbf{r}) \right] \psi_i(\mathbf{r}) = \varepsilon_i \psi_i(\mathbf{r}) \quad (4)$$

where ε_i are the Kohn-Sham eigenvalues. The ground-state density is then easily con-

constructed from the N lowest-energy occupied Kohn-Sham orbitals:

$$\rho_0(\mathbf{r}) = \sum_{i=1}^N |\psi_i(\mathbf{r})|^2 \quad (5)$$

The key to this method and the single most important concept for this chapter lies in the definition of the effective potential, $V_{\text{eff}}(\mathbf{r})$, which is defined as:

$$V_{\text{eff}}(\mathbf{r}) = V_{\text{ext}}(\mathbf{r}) + V_{\text{H}}(\mathbf{r}) + V_{\text{XC}}(\mathbf{r}) \quad (6)$$

Here, $V_{\text{H}}(\mathbf{r})$ is the classical electrostatic Hartree potential, which describes the repulsion of an electron from the mean-field charge distribution of all other electrons:

$$V_{\text{H}}(\mathbf{r}) = \frac{e^2}{4\pi\epsilon_0} \int \frac{\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}' \quad (7)$$

All the complex many-body effects of quantum exchange and correlation are bundled into the final term, $V_{\text{XC}}(\mathbf{r})$, known as the exchange-correlation (XC) potential. It is formally defined as the functional derivative of the exchange-correlation energy, $E_{\text{XC}}[\rho]$, with respect to the density:

$$V_{\text{XC}}(\mathbf{r}) = \frac{\delta E_{\text{XC}}[\rho(\mathbf{r})]}{\delta \rho(\mathbf{r})} \quad (8)$$

The exchange-correlation energy $E_{\text{XC}}[\rho]$ contains the difference between the true kinetic energy of the interacting system and the kinetic energy of the non-interacting KS system, as well as all non-classical contributions to the electron-electron interaction. The exact form of $E_{\text{XC}}[\rho]$ is unknown, and it is here that approximations must be made. The accuracy of any DFT calculation rests entirely on the quality of the chosen approximation for this functional. This is the central reason why advanced correction schemes are often necessary. The most widely used families of approximations are:

- **Local Density Approximation (LDA):** This simplest approximation assumes the exchange-correlation energy density at a point $\mathbf{r}$ is the same as that of a uniform electron gas with a density equal to the local density $\rho(\mathbf{r})$. Despite its simplicity, LDA is often remarkably successful but tends to overbind molecules and underestimate band gaps. The XC energy functional is given by:

$$E_{\text{XC}}^{\text{LDA}}[\rho] = \int \rho(\mathbf{r}) \epsilon_{\text{XC}}^{\text{hom}}(\rho(\mathbf{r})) d^3\mathbf{r} \quad (9)$$

where $\epsilon_{\text{XC}}^{\text{hom}}(\rho)$ is the exchange-correlation energy per particle of a homogeneous electron gas of density ρ , a quantity that can be calculated accurately (e.g., via Quantum Monte Carlo) and parametrised [52].

- **Generalized Gradient Approximation (GGA):** These functionals improve upon LDA by considering not only the local density $\rho(\mathbf{r})$ but also the magnitude of its local gradient, $|\nabla\rho(\mathbf{r})|$. The functional takes the general form $E_{\text{XC}}^{\text{GGA}}[\rho, \nabla\rho]$. This allows the functional to better account for the inhomogeneity of the electron density in real materials, generally leading to more accurate geometries and energies. Several widely used forms exist, the foundational one being in [53, 54].

Since V_{eff} depends on the density that is constructed from its own orbital solutions, the KS equations must be solved iteratively in a self-consistent field (SCF) procedure.

2.2 Spin-Polarized Density Functional Theory

To describe magnetic materials, where the density of spin-up and spin-down electrons is imbalanced, the DFT formalism must be extended. This is achieved by treating the spin-up ($\uparrow$) and spin-down ($\downarrow$) densities as two distinct variables. This is the foundation of spin-DFT [55]. The scalar density $\rho(\mathbf{r})$ is replaced by two spin densities:

$$\rho_{\uparrow}(\mathbf{r}) \quad \text{and} \quad \rho_{\downarrow}(\mathbf{r}) \quad (10)$$

From these, one can define the total charge density, $\rho(\mathbf{r}) = \rho_{\uparrow}(\mathbf{r}) + \rho_{\downarrow}(\mathbf{r})$, and the scalar magnetization density, $m(\mathbf{r}) = \rho_{\uparrow}(\mathbf{r}) - \rho_{\downarrow}(\mathbf{r})$. The Hohenberg-Kohn theorems are generalized to state that the ground state is a unique functional of both $\rho(\mathbf{r})$ and $m(\mathbf{r})$ (or, equivalently, $\rho_{\uparrow}$ and $\rho_{\downarrow}$). This leads to a set of two coupled Kohn-Sham equations, one for each spin channel $\sigma \in \{\uparrow, \downarrow\}$:

$$\left[-\frac{\hbar^2}{2m_e} \nabla^2 + V_{\text{eff},\sigma}(\mathbf{r}) \right] \psi_{i,\sigma}(\mathbf{r}) = \epsilon_{i,\sigma} \psi_{i,\sigma}(\mathbf{r}) \quad (11)$$

The spin-dependent effective potential now includes a spin-dependent XC potential:

$$V_{\text{eff},\sigma}(\mathbf{r}) = V_{\text{ext}}(\mathbf{r}) + V_{\text{H}}(\mathbf{r}) + V_{\text{XC},\sigma}(\mathbf{r}) \quad (12)$$

where the XC potential for each spin is given by the functional derivative of the XC energy, which is now a functional of both spin densities:

$$V_{\text{XC},\sigma}(\mathbf{r}) = \frac{\delta E_{\text{XC}}[\rho_{\uparrow}, \rho_{\downarrow}]}{\delta \rho_{\sigma}(\mathbf{r})} \quad (13)$$

This formulation, known as the Local Spin-Density Approximation (LSDA) when combined with an LDA-type functional [55], is the simplest method to describe magnetic systems from first principles. It naturally allows for an energy difference between parallel (ferromagnetic) and antiparallel (antiferromagnetic) spin alignments, providing access to magnetic moments, exchange interactions, and spin-dependent band structures. For more complex non-collinear magnetic systems, the density and potential become 2×2 matrices in spin space [56]. However, for the collinear magnetic states investigated in this thesis, the spin-polarized formalism of Eq. (11) provides the necessary foundation. Since the effective potential V_{eff} depends on the density $\rho(\mathbf{r})$, which in turn depends on the Kohn-Sham orbitals ψ_i that are solutions of the KS equations shown in Eq. (4), the problem must be solved iteratively in a self-consistent field procedure.

2.3 Beyond Standard DFT: Treating Correlation and Self-Interaction

While the Kohn-Sham formalism is formally exact, its practical application relies on approximations for the exchange-correlation functional, E_{XC} . Standard approximations like LDA and GGA, which are based on the model of a slowly-varying electron gas, have proven remarkably successful for a wide range of materials, particularly simple metals and semiconductors. However, they exhibit systematic failures in systems dominated by strong electron-electron interactions, very commonly referred to as "strongly correlated" materials. The central issue arises in materials containing atoms with localized, partially-filled d - or f - orbitals, such as transition metal oxides. In these systems, the strong on-site Coulomb repulsion between electrons in these localized shells is a dominant physical effect. LDA and GGA intrinsically tend to overly delocalize the electron density because of spurious self-interaction of the independent electrons, failing to capture this strong localization. This deficiency leads to several well-known failures:

- A severe underestimation of the band gap, famously predicting many Mott insulators

(materials that are insulating due to strong correlation) to be metallic.

- An incorrect description of the magnetic ground state or a significant underestimation of the local magnetic moments.
- An inability to describe correlation-driven metal-insulator transitions.

Since the magnetism in RuO₂ is hypothesized to be an itinerant phenomenon driven by correlation effects within the Ru 4*d* shell, it is imperative to employ methods that go beyond the standard LDA/GGA framework.

2.3.1 The DFT+*U* Method: A Correction for On-Site Correlation

The DFT+*U* method is a pragmatic and widely-used approach to correct for the deficiencies of standard functionals in strongly correlated systems [57]. The core idea is to augment the standard DFT functional with a term borrowed from the Hubbard model, which explicitly accounts for the strong on-site Coulomb repulsion of localized electrons. The total energy functional in the DFT+*U* approach is written as:

$$E_{\text{DFT}+U}[\rho, \{\hat{n}^I\}] = E_{\text{DFT}}[\rho] + E_{\text{Hub}}[\{\hat{n}^I\}] - E_{\text{dc}}[\{\hat{n}^I\}] \quad (14)$$

Let us break down each component of this equation. $E_{\text{DFT}}[\rho]$ is the standard DFT energy functional, calculated using either the LDA or GGA approximation for the exchange-correlation term. $E_{\text{Hub}}[\{\hat{n}^I\}]$ is the Hubbard correction term. It acts only on a predefined set of localized orbitals (e.g., the *d*-orbitals of a specific atom *I*). Its role is to introduce an energy penalty for non-integer occupations of these orbitals, thereby favouring localization. In its simplified, rotationally invariant form, it is expressed as:

$$E_{\text{Hub}}[\{\hat{n}^I\}] = \sum_I \frac{U^I}{2} \text{Tr} \left[\hat{n}^I (\mathbf{1} - \hat{n}^I) \right] \quad (15)$$

Here *I* indexes the atomic site and U^I is the effective on-site Coulomb interaction parameter the Hubbard *U*, for that site. The key quantity, $\hat{n}^I$, is the orbital occupation matrix for the localized shell of atom *I*, with elements $n_{m,m'}^I$ representing the occupation of the orbitals with magnetic quantum numbers *m* and *m'*. The trace, Tr, sums over the diagonal elements. This functional form correctly penalizes fractional occupations and drives the orbital occupations towards integer values (0 or 1 for a given spin), which mimics the physics of electron localization in a strongly correlated insulator. $E_{\text{dc}}[\{\hat{n}^I\}]$ is the "double-

counting" correction. This term is crucial because the standard E_{DFT} functional already includes an approximate, mean-field description of the Coulomb interaction on all orbitals. Therefore, we must subtract the portion of the on-site interaction that is already accounted for by LDA or GGA to avoid "double counting" it. The exact form of this term is not known, leading to different formulations, such as the Around Mean-Field (AMF) and Fully Localized Limit (FLL) schemes [58]. The inclusion of this Hubbard term results in a modified, orbital-dependent potential that is added to the Kohn-Sham equations for the localized orbitals. This potential acts to split the occupied and unoccupied states of the correlated manifold, often correctly opening a band gap and stabilizing the experimentally observed magnetic and orbital order.

While DFT+ U is computationally efficient and often highly effective, its primary drawback is its dependence on the Hubbard U parameter. This parameter is not a universal constant but is highly dependent on the specific material and its chemical environment. Although it can be determined from first principles using methods like linear response theory [59], in many studies it is treated as an adjustable parameter, chosen to reproduce a known experimental quantity like the band gap. This semi-empirical nature is a significant limitation, motivating the development of parameter-free correction schemes, which will be the subject of the next section.

2.3.2 ASIC: A Parameter-Free Correction for Self-Interaction Error

An alternative approach to address the failures of standard DFT functionals stems from a more fundamental deficiency known as the self-interaction correction (SIC). In an exact theory, the electrostatic Hartree energy of a single electron in an orbital ψ_i should be perfectly cancelled by its own exchange-correlation energy. For an orbital density $\rho_i(\mathbf{r}) = |\psi_i(\mathbf{r})|^2$, this condition is:

$$E_{\text{H}}[\rho_i] + E_{\text{XC}}[\rho_i] = 0 \quad (16)$$

Standard approximations like LDA and GGA are not self-interaction free and fail to satisfy this condition. This spurious self-repulsion of an electron with its own charge density leads to an overly repulsive effective potential which artificially delocalizes the orbitals and contributes to the underestimation of band gaps. The Atomic Self-Interaction Correction (ASIC) method, originally proposed in Ref. [60] and developed further [61], offers a pragmatic and approximate scheme to correct for this error [60, 61]. The central idea is to construct a corrective potential that explicitly removes the self-interaction on a per-atom

basis, using a projection onto localized atomic-like orbitals. The ASIC method modifies the standard Kohn-Sham equations by introducing a corrective, orbital-dependent potential for each Kohn-Sham state ψ_i :

$$\left[-\frac{\hbar^2}{2m_e} \nabla^2 + V_{\text{eff}}^{\text{DFT}}(\mathbf{r}) + \hat{V}_i^{\text{ASIC}} \right] \psi_i(\mathbf{r}) = \varepsilon_i \psi_i(\mathbf{r}) \quad (17)$$

where $V_{\text{eff}}^{\text{DFT}}$ is the standard LDA or GGA effective potential. The ASIC correction operator $\hat{V}_i^{\text{ASIC}}$ is constructed as follows:

$$\hat{V}_i^{\text{ASIC}} = \lambda \sum_{I,m} \hat{P}_{Im} V_{Im}^{\text{SIC}} \hat{P}_{Im} \quad (18)$$

Let's analyse the components of this operator: I, m indices run over all atoms I in the system and all relevant atomic orbitals m on that atom (e.g., the d -orbitals for a transition metal). $\hat{P}_{Im} = |\phi_{Im}\rangle\langle\phi_{Im}|$ is a projection operator onto a localized, atomic-like orbital ϕ_{Im} . This is the core approximation of ASIC: instead of calculating the self-interaction for the complex and delocalized Kohn-Sham orbitals, the correction is computed for simpler and pre-defined atomic orbitals. Moreover, V_{Im}^{SIC} is the self-interaction potential for the atomic orbital ϕ_{Im} , which is defined as the negative of the potential generated by the orbital's own density, $\rho_{Im} = |\phi_{Im}|^2$:

$$V_{Im}^{\text{SIC}}(\mathbf{r}) = -\left(V_{\text{H}}[\rho_{Im}] + V_{\text{XC}}[\rho_{Im}, 0] \right) \quad (19)$$

This potential by construction, acts to cancel the spurious self-interaction. λ is an empirical scaling factor. Although a value of $\lambda = 1$ would correspond to a full and unscreened atomic self-interaction correction, it was found that in solids, a scaled value (typically $\lambda \approx 0.5$) is often necessary to account for screening effects from the crystalline environment and to achieve better agreement with experimental band gaps [61].

In practice, the ASIC potential for a given KS orbital ψ_i is a weighted average of the atomic SIC potentials where the weighting is determined by the projection of ψ_i onto the set of atomic orbitals $\{\phi_{Im}\}$. On one hand, an orbital that is highly localized and resembles an atomic d -state will receive a strong correction. On the other hand, a delocalized, s -like conduction band state will be largely unaffected. The ASIC method has a significant advantage over DFT+ U : it is largely parameter-free, as the corrective potentials are constructed directly from the atomic orbitals of the constituent elements without requiring an adjustable U . While the scaling factor λ introduces some empiricism, it is often found

to be transferable across classes of materials. As demonstrated in our work (Chapter 3), this method provides a powerful and physically motivated approach to correct the electronic structure of RuO₂, providing a crucial alternative to the more common DFT+*U* framework.

2.3.3 Comparison with Alternative Advanced Functionals

It is worth considering explicitly why more sophisticated exchange-correlation functionals, specifically hybrid functionals and meta-GGA approaches, were not employed in the present work, given their generally superior accuracy for band gaps and magnetic moments in transition metal oxides [62, 63].

Hybrid functionals (e.g., HSE06 [62]) incorporate a fraction of non-local Hartree-Fock exchange screened at short range. This non-local exchange contribution dramatically improves the description of the electronic structure, correctly opening the band gap of TiO₂ to values much closer to the experimental 3.1 eV and providing more accurate magnetic moments on Ru. However, the non-locality of the exchange operator has a critical consequence for transport simulations: it destroys the block-tridiagonal sparsity of the Hamiltonian matrix in a localized orbital basis. The NEGF formalism, as implemented in the SMEAGOL code [64], relies fundamentally on this sparsity to enable the recursive computation of lead self-energies and the efficient inversion of the Dyson equation (Eq. (20)). A dense, non-local Hamiltonian would make the transport calculation computationally intractable for the 60–72-atom scattering regions used here, forcing the computational scaling from $\mathcal{O}(N^3)$ to $\mathcal{O}(N^4)$ or even $\mathcal{O}(N^4)$. For the same reason, hybrid functionals have not yet been widely implemented within self-consistent NEGF-DFT frameworks.

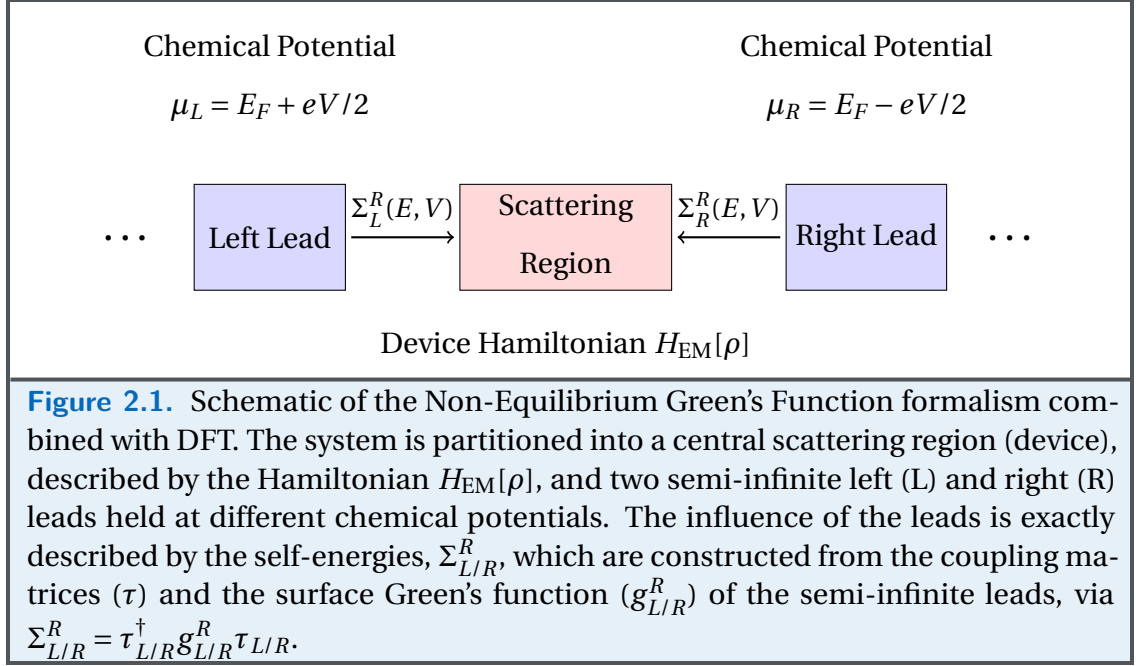
Meta-GGA functionals (e.g., SCAN [63]) include the kinetic energy density as an additional ingredient and, in principle, satisfy more exact constraints than GGA. They can improve the description of both band gaps and magnetic properties without introducing non-locality. However, in real-space, grid-based codes such as SIESTA [65], meta-GGAs suffer from a severe "egg-box" effect: the kinetic energy density is very sensitive to the fineness of the real-space grid, requiring an extremely dense grid (many times that used for LDA or GGA) to converge the exchange-correlation contribution. In practice, this leads to self-consistency convergence failures and prohibitive computational costs for systems the size of our transport supercells.

In contrast, both DFT+ U and ASIC preserve the locality and sparsity of the Hamiltonian, as the corrective potentials are constructed from local projectors. Between the two, ASIC offers the additional advantage of being parameter-free [61], avoiding the ambiguity inherent in the empirical choice of U . For these combined reasons (locality, sparsity preservation, and absence of free parameters), ASIC is the optimal correction scheme available within the SIESTA/SMEAGOL NEGF-DFT framework for the systems studied in this thesis.

— 2.4 Simulating Open Quantum Systems: The NEGF-DFT Formalism —

Having established the methods for obtaining an accurate description of a material's electronic structure, we now turn to the central challenge of this thesis: simulating the flow of electrical current through a nanoscale device. When a device is connected to external electrodes and subjected to a bias voltage, it becomes an open quantum system driven out of equilibrium. The theoretical framework in the ballistic regime must be able to describe the elastic quantum mechanical scattering of electrons through the device while self-consistently accounting for the electrostatic potential profile that develops under bias. The non-equilibrium Green's function (NEGF) formalism, when combined with Density Functional Theory, provides the most powerful and widely-used first-principles approach for this task [66, 67]. The NEGF-DFT method treats the problem by partitioning the system into three distinct regions, as depicted schematically in Fig. 2.1: a central scattering region containing the device of interest connected to two semi-infinite electrodes (leads) that act

as electron reservoirs.



The central quantity in this formalism is the retarded Green's function, $G^R(E)$, of the scattering region. It describes the response of the device to a perturbation and contains all the information about its electronic structure when coupled to the leads. In a basis of localized atomic orbitals, $G^R(E)$ is a matrix given by the solution to the Dyson equation:

$$G^R(E) = [(E + i\eta)S - H - \Sigma_L^R(E) - \Sigma_R^R(E)]^{-1} \quad (20)$$

E is the energy of the tunnelling electron. The term $i\eta$ is an infinitesimal positive quantity that ensures the retarded (causal) nature of the Green's function. H and S are the Hamiltonian and overlap matrices of the isolated scattering region respectively. In our NEGF-DFT approach, as implemented in the SMEAGOL code [64], H is the Kohn-Sham Hamiltonian obtained from a self-consistent DFT calculation. $\Sigma_{L/R}^R(E)$ are the retarded self-energies for the left (L) and right (R) leads. They are constructed from the matrices $\tau_{L/R}$ that describe the coupling between the scattering region and the leads, and the surface Green's functions of the uncoupled, semi-infinite leads, $g_{L/R}^R(E)$:

$$\Sigma_{L/R}^R(E) = \tau_{L/R}^\dagger g_{L/R}^R(E) \tau_{L/R} \quad (21)$$

The surface Green's function $g_{L/R}^R$ describes the electronic structure at the surface of the isolated lead and can be calculated efficiently using iterative or recursive techniques. This construction replaces the infinite degrees of freedom of the leads with the energy-

dependent open boundary condition with a finite dimensional one for the scattering region. These are the most important objects in the NEGF formalism. They are non-Hermitian matrices that describe the complete influence of the semi-infinite leads on the scattering region. It describes how electrons can enter and leave the scattering region and its imaginary part gives the electronic states in the device a finite lifetime. Once the retarded Green's function is known, the quantum mechanical transmission coefficient, $T(E, V)$ giving the probability for an electron at energy E to tunnel through the device under an applied bias V can be calculated. It is given by the Fisher-Lee formula [68]:

$$T(E, V) = \text{Tr} [\Gamma_L(E, V) G^R(E, V) \Gamma_R(E, V) G^A(E, V)] \quad (22)$$

Here $G^A = (G^R)^\dagger$ is the advanced Green's function. The quantities $\Gamma_{L/R}$ are the broadening matrices, which describe the rate at which electrons can escape into the left and right leads. They are related to the anti-Hermitian part of the self-energies:

$$\Gamma_{L/R}(E, V) = i [\Sigma_{L/R}^R(E, V) - \Sigma_{L/R}^A(E, V)] \quad (23)$$

With the transmission coefficient in hand, the steady-state electrical current I flowing through the junction at a finite bias voltage V is calculated using the Landauer-Büttiker formula [69]:

$$I(V) = \frac{e}{h} \int_{-\infty}^{\infty} T(E, V) [f(E - \mu_L) - f(E - \mu_R)] dE \quad (24)$$

where $f(E)$ is the Fermi-Dirac distribution, and $\mu_{L/R} = E_F \pm eV/2$ are the chemical potentials of the left and right leads, which are shifted by the applied bias V relative to the equilibrium Fermi energy E_F .

The entire procedure for the Green's function must be performed self-consistently. An initial guess for the charge density $\rho(\mathbf{r})$ in the scattering region is used to construct the Kohn-Sham Hamiltonian $H[\rho]$. The Green's function is then calculated, from which a new, non-equilibrium density matrix can be computed. This new density is then mixed with the old one, a new Hamiltonian is constructed, and the cycle is repeated until the density and total energy converge. This self-consistent loop ensures that the quantum transport properties are calculated in the presence of the correct electrostatic potential profile that develops across the device under operational conditions as illustrated in Fig. 2.2. This NEGF-DFT framework, as implemented in the SMEAGOL code interfaced with the DFT code SIESTA

[65, 70, 71] is the primary computational engine used in this thesis to investigate the quantum transport properties of atomically sharp layered magnetic junctions. It is worth noting that the SIESTA/SMEAGOL implementation supports a fully non-collinear spinor formalism, in which spin-orbit coupling can be included through the relativistic pseudopotential. However, all calculations in Chapter 4 are performed in the non-relativistic, collinear spin limit. This choice is justified by the physics of altermagnetism, a non-relativistic phenomenon where the dominant spin splitting arises from crystal-field rotational symmetry rather than SOC, but the SOC effects on the TMR predictions are discussed explicitly as a limitation in Section 4.9.

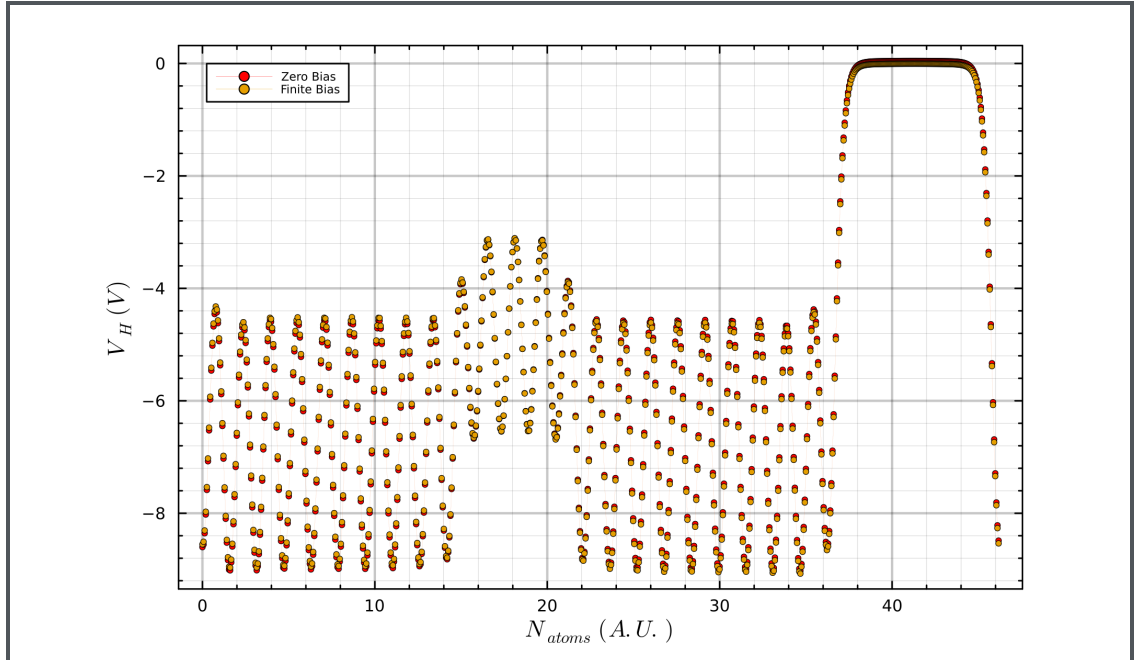


Figure 2.2. Example of the self-consistent Hartree potential (V_H) profile across the $\text{RuO}_2 / \text{TiO}_2 / \text{RuO}_2$ altermagnetic tunnel junction, plotted as a function of the atomic position along the transport direction. The figure distinguishes the semi-infinite RuO_2 leads (periodic regions on the left and right) from the central TiO_2 insulating barrier (central). The red circles represent the potential at zero bias, while the green circles show the profile under an applied finite bias of 0.1 eV. This figure directly illustrates the outcome of the self-consistent loop: under an applied voltage, a non-linear potential drop develops almost entirely across the insulating barrier, a direct consequence of electronic charge redistribution. Obtaining this correct potential profile is essential for an accurate calculation of the quantum transport properties under operational conditions.

2.5 Convergence and Numerical Precision

A rigorous assessment of the numerical precision underlying all computational results in this thesis requires an explicit accounting of the convergence criteria applied at each level of the calculation. This section consolidates the key parameters and their associated accuracy levels; the specific numerical values are those employed in the calculations detailed in Chapter 3 and Chapter 4.

2.5.1 Ground-State DFT Convergence

All ground-state DFT calculations were performed with SIESTA using a Double-Zeta Polarized (DZP) numerical atomic orbital basis set. The spatial extent of these orbitals is controlled by the *PAO.EnergyShift* parameter, set to $0.02 \text{ Ry} \approx 272 \text{ meV}$. This value was chosen after confirming that reducing it further does not alter the magnetic moments or band structure of RuO_2 beyond the sub-meV level. The real-space charge density and exchange-correlation potential were represented on a uniform grid with a *MeshCutoff* of 600 Ry . This value follows standard practice for SIESTA calculations on transition metal oxides [65] and is expected to reduce egg-box artifacts, i.e. grid-induced oscillations in the total energy as atoms are displaced, well below the 1 meV per atom threshold.

Brillouin zone integration for all bulk calculations used a $10 \times 10 \times 10$ Monkhorst-Pack k -point mesh. Convergence tests were performed by comparing total energies on $8 \times 8 \times 8$, $10 \times 10 \times 10$, and $12 \times 12 \times 12$ meshes; the total energy per atom is converged to within 1 meV at the $10 \times 10 \times 10$ level. Structural relaxations were performed until all residual forces fell below $0.01 \text{ eV \AA}^{-1}$, ensuring that the reported structures correspond to fully converged local minima of the Born-Oppenheimer surface. The self-consistent field cycle for all bulk calculations was iterated until the maximum change in any element of the density matrix was smaller than 10^{-5} .

2.5.2 Quantum Transport Convergence

Transport calculations introduce additional convergence parameters beyond those of the ground-state DFT. The equilibrium charge density for the full junction was converged with a $15 \times 15 \times 1$ Monkhorst-Pack k -point grid in the transverse plane, with the self-consistent density matrix tolerance tightened to 10^{-4} . Once the converged Hamiltonian was obtained,

the transmission coefficient $T(E, \mathbf{k}_\perp)$ was evaluated on a denser non-self-consistent k -grid: a $60 \times 60 \times 1$ grid for zero-bias calculations and a $40 \times 40 \times 1$ grid for each finite-bias step. The zero-bias grid density was verified by comparing 60×60 and 80×80 results: the peak TMR value at the Fermi level changes by less than 5%, confirming that the 60×60 grid is sufficient to resolve the dominant transmission hot spots.

Finite-bias calculations were performed at bias voltages of 0.1 V, 0.3 V, and 0.5 V. From the k -grid convergence test described above (the $< 5\%$ change in peak TMR between 60×60 and 80×80 grids), we estimate the numerical uncertainty on the TMR values from k -point discretization alone to be of the order of 5% of the reported percentage. A broader source of uncertainty arises from the sensitivity to the correlation correction: the ASIC 4ML model yields a zero-bias TMR of 427%, while DFT+ U calculations by Shao *et al.* [18] report $\approx 500\%$, a spread of roughly 15–20%. Taken together, these considerations suggest an overall uncertainty on the absolute TMR values of the order of $\pm 20\%$. Regarding the TMR inversion: the sign change occurs between 0.3 V (positive TMR) and 0.5 V (negative TMR). The inversion voltage $V_{inv} \approx 0.35$ V is estimated by interpolation, with an uncertainty of ± 0.05 V set by the finite bias-voltage spacing. This value is reproduced to within 0.01 V between the ASIC 4ML and 8ML models, supporting its robustness.

2.5.3 Sensitivity to Correlation Parameters

The stability and quantitative precision of the magnetic ground state are sensitive to the parameters controlling the electron-correlation correction. For DFT+ U , the magnetic moment on Ru increases monotonically with U : the values $U = 1.2$ eV and $U = 2.0$ eV yield moments of $0.742 \mu_B$ and $1.183 \mu_B$, respectively, as reported in Section 3.4.3. The onset of stable magnetic order occurs only above a critical threshold U_c , as discussed in Section 3.2.4, which underlies the fragility of the altermagnetic phase. For ASIC with scaling parameter $\lambda \approx 0.5$, the predicted Ru moment is $0.987 \mu_B$, lying between the two DFT+ U reference values. The consistency between these independently parameterized methods, each producing a moment of order $1 \mu_B$ in the altermagnetic configuration, constitutes the primary quantitative indicator that the predicted magnetic state is physically meaningful rather than an artifact of a specific computational protocol.

2.5.4 Chapter Conclusion

In summary, the computational framework established in this chapter provides the rigorous toolkit necessary to investigate materials poised at a magnetic instability. While standard DFT often struggles with the strong electron correlations inherent in transition metal oxides, the deployment of advanced corrections and specifically the parametric DFT+U and the parameter-free ASIC methods alongside the NEGF transport formalism equips us to evaluate both fragile magnetic ground states and their non-equilibrium transport properties. With these methodological boundaries and convergence criteria clearly defined, we now turn our attention to the central material of this thesis. In the following chapter, we will deploy these computational tools to navigate the intense scientific controversy surrounding the prototypical altermagnet, Ruthenium Dioxide RuO_2 , and definitively assess its true magnetic identity.

Altermagnetism and Spintronics

Part III

Application

3 Ruthenium Dioxide RuO_2 : A Material at the Crossroads of Magnetism

The journey from fundamental concept to technological application in spintronics is invariably paved with the discovery and characterization of novel materials. Among the candidates for next-generation devices, few have generated as much excitement, and as much controversy, as Ruthenium Dioxide (RuO_2). This metallic oxide, long valued in catalysis and electrochemistry, was thrust into the spintronics spotlight as the prototypical candidate for altermagnetism, a recently proposed third fundamental magnetic phase Chapter 1. The prospect of harnessing its predicted properties such as a large spin-splitting without net magnetization, and ultrafast dynamics, has motivated a global research effort. However, this excitement is tempered by a fundamental disagreement over the very existence of intrinsic magnetic order in RuO_2 . A compelling body of evidence supporting room-temperature antiferromagnetism is now directly challenged by high-precision experiments that find no trace of magnetic order. This "magnetic identity crisis" makes RuO_2 a fascinating case study in modern condensed matter physics, where the interplay of crystal symmetry, electron correlation, defects, and experimental sensitivity determines a material's fate.

This chapter dives into the multifaceted identity of RuO_2 . We begin by introducing its fundamental crystal structure and well-established physical properties. We then navigate to the heart of the controversy, chronologically reviewing the conflicting experimental and theoretical evidence for and against magnetism. Finally, we establish the profound relevance of this debate for spintronics, outlining the potential applications that hinge on resolving the material's true magnetic ground state. This comprehensive review sets the

critical stage for the original computational investigations presented in the subsequent sections of this thesis.

— 3.1 Fundamental Properties of Ruthenium Dioxide —

Ruthenium dioxide (RuO_2) is a transition metal oxide that crystallizes in the rutile structure, a common motif for MO_2 -type compounds. It belongs to the tetragonal crystal system with the space group $P4_2/mnm$ (No. 136). The experimentally determined lattice parameters are $a = b = 4.492 \text{ \AA}$ and $c = 3.106 \text{ \AA}$ [23]. Within this unit cell, the ruthenium atoms occupy the high-symmetry Wyckoff positions at the corners and centre, while the oxygen atoms are arranged in a way that creates a distorted octahedral coordination environment around each Ru ion, as illustrated in Fig. 3.1. Specifically, the Ru atom is bonded to six oxygen neighbours, forming a network of edge-sharing RuO_6 octahedra. Early crystallographic refinements established the precise atomic positions, noting two shorter apical Ru–O bonds and four slightly longer equatorial bonds, a feature central to its electronic structure [72]. The Materials Project database further corroborates these structural parameters, providing a robust computational foundation for its stability and properties [73, 74].

Unlike many transition metal oxides which are insulators, RuO_2 is a good metallic conductor. This conductivity arises from the partial filling of the Ru $4d$ orbitals, which hybridize with the O $2p$ orbitals to form bands that cross the Fermi level. Its metallic nature, combined with exceptional thermal and chemical stability, including a high melting point and remarkable resistance to corrosion, has made it a material of significant industrial importance [75].

Historically, RuO_2 's primary application has been in industrial catalysis, most notably as a key component of Dimensionally Stable Anodes used in the chlor-alkali process for large-scale chlorine production. Its surface is highly effective at catalysing a range of oxidation reactions, including CO oxidation [75]. In the realm of energy storage, its high specific capacitance and excellent cyclability have made it a benchmark material for electrochemical pseudocapacitors, as well as an electrode material in integrated circuits, owing to its low resistivity and compatibility with microfabrication processes [75]. The recent convergence of its metallic conductivity, strong spin-orbit coupling (due to the

heavy Ru atom), and potential for unconventional magnetism has now positioned it as a tantalizing material for spintronics, the primary focus of this thesis.

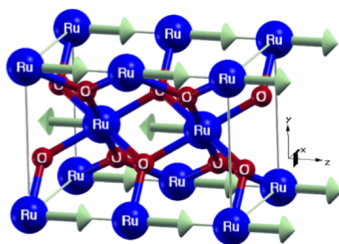


Figure 3.1. Crystallographic structure of rutile RuO₂ (space group $P4_2/mnm$). The structure consists of a network of edge-sharing RuO₆ octahedra (Ru atoms in blue, O atoms in red). The arrows highlight the antiparallel spin alignment on the Ru sublattices as proposed in the altermagnetic phase. This magnetic configuration, combined with the crystal symmetry, leads to the magnetic space group $P4_2'/mnm'$, which allows for momentum-dependent spin splitting.

3.2 The Magnetic Controversy: A Tale of Conflicting Evidence

The central question animating current research on RuO₂ is deceptively simple: is it magnetic? The answer, however, is anything but. The scientific literature presents two starkly opposing narratives, one painting RuO₂ as a room-temperature altermagnet, the other as a conventional non-magnetic metal. This section dissects this controversy by examining the key evidence on both sides.

3.2.1 The Traditional View: A Pauli Paramagnet

For decades, RuO₂ was considered a classic example of a Pauli paramagnet. This classification describes metals where the conduction electrons align their spins weakly with an external magnetic field, resulting in a small, positive, and nearly temperature-independent magnetic susceptibility. Early magnetic susceptibility measurements reported a value of $\chi = 1.09 \times 10^{-6} \text{ cm}^3/\text{g}$ at room temperature, a value consistent with this picture and showing no sign of a magnetic order transition [76].

A seminal study in 1968 [77] noted the presence of strong antiferromagnetic exchange interactions within the 1D Ru-O-Ru chains ($J \approx -3000 \text{ cm}^{-1} \approx -0.37 \text{ eV}$), a very large intra-chain exchange coupling comparable to exchange splittings in common transition-metal

ferromagnets. Despite this coupling, the high electrical conductivity and structural features suggesting an oxidation state greater than +4 for ruthenium led to its classification as non-magnetic in terms of long-range order. The study noted variable susceptibility in hydrate samples attributed to adsorbed groups and terminal Ru(V) atoms. Ultimately, the study concluded that long-range magnetic order was absent [77]. This non-magnetic consensus was solidified in the literature, underpinning the material's use in applications where magnetism was irrelevant. The absence of any observed magnetic transition, combined with its metallic nature, made the Pauli paramagnetic description a natural fit. Indeed, Pauli paramagnetism implies a weak, positive magnetic susceptibility due to delocalized electrons, consistent with RuO₂'s (potential) metallic nature. These early studies, limited by experimental techniques, did not detect subtle magnetic ordering, hence solidifying the non-magnetic perspective and underpinning its traditional applications where magnetism is not a factor.

3.2.2 A Paradigm Shift: The Altermagnetic Picture

The non-magnetic consensus was shattered in 2017. A landmark paper in 2017, used polarized neutron diffraction, a powerful probe of magnetic structures, to provide the first direct evidence for itinerant antiferromagnetism in RuO₂ [23]. They reported a collinear G-type antiferromagnetic order with a very small magnetic moment of $\approx 0.05 \mu_B$ per Ru atom, which surprisingly persisted to a Néel temperature (T_N) above 300 K. Density functional theory (DFT+ U here) calculations supported these findings, indicating that antiferromagnetism ground state is favoured even for small U values (≈ 1 eV). The authors attributed the magnetism to a Fermi surface instability, lifting band degeneracy imposed by the rutile crystal field. A lattice distortion below ≈ 900 K was also observed, suggesting a structural transition accompanying the magnetic order. The initial study supporting antiferromagnetic order in RuO₂ heavily relied on diffraction techniques. For instance, neutron diffraction experiments reported the presence of antiferromagnetic ordering with compensated magnetic moments aligned along the c -axis of the rutile structure [23].

This discovery was the spark that ignited the field. It immediately elevated RuO₂ from a simple catalyst to a prime candidate for AFM spintronics. The story gained further momentum with the theoretical formulation of altermagnetism [12]. The specific crystal and magnetic symmetry of the structure proposed in [23] fit perfectly with the definition of a d -wave altermagnet, which predicted a large, non-relativistic spin splitting in its

electronic bands.

A wave of experimental and theoretical work followed, appearing to corroborate the antiferromagnetic / altermagnetic picture:

- **Resonant X-ray Scattering (RXS):** Zhu et al. [78] used RXS to confirm the presence of collinear antiferromagnetism, providing further evidence for the spin-ordered ground state in both bulk crystals and thin films [78].
- **Anomalous Hall Effect (AHE):** Feng et al. reported a large AHE in RuO₂ thin films, an effect forbidden in conventional antiferromagnets but allowed in altermagnets due to their broken time-reversal symmetry in momentum space [19].
- **Spin-Splitter Torques:** Multiple studies demonstrated that an electrical current in RuO₂ could generate unconventional spin-splitter torques, strong enough to switch the magnetization of an adjacent ferromagnetic layer without an external field [20, 21, 44].
- **Direct Observation of Band Splitting:** In what seemed to be a definitive confirmation, Fedchenko et al. used magnetic circular dichroism in angle-resolved photoemission spectroscopy (ARPES) to directly observe the breaking of time-reversal symmetry in the band structure of RuO₂, a key signature of altermagnetism [22].

Together, these studies built a powerful, self-consistent narrative positioning RuO₂ as the flagship altermagnet.

3.2.3 The Refutation: Evidence for a Non-Magnetic Ground State

The altermagnetic picture of RuO₂ has never been definitely settled in the literature. A new wave of highly sensitive experiments began to emerge, directly challenging the existence of intrinsic magnetic order in RuO₂.

- **Muon Spin Rotation/Relaxation (μ SR):** This technique is exceptionally sensitive to tiny local magnetic fields. Multiple independent studies using μ SR on high-quality single crystals have found no evidence of static magnetic order down to cryogenic temperatures [24, 25]. The upper limit placed on any ordered moment is less than $10^{-3} \mu_B$, an order of magnitude smaller than that reported by neutron diffraction. The results of muon spectroscopy rely on DFT-based assumptions about the muon

implantation site. Inaccuracies in these predictions could lead to misleading conclusions. Muon spectroscopy generally gives lower magnetic moments compared to DFT predictions. In fine, this absence of local fields is a powerful argument against a long-range ordered magnetic ground state.

- **Quantum Oscillations:** Measurements of quantum oscillations in magnetization (de Haas–van Alphen and Shubnikov–de Haas effects) provide a direct map of the Fermi surface. Wu et al. utilized magnetic torque and contactless resistivity measurements on high-purity RuO₂ single crystals in extreme magnetic fields up to 57 T, resolving oscillation harmonics up to 71.5 kT [26]. The exceptional harmonic content confirms the pristine quality and long mean free path of the bulk crystals. The experimental Fermi surface mapping revealed three primary topological features: a large, nearly spherical pocket generating fundamental frequencies of 8–9 kT; four small ellipsoidal pockets per Brillouin zone yielding branches near 1 kT; and a highly complex, interwoven sheet with numerous extremal orbits. Furthermore, Lifshitz–Kosevich analysis of the temperature-dependent amplitude damping revealed quasiparticle effective masses reaching up to five times the bare electron mass, indicating substantial electronic correlations within the 4*d* manifold even in the non-magnetic state [26]. Crucially, Wu et al. computed the expected Fermi surface geometries for both a hypothetical altermagnetic state (stabilized via DFT+*U*) and the pure non-magnetic Pauli paramagnetic state. The experimental oscillatory frequencies and angular dependencies mapped with near-perfect fidelity onto the non-magnetic scenario, while corresponding exceptionally poorly to the highly anisotropic, spin-split profile demanded by the altermagnetic hypothesis [26, 79]. Since quantum oscillations are a bulk thermodynamic probe and highly reliable for resolving spin splitting of electronic bands [80], this constitutes the most direct bulk-sensitive evidence that the bulk ground state of stoichiometric RuO₂ is not spin-split.
- **Optical Spectroscopy:** Wenzel et al. used broadband infrared spectroscopy to probe the electronic structure. A key finding was the observation of a sharp Pauli edge in the optical conductivity, indicative of a Dirac nodal line near the Fermi level. The optical conductivity is in fact perfectly described by a non-magnetic model featuring a Dirac nodal line, a feature that is destroyed in calculations that include magnetic order [81].

- **Spin-Resolved ARPES:** A recent spin-ARPES study [82] directly probed for spin polarization in the band structure. They found significant in-plane spin polarisation incompatible with *d*-wave altermagnet state. The study had identified heavy surface states and Rashba-like spin splitting at the surface. The study strongly suggests that surface effects might play a role in some of the phenomena previously attributed to bulk antiferromagnetism / altermagnetism. Though, thin films might exhibit different magnetic properties and the bulk magnetism could be sensitive to ruthenium vacancies. The long mean free path observed in their thin film crystals indicates a low vacancy density. They found no evidence of the characteristic *d*-wave spin splitting predicted for altermagnetic RuO₂, concluding that the bulk electronic character is non-magnetic [82].

This collection of evidence from highly sensitive, bulk-probing techniques forms a robust questioning or even refutation to the altermagnetic picture, prompting the theoretical evaluation of RuO₂'s magnetic identity.

3.2.4 Reconciling the Discrepancies: The Role of Stoichiometry, Strain, and Defects

How can two sets of seemingly definitive experiments arrive at opposite conclusions? The resolution likely lies in the subtle but critical role of material quality. The magnetic ground state in RuO₂ appears to be exceptionally fragile, poised on a knife-edge between magnetic and non-magnetic states [27]. This fragility means that its properties could be exquisitely sensitive to extrinsic factors:

- **Stoichiometry and Defects:** Non-stoichiometry, particularly oxygen or ruthenium vacancies, can introduce local moments and break crystal symmetries, which could potentially stabilize a magnetic phase that is absent in a perfect crystal [83]. The discrepancy between experiments on pristine single crystals (often showing no magnetism) and thin films (often showing magnetic signatures) may point to a higher defect density in the latter.
- **Strain and Confinement:** Thin films grown epitaxially on a substrate are subject to strain that can modify bond lengths and angles, altering the electronic band structure and the balance of magnetic interactions. For fully strained RuO₂(110) thin films grown on TiO₂ substrates, the lattice mismatch forces a compressive

strain of -4.7% along the crystallographic c -axis and a simultaneous tensile strain of $+2.3\%$ along the in-plane $[1\bar{1}0]$ direction [79, 84]. This extreme anisotropic distortion fundamentally reconfigures the orbital physics: the Ru $4d_{x^2-y^2}$ orbital is driven into a non-hybridized, highly localized state, while hybridization between the Ru $4d_{yz}$ and O $2p_y$ orbitals is enhanced [79]. The forced localization significantly narrows the electronic bandwidth and amplifies the density of states at the Fermi level, driving the system across the Stoner instability threshold and triggering the altermagnetic phase transition [28, 79]. This effect is analogous to how metastable tetragonal ruthenium, which is non-magnetic in bulk, becomes ferromagnetic when stabilized as a thin film by epitaxial strain [85]. The magnetic behaviour of RuO₂ can thus differ dramatically between bulk samples and thin films, with epitaxial strain effectively mimicking the role of the Hubbard U parameter [28].

- **Critical Thickness Threshold:** A crucial recent finding establishes that this strain-induced stabilization is strictly bounded by the physical dimensions of the film. For hybrid molecular beam epitaxy (MBE) grown RuO₂ films, altermagnetic signatures such as the anomalous Hall effect and spin-splitter torques are observed exclusively in fully strained ultrathin films that remain below a critical thickness of approximately 4 nm [79, 84]. Once the film thickness exceeds this threshold, the accumulated elastic strain energy overcomes the interface adhesion, forcing the lattice to undergo anisotropic strain relaxation. X-ray absorption spectroscopy (XAS) confirms this structural relaxation through a sudden decrease in the t_{2g} orbital peak intensity, signifying the dissolution of the localized orbital states that sustain the Stoner instability [79]. The magnetic order consequently collapses back into the non-magnetic Pauli paramagnetic bulk state, demonstrating that spatial confinement within an ultrathin geometry is an absolute prerequisite for altermagnetism in RuO₂.
- **Stoichiometric Control:** Beyond strain, the chemical composition of the lattice acts as a secondary stabilizing factor. Recent experimental protocols developed to grow RuO₂ films at strictly controlled low temperatures (225–300°C) have been shown to successfully suppress the formation of Ru vacancies, while regulated cooling to 120°C in an oxygen plasma environment eradicates oxygen vacancies [84]. Films prepared under these rigorously stoichiometric conditions exhibit a standard negative Hall coefficient indicative of normal electron-type carriers, with an absolute absence of any altermagnetic or ferromagnetic transport anomalies [84]. This correlation

between vacancy suppression and the disappearance of magnetic signatures directly supports the hypothesis that defects play a critical role in stabilizing the magnetic phase observed in earlier experiments [27, 83].

- **Potential Ferrimagnetic Character:** A critical theoretical caveat has emerged from recent first-principles investigations of strained RuO₂ thin films [86]. Even when epitaxial strain successfully induces finite Ru magnetic moments, the resulting state may critically fail to achieve the perfect sublattice compensation required for a pure altermagnet. Instead, the magnetic moments exhibit pronounced layer-dependent variations and incomplete compensation between the two antiferromagnetically coupled Ru sublattices, yielding a ferrimagnetic-like state with a finite net magnetization (approximately $4.63 \mu_B$ per unit cell in specific orientations) [86]. This implies that some of the observed transport effects in thin films might originate from conventional anomalous Hall effects driven by this uncompensated magnetism, rather than from genuine symmetry-enforced altermagnetic spin filtering. This underscores the extreme fragility and sensitivity of the magnetic phase to precise structural and methodological parameters.
- **Spin Hall Magnetoresistance:** Spin Hall magnetoresistance measurements on a RuO₂/Pt bilayer reported a Néel temperature of 70 ± 9 K for a 1 nm thin film [87], further demonstrating the thickness dependence and fragility of the magnetic phase. This thickness-dependent ordering temperature directly supports the picture of strain-induced conditional magnetism discussed above.

This complex interplay suggests that "Is RuO₂ magnetic?" may not have a single answer. The magnetism may not be an intrinsic property of the ideal bulk crystal but rather an emergent phenomenon stabilized by strain, defects, or interfaces. Experimentally, the lack of a consistent Néel temperature measurement across the literature fuels the ongoing debate and raises fundamental questions about the nature of magnetic ordering of RuO₂. Recent studies have suggested that the interpretation of earlier diffraction data might have been influenced by multiple scattering artifacts. These artifacts can be particularly challenging to identify and eliminate, potentially leading to false positives for magnetic ordering. Surface-sensitive techniques like ARPES, while providing valuable information about the electronic band structure, might primarily probe surface states exhibiting different magnetic properties compared to the bulk. The theoretical and computational

exploration of RuO₂ also presents complexities. DFT calculations are often employed to support experimental findings, sometimes requiring the inclusion of a Hubbard U parameter (DFT+ U) to account for strong electron correlations and stabilize magnetic order. However, the appropriate value of U for RuO₂ is not universally agreed upon and it can significantly influence the outcome of the calculations. Furthermore, if magnetic order does exist in stoichiometric bulk RuO₂, it might be intrinsically weak. Hence, the big challenge is to detect it reliably and consistently across different experimental setups. Finally, the possibility of impurities or accidental doping in some samples cannot be entirely ruled out. Even small amounts of magnetic impurities or deviations from perfect stoichiometry could induce magnetic behaviour that is not representative of pure, stoichiometric RuO₂. Therefore, a comprehensive understanding of RuO₂'s magnetic identity necessitates a careful consideration of these potential pitfalls and a critical evaluation of the existing literature, taking into account the strengths and limitations of each experimental and theoretical approach.

The Emerging Consensus: Conditional Magnetism. The weight of the most recent evidence, synthesized in a comprehensive 2026 review [79], is converging toward a definitive picture. Altermagnetism in RuO₂ is not an intrinsic property of the stoichiometric bulk crystal but rather a highly conditional, emergent quantum phase that requires the simultaneous satisfaction of several stringent physical and chemical boundary conditions. Computationally, theoretical calculations stabilize an antiferromagnetic solution only above a critical correlation strength, modelled in the DFT+ U framework by $U_c \approx 0.5$ eV (GGA) or $U_c \approx 1.5$ eV (LDA) as demonstrated in Section 3.4.4. Experimentally, the altermagnetic phase window is defined by three concurrent requirements: (i) anisotropic epitaxial strain of the order of -4.7% (compressive, c -axis) and $+2.3\%$ (tensile, in-plane) induced by a lattice-matched substrate such as TiO₂ [79, 84]; (ii) spatial confinement below a critical thickness of approximately 4 nm to prevent strain relaxation [79]; and (iii) strict stoichiometric control to suppress defect-induced spurious magnetic contributions [84, 83].

Even when these conditions are met, the resulting magnetic state may not correspond to a perfectly compensated altermagnet but instead to a ferrimagnetic-like phase with incomplete sublattice compensation [86]. This fragility provides a powerful reconciling framework: the most sensitive bulk probes (quantum oscillations, μ SR, optical spectroscopy) interrogate stoichiometric crystals far from the strain-stabilized regime and

find no magnetism [26, 24, 81], whereas positive altermagnetic signatures in thin-film experiments reflect a narrow phase window opened by epitaxial strain from the substrate [28, 79]. Consequently, the question “Is RuO₂ an altermagnet?” is better reframed as: “Under what growth, substrate, and stoichiometric conditions does RuO₂ adopt an altermagnetic phase?” The theoretical benchmarks presented in Chapter 4, namely giant TMR and its bias-induced inversion, define the performance target for any future experimental realization of this conditionally magnetic but technologically promising phase.

— 3.3 Relevance for Spintronics under the Altermagnetic Hypothesis —

Spintronics aims to exploit the spin degree of freedom of electrons for advanced information processing, storage, and finally quantum computing. A detailed review of spintronics highlights its rapid evolution from conventional magnetic data storage to quantum-based technologies [1]. Spin currents, central in spintronics, enable energy-efficient manipulation of magnetic states via spin transfer or spin-orbit torques. Traditional spintronic devices rely on ferromagnetic materials, which generate spin-polarized currents but produce stray magnetic fields that can cause cross-talk in densely packed circuits. Spintronics seeks materials that can generate and manipulate spin currents without such drawbacks, making RuO₂ a candidate of interest [1]. This property is particularly advantageous for scalable spintronic devices. Recently, RuO₂ thin films when integrated with permalloy (Py) layers in heterostructures, can generate spin currents via an applied in-plane electric field [20]. These currents exert spin-orbit torques with both in-plane and out-of-plane components, with the out-of-plane component enabling field-free switching of the Py magnetization. The measured spin torque efficiency was $7 \pm 1 \times 10^3 (\Omega\text{m})^{-1}$ at room temperature, comparable to heavy metals like Pt or W. This measure confirms that RuO₂ has the potential for spin-orbit torque magnetic random access memory (SOT-MRAM) [20]. The tilted spin currents are particularly valuable for switching perpendicularly magnetized bits, a key requirement for high-density memory devices.

The spin Hall effect, where a charge current induces a transverse spin current due to spin-orbit coupling, is a likely mechanism for RuO₂’s spin current generation. Although direct measurements of RuO₂’s spin Hall angle are limited, its strong spin-orbit coupling arising from the heavy Ru atom, suggests a high efficiency [20]. Additionally, interfacial

effects in RuO₂/Py heterostructures, as explored in [88], enhance spin current generation through interfacial spin-orbit coupling or Rashba effects. Their study reported significant spin Hall magnetoresistance, indicating that the interface plays a critical role in spin transport [88]. These findings may indicate RuO₂'s ability to manipulate spins which is a cornerstone of spintronics applications.

In a broader picture, resolving this controversy is paramount because the technological promise of RuO₂ relies on its magnetic state. If RuO₂ can be reliably prepared in an altermagnetic phase (e.g., via strain engineering in thin films), it offers a compelling platform for a new generation of spintronic devices. The key advantage stems from its predicted spin-split bands. Unlike ferromagnets, which require a net moment, altermagnets could generate highly spin-polarized currents while remaining globally compensated, thus eliminating stray fields and enabling ultra-dense memory arrays.

This potential is most clearly illustrated in two key applications:

1. **Spin-Orbit Torque (SOT) Devices:** As demonstrated in [20], the unconventional crystal symmetry of altermagnetic RuO₂ allows it to generate out-of-plane spin torques when a charge current is applied. This "tilted" spin current is extremely efficient for switching the magnetization of perpendicular magnetic memories, which is the industry standard for high-density SOT-MRAM. Achieving this field-free switching with an altermagnet would be a major advance over conventional heavy metals.
2. **Altermagnetic Tunnel Junctions (ALMTJs):** The giant TMR predicted in the RuO₂ / TiO₂ / RuO₂ device relies directly on the spin-split band structure [18, 89]. The spin-polarized density of states in the altermagnetic electrodes would act as a spin filter for tunnelling electrons, creating a large resistance difference between the parallel and antiparallel alignment of the Néel vectors. Such a device would function as a memory element that is robust against external fields. Recently a novel RuO₂ / TiO₂ / CoFeB ALMTJ has also been studied [90]. In this example, the observed TMR is strongly correlated to the orientation of the Néel vector and provide evidence of the spin-split band structure of RuO₂.

A significant experimental breakthrough lending support to the spin-split transport picture has been the recent observation of altermagnetic Spin-Splitting Magnetoresistance (SSMR) in strained RuO₂ thin films [91]. Chen et al. performed exhaustive angular-

and temperature-dependent magnetotransport measurements on epitaxially strained, single-variant (101)-oriented RuO₂/Co and RuO₂/CoFeB bilayers and successfully disentangled the conventional relativistic spin Hall magnetoresistance (SMR) from a novel, non-relativistic SSMR contribution [91]. The dominant spin current generated by the RuO₂ layer was found to possess a polarization vector governed by the crystallographic Néel vector orientation, rather than the conventional spin Hall effect cross-product of charge current and electric field. The SSMR manifests experimentally as a distinct phase shift in the angular magnetoresistance profile, providing a purely all-electrical fingerprint for mapping the hidden Néel vector orientation of the altermagnet, without recourse to neutron or X-ray synchrotron facilities [91]. The robust, scattering-dependent temperature profile of the SSMR signal confirms the existence of a long-range, macroscopic, non-relativistic spin-split quantum state in the strained thin films, offering direct experimental validation of the band-structure predictions central to the altermagnetic hypothesis.

These potential applications, summarized in Tab. 3.1, represent the significant prize at stake. However, they are largely predicated on the existence and stability of the altermagnetic phase. Therefore, a primary goal of this thesis is to provide a definitive computational benchmark from first principles: what are the spin-dependent ballistic transport properties of a defect-free (epitaxially grown) altermagnetic RuO₂ junction? The answer would potentially provide a target for experimentalists towards fulfilling the long RuO₂ promise

for spintronic applications.

Application	Description	Key Reference
SOT-MRAM	RuO ₂ generates unconventional out-of-plane spin torques for efficient, field-free switching of perpendicular magnetic bits, potentially enabling high-density, low-power memory.	[20]
Altermagnetic Tunnel Junctions	The spin-split bands act as spin filters, promising giant TMR in an MTJ that is immune to external magnetic fields, ideal for robust memory elements.	[18]
Spin Hall Transistors	The efficient generation of pure spin currents via the altermagnetic spin-splitter effect could form the basis for ultra-low-power logic devices.	[17]
Terahertz Spintronics	The intrinsically ultrafast (THz) dynamics of the AFM order could be harnessed for high-speed data processing and communication.	[9]

Table 3.1. Summary of potential spintronic applications enabled by the unique properties of the altermagnetic phase of RuO₂. All applications are contingent on the stable realization of its magnetic order.

This detailed exploration of the RuO₂ controversy provides the essential context for

the original research presented afterwards. The following section will detail our own computational investigation aimed at characterizing the electronic and magnetic properties of RuO₂ from first principles.

— 3.4 Computational Investigation of the RuO₂ Ground State —

The conflicting experimental evidence surrounding the magnetic nature of RuO₂ underscores the need for a robust theoretical investigation. First-principles calculations allow for a systematic investigation of the electronic and magnetic ground state, but as the controversy suggests, the predictions for this fragile system are highly sensitive to the chosen methodology. This section details our original first-principles investigation into RuO₂. We begin by presenting a comprehensive overview of the computational methodology employed, providing the specific numerical parameters and justifications essential for reproducibility and rigour. We then apply this methodology to explore the stability of magnetism under standard DFT, its response to lattice strain, and the impact of advanced correlation and self-interaction corrections. Through this analysis, we aim to identify the most physically realistic model for altermagnetic RuO₂, thereby providing a reliable foundation for the quantum transport simulations in the next chapter.

3.4.1 Computational Methodology

All first-principles calculations, unless otherwise specified, were performed using the SIESTA (Spanish Initiative for Electronic Simulations with Thousands of Atoms) code [65]. SIESTA implements DFT using a basis set of strictly localized numerical atomic orbitals which allows for efficient scaling to large systems, such as the transport calculations presented later in this thesis. For comparative purposes, some strain calculations were also performed with the Vienna Ab initio Simulation Package (VASP), which uses a plane-wave basis set.

SIESTA

The calculations were performed using two standard exchange-correlation functionals: the Local Density Approximation (LDA) in the Ceperley-Alder (CA) parametrisation [51] and the Generalized Gradient Approximation (GGA) in the Perdew-Burke-Ernzerhof (PBE) form [54]. The interaction between the ionic cores and the valence electrons was described

by norm-conserving Troullier-Martins pseudopotentials [92]. To accurately account for the interaction between valence and shallow core electrons in the heavy Ruthenium atom, a semi-core pseudopotential was generated. This is a critical choice for systems with overlapping core and valence charge densities. The explicit valence electron configuration for Ru was $4s^2 4p^6 4d^7 5s^1$, treating the outer-core $4s$ and $4p$ states as valence. Oxygen was treated with a standard $2s^2 2p^4$ valence configuration. In the SIESTA calculations, the Kohn-Sham orbitals were expanded using a numerical atomic orbital basis set of double-zeta plus polarization (DZP) quality. The spatial confinement of these orbitals, a crucial parameter controlling basis set quality, was defined by a *PAO.EnergyShift* of 0.02 Ry (≈ 272 meV). This value was chosen to ensure a robust basis that accurately describes the electronic structure without being computationally prohibitive. The charge density and potentials were represented on a real-space grid. Besides, the fineness of this grid is determined by an equivalent plane-wave cutoff energy, *MeshCutoff*. A high value of 600 Ry was used for all calculations to ensure a highly accurate representation of the charge density and to eliminate any "egg-box" effects. Moreover, the Brillouin zone was sampled using a $10 \times 10 \times 10$ Monkhorst-Pack k-point mesh for all bulk calculations. This mesh density was determined to be sufficient to converge the total energy of the unit cell to within 1 meV per atom, ensuring a reliable description of the electronic ground state and its magnetic properties. For the band structure plots, electron energies were calculated along high-symmetry paths with a density of 200 k-points per segment. Finally, the electronic self-consistency cycle was iterated until the maximum difference between elements of the output and input density matrix was less than 10^{-5} . This tight tolerance ensures that the electronic ground state is fully converged. For any structural relaxations, atomic forces were relaxed to below $0.01 \text{ eV \AA}^{-1}$.

VASP Calculation Parameters

For the comparative 2D strain map shown in Fig. 3.3, calculations were performed with VASP. These calculations employed the Projector-Augmented Wave (PAW) method. The PBE functional was used with the recommended PAW pseudopotentials for Ru and O from the VASP library. A plane-wave kinetic energy cutoff (ENCUT) of 520 eV was used, and the Brillouin zone was sampled with a comparable k-point density to the SIESTA calculations.

Advanced Methods: DFT+U and ASIC

To account for strong correlation effects on the Ru 4*d* orbitals, the DFT+*U* method was employed. The specific implementation in this version of SIESTA corresponds to the simplified, rotationally invariant formalism introduced in [93]. In this approach, the total energy functional of DFT is corrected with a Hubbard term that depends only on the effective interaction parameter, $U_{\text{eff}} = U - J$. The energy correction is given by:

$$E_{\text{DFT}+U} = E_{\text{DFT}} + \frac{U_{\text{eff}}}{2} \sum_{\sigma} \text{Tr}[\mathbf{n}^{\sigma}(1 - \mathbf{n}^{\sigma})] \quad (1)$$

where E_{DFT} is the standard DFT total energy and $\mathbf{n}^{\sigma}$ is the atomic orbital occupation matrix of the localized *d*-states for spin channel σ . The implementation requires defining a separate species for the atoms receiving the +*U* correction (e.g., RuU) and using a minimal, single-zeta basis set for projecting the charge density onto the localized orbitals.

As a parameter-free alternative, the Atomic Self-Interaction Correction (ASIC) was also used in our investigation as detailed in Section 2.3, as implemented in SIESTA.

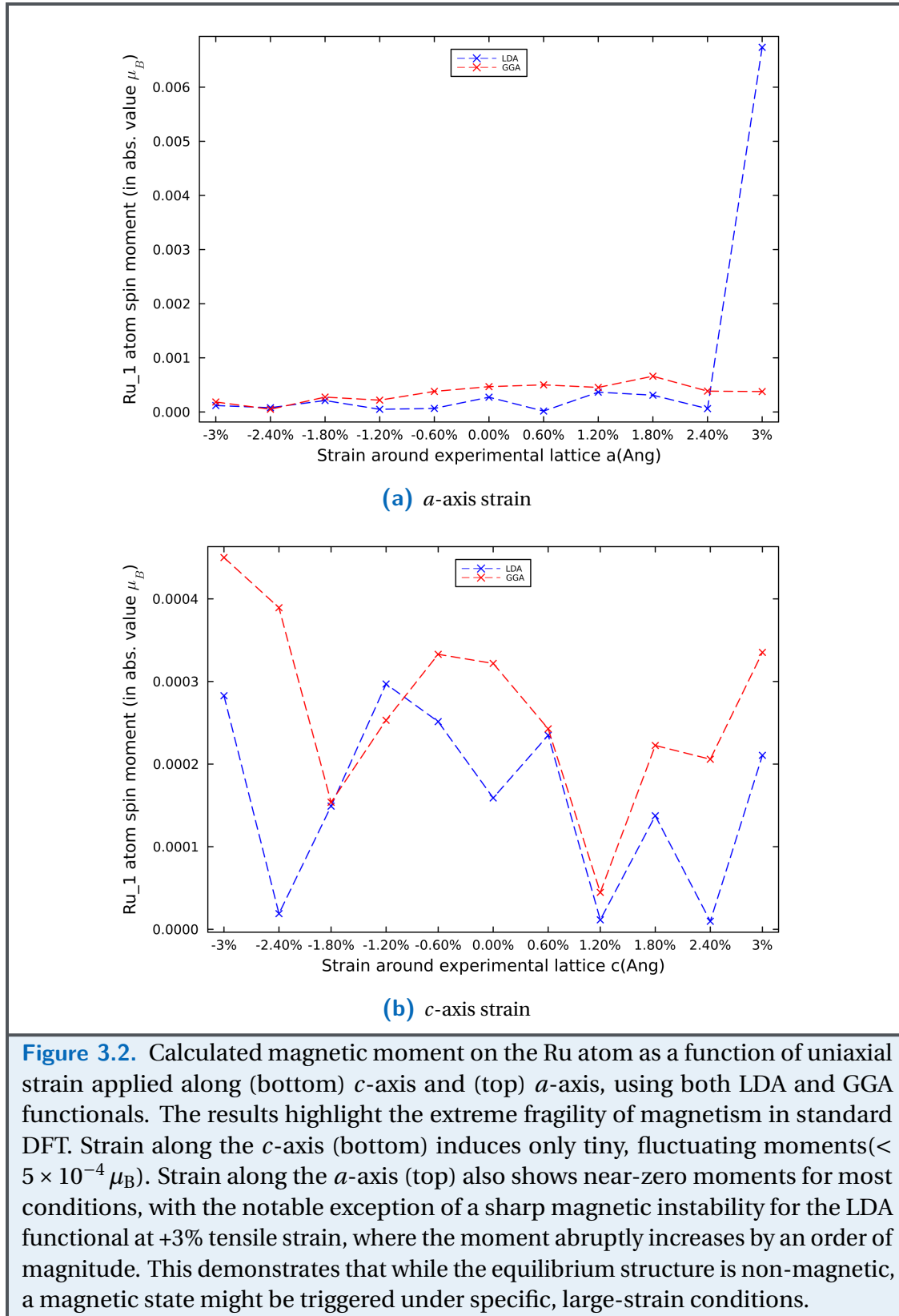
It is instructive to consider how results from more advanced exchange-correlation functionals compare to the DFT+*U* and ASIC approaches employed here. Hybrid functionals such as HSE06 would, in principle, improve the description of the Ru 4*d* electronic structure and are expected to yield larger spin splittings and more accurate magnetic moments for RuO₂ relative to GGA. However, as discussed in detail in Section 2.3.3, the non-local exchange operator intrinsic to hybrid functionals destroys the block-tridiagonal sparsity of the Hamiltonian that is essential for the NEGF transport calculations presented in Chapter 4. Consequently, hybrid functionals were not applied to the full junction geometry. Meta-GGA functionals (e.g., SCAN) are semi-local and in principle more accurate than GGA, but suffer from severe grid sensitivity in the real-space framework of SIESTA [65], as detailed in Section 2.3.3. In contrast, DFT+*U* and ASIC both preserve Hamiltonian sparsity and are therefore compatible with the NEGF formalism. The consistency between our ASIC results (presented in Section 3.4.4) and the independent DFT+*U* calculations of [18, 23] strengthens confidence that the physical picture obtained is not an artifact of a particular functional, but reflects the genuine role of correlation in determining the magnetic ground state of RuO₂.

3.4.2 Magnetic Instability in Standard DFT: The Effect of Strain

We begin by examining the magnetic stability of RuO₂ within the standard local density (LDA) [51, 55, 94, 95] and generalized gradient (GGA) approximations [53, 96, 97, 98]. These functionals often serve as a reliable baseline for systems without strong electron correlation. All calculations were performed using the SIESTA package (localised orbital basis) [65], starting from an initial antiparallel magnetic configuration to test for a stable magnetic solution. To ensure accuracy, especially for the potential delicate magnetic state, we employed pseudopotentials that include semi-core states, a choice verified to provide band structure consistent with plane wave codes like widely used DFT platform, e.g. VASP.

The Fig. 3.2 shows the magnitude of the calculated magnetic moment on a single Ru atom as a function of uniaxial strain along the a and c axes. The most striking result is the extremely small magnitude of the induced moment. For both standard LDA and GGA, the moments are on the order of $10^{-4} \mu_B$ confirming muon spectroscopy reported in Section 3.2.3. This value is nearly three orders of magnitude smaller than the $\approx 0.05 \mu_B$ reported in the initial neutron diffraction experiments in [23]. This result strongly suggests that in the framework of standard DFT, the ground state of an ideal, stoichiometric RuO₂ crystal is fundamentally non-magnetic. The tiny, fluctuating moments observed are best interpreted not as a true magnetic state, but as numerical noise or the remnant of a solution that is energetically unstable against the paramagnetic state. The erratic oscillations of the moment with infinitesimal changes in the lattice parameters reinforce this conclusion. Rather than indicating a smooth evolution of a robust magnetic state, this behaviour highlights an extreme sensitivity and fragility. A small perturbation in the lattice can enhance or completely destroy this vestigial moment, indicating that the magnetism is not

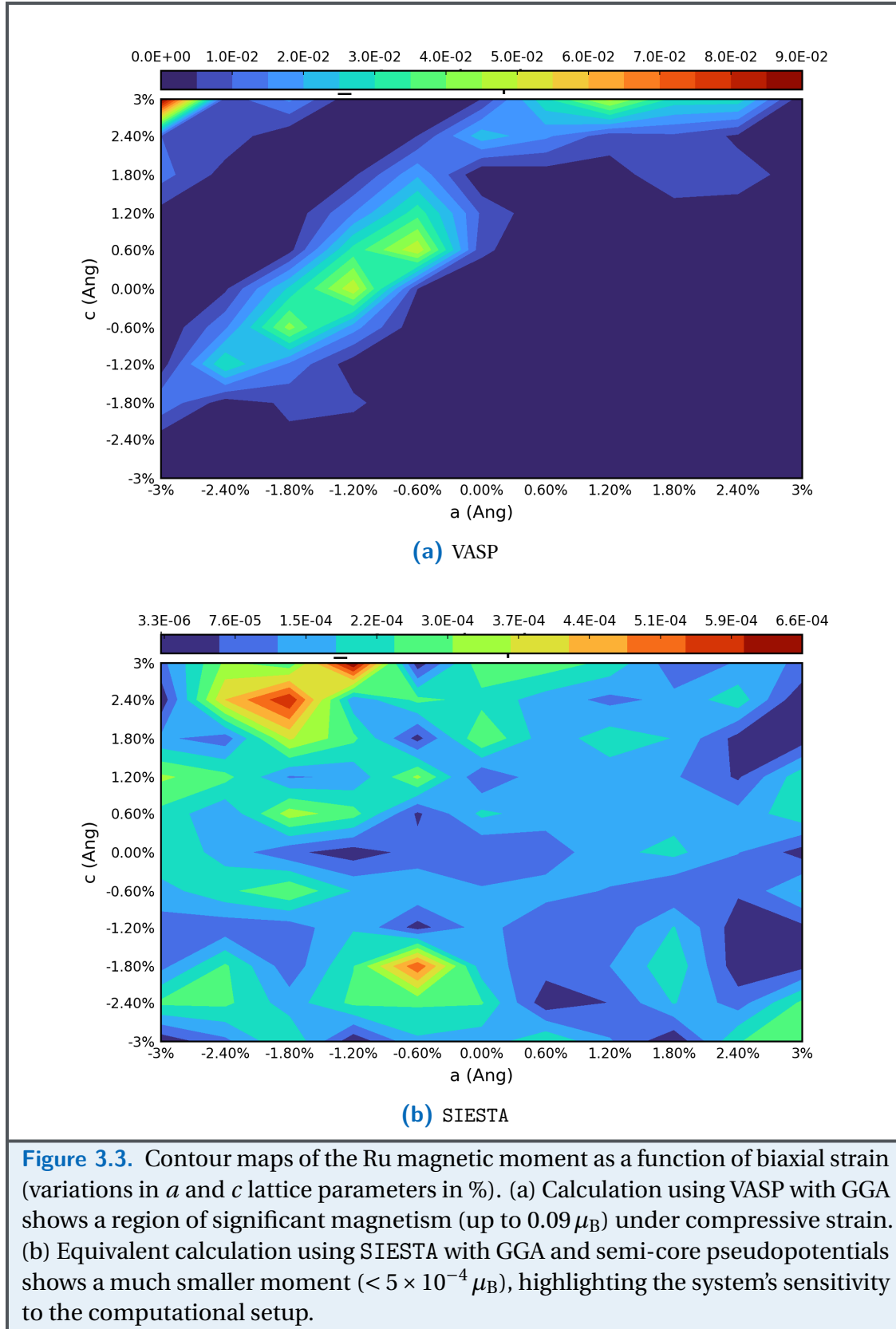
a reliable or intrinsic property of the ideal crystal under standard DFT.



This picture is further clarified by the two-dimensional strain maps shown in Fig. 3.3, which compare results from a VASP calculation using GGA with our SIESTA calculations. Here, a significant discrepancy emerges, highlighting the sensitivity of the system to the

computational implementation. The VASP-GGA calculation (Fig. 3.3) reveals a distinct "island" of stable magnetism under compressive biaxial strain, where the moment can reach values as high as $0.09 \mu_B$. This result is particularly tantalizing, as it suggests that epitaxial strain could be a viable pathway to stabilize the magnetic phase. However, it also shows that under other strain conditions, even compressive ones, the moment is quenched. In sharp contrast, our SIESTA calculations with carefully validated semi-core pseudopotentials show much smaller moments across the entire strain map (Fig. 3.3), with maximum values remaining in the $10^{-4} \mu_B$ range. This stark difference underscores the fragility of the magnetic state; its very existence in a simulation can depend on the specifics of the pseudopotential and basis set. The VASP result provides a hopeful indication that magnetism is accessible under strain, but our localised basis SIESTA results align with the

primary conclusion: for the ideal crystal, magnetism is not the inherent ground state.



3.4.3 A Survey of Computational Approaches in the Literature

The theoretical investigation of RuO₂ is marked by significant complexity with computational outcomes varying widely depending on the chosen methodology. The prediction of RuO₂'s magnetic ground state, i.e. whether it exhibits a non-magnetic, ferromagnetic, or antiferromagnetic phase (particularly the recently proposed altermagnetic phase), relies critically on the computational approach. This subsection provides a comprehensive survey of key computational studies on RuO₂ detailing their methodologies, findings, and contributions to the ongoing debate over RuO₂'s magnetic properties. By integrating seminal works, recent advancements and contrasting perspectives, this review aims to contextualize the theoretical landscape and highlights the challenges in modelling this intriguing material.

The study from 2017 we have seen in Section 3.2 and in more details in Section 3.2.2 sparked the renewed interest in RuO₂'s magnetic properties by proposing that it exhibits itinerant antiferromagnetism with a magnetic moment of $0.05 \mu_B$ [23]. Using density functional theory (DFT) with GGA and Hubbard U corrections (DFT+ U), implemented in the Vienna Ab initio Simulation Package (VASP) [99, 100, 101] with projector-augmented wave (PAW) pseudopotentials [102], they explored the electronic structure of bulk rutile RuO₂ (space group $P4_2/mnm$). Their calculations revealed that standard GGA predicts a non-magnetic, Pauli paramagnetic ground state, consistent with historical views [76]. However, introducing a modest Hubbard U of 1 – 2 eV on the Ru $4d$ orbitals stabilized a G-type antiferromagnetic order, characterized by antiparallel spin alignments on neighbouring Ru atoms. This magnetic state was accompanied by a subtle lattice distortion below approximately 900 K, with a small room-temperature magnetic moment of $M_{Ru} = 0.05 \mu_B$ per Ru atom measured by polarized neutron diffraction experiments. The DFT+ U magnetic moments estimated with $U = 1.2, 2.0$ eV are $M_{Ru} = 0.742 \mu_B, 1.183 \mu_B$. The study attributed the magnetism to a Fermi surface instability that lifts the band degeneracy imposed by the rutile crystal field, a mechanism distinct from conventional antiferromagnetism driven by strong electron correlations. This work established DFT+ U as a standard approach for modelling magnetic RuO₂, influencing subsequent studies predicting significant spintronic effects.

Building on this foundation, [18] investigated the potential of RuO₂ in spintronic applications, specifically in antiferromagnetic tunnel junctions (AFMTJs). Using DFT+ U

with $U = 2.0$ eV, implemented in the Quantum ESPRESSO code [103, 104, 105] with norm-conserving pseudopotentials, they designed a RuO₂ / TiO₂ / RuO₂ (001) AFMTJ. Their first-principles quantum transport calculations, employing the non-equilibrium Green's function (NEGF) formalism, predicted a giant TMR effect of $\sim 500\%$, driven by the momentum-dependent spin polarization of RuO₂'s non-spin-degenerate Fermi surface. The TMR arises from the relative orientation of the Néel vectors in the RuO₂ electrodes, with parallel and antiparallel configurations yielding distinct transmission probabilities. The study highlighted RuO₂'s altermagnetic properties, characterized by zero net magnetization but significant spin splitting, making it a promising candidate for high-performance spintronic devices. The use of $U = 2.0$ eV was justified by its ability to reproduce the magnetic ground state observed in experiments, aligning with [23].

Similarly, [17] explored RuO₂'s potential as an electrical spin splitter, a device that generates spin-polarized currents without net magnetization. Using ab initio calculations with DFT+ U ($U = 2.0$ eV) in the VASP code, they demonstrated that RuO₂'s anisotropically spin-split bands enable highly efficient spin-current generation at room temperature. Their calculations focused on the (001) direction, where RuO₂ exhibits spin-independent conductance but a non-spin-degenerate Fermi surface, a hallmark of altermagnetism [14]. The predicted spin-splitter effect, with spin polarization efficiencies comparable to ferromagnetic materials, underscores RuO₂'s potential for spintronic applications beyond TMR, such as spin filters and spin-orbit torque devices. The consistent use of DFT+ U across these studies has created a coherent theoretical framework supporting the altermagnetic hypothesis, with electron correlation playing a pivotal role in stabilizing the magnetic phase.

However, the reliance on the Hubbard U parameter introduces significant ambiguity, as its value is often chosen empirically rather than calculated from first principles. The choice of U (typically 1 – 2 eV for RuO₂) is critical, as standard DFT without U predicts a non-magnetic ground state [76]. This raises questions about the predictive power of DFT+ U , as the magnetic state may be an artifact of the chosen U . To address this, some studies have explored alternative computational approaches or the impact of structural and chemical modifications on RuO₂'s magnetic properties.

The fragility of RuO₂ magnetic state has been recently explored [27]. The study conducted a detailed analysis of RuO₂'s magnetic interactions using DFT calculations with

the GGA-PBE functional in VASP. Their results suggest that the magnetic ground state is exceptionally fragile, with ferromagnetic and various antiferromagnetic configurations nearly degenerate in energy, and the non-magnetic state being the most stable. They argued that quantum fluctuations, not captured in standard DFT, could suppress long-range magnetic order, providing a theoretical explanation for experimental findings from muon spin relaxation (μ SR) and quantum oscillation measurements that indicate a non-magnetic ground state [26]. Their calculations used a range of U values 0 – –3 eV to test the stability of the magnetic state, finding that only higher U values stabilize the AFM order. This further highlights and emphasizes the sensitivity to electronic structure and computational parameters.

Combining broadband infrared spectroscopy with DFT band-structure calculations in [81] has challenged the altermagnetic claims for RuO₂. Using the full-potential linearized augmented plane-wave (FLAPW) method in the WIEN2k code [106] without Hubbard U corrections, they modelled RuO₂ as a metallic, non-magnetic Fermi-liquid state. Their calculations, performed with a 500 eV plane-wave cutoff and a $12 \times 12 \times 8$ k-point mesh accurately reproduced the experimental optical conductivity. Therefore, it suggests that the electronic structure of bulk RuO₂ is best described by a non-magnetic model. Indeed, this finding aligns with historical views of RuO₂ as a Pauli paramagnet [76] and contrasts with the altermagnetic predictions. The critical role of electron correlation is emphasized in stabilizing magnetism. Their study further incorporated experimental data showing a Dirac nodal line 45 meV below the Fermi level. Founding no evidence of spin splitting in the band structure is consistent with a non-magnetic ground state. The work is very recent at the time of this manuscript but appears to provide strong computational and experimental evidence against the altermagnetic hypothesis. Overall it is clear that careful considerations are needed when choosing computational parameters when modelling RuO₂ for spintronics application.

The influence of structural factors, such as strain and defects, has also been a focal point in recent computational studies. In Ref. [107], the authors employed the full-potential linear muffin-tin orbital (FP-LMTO) method within the LDA framework to study RuO₂'s structural, electronic, and mechanical properties under pressure. Their calculations, using a $6 \times 6 \times 8$ k-point mesh and a double-zeta basis set, predicted a metallic, non-magnetic ground state for the rutile phase at ambient conditions, consistent with early experimental data [76]. However, they noted that pressure-induced phase transitions could alter the

electronic properties, suggesting structural sensitivity. More recently, a study presented in Section 3.2.4 proposed that confinement-induced strain in RuO₂ thin films could stabilize the altermagnetic phase without requiring a large Hubbard U [28]. Using DFT calculations in the Quantum ESPRESSO code with GGA-PBE functionals, they demonstrated that compressive strain modifies the electronic structure, lifting band degeneracy and inducing spin splitting. This provides a potential explanation for the discrepancy between non-magnetic bulk and magnetic thin-film experiments. The structural modifications might mimic the effects of electron correlation.

Chemical substitution offers another avenue for tuning the properties of RuO₂, complementing the insights gained from strain engineering. An experimental study investigated Cr-doped RuO₂ and observed a significant AHE [108]. Later, based on the prevailing assumption that pure RuO₂ was an altermagnet, they interpreted their results, supported by their own DFT+ U calculations, as a reconstruction of the collinear antiferromagnetic order, where the Cr dopants rotate the Néel vector and break crystal symmetries to enable the AHE. However, this interpretation has been critically re-examined in a recent theoretical study [48]. Their work provides an alternative explanation that aligns with the growing evidence for a non-magnetic ground state in pristine RuO₂. Through extensive supercell DFT calculations, the authors argue that the Cr dopants do not induce itinerant magnetism in the Ru sublattice. Instead, their findings indicate that the magnetism is highly localized on the Cr atoms themselves. Consequently, the Ru host remains essentially non-magnetic, acting merely as a polarizable medium. Besides, they show that the Cr atoms tend to form ferromagnetic clusters within the non-magnetic RuO₂ matrix. In fine, they conclude that the observed AHE does not originate from a modified altermagnetic order in RuO₂, but rather originates directly from the intrinsic anomalous Hall conductivity of these magnetic Cr clusters. This re-interpretation is pivotal, as it shifts the focus from manipulating an intrinsic altermagnetic state to understanding the behaviour of magnetic impurities within a non-magnetic host. It further highlights that phenomena initially attributed to bulk altermagnetism in RuO₂ may, in fact, be governed by extrinsic factors such as defects or, in this case, dopants. These two studies highlight the sensitivity of RuO₂'s magnetic landscape to chemical modifications, complementing the strain-based insights.

The computational predictions of altermagnetism in RuO₂ have been both supported and challenged by experimental studies as shown in Section 3.2.2. Conversely, comprehensive experimental studies using μ SR spectroscopy and neutron diffraction have found

no magnetic order in bulk RuO₂, with magnetic moments below $1.14 \times 10^{-4} \mu_B$ per Ru, significantly lower than computational predictions. Additionally, time-domain terahertz spectroscopy on RuO₂ films failed to detect antiferromagnetic spin splitting, casting further doubt on the antiferromagnetic hypothesis [109]. These discrepancies underscore the need to reconcile computational models with experimental observations, particularly regarding the role of electron correlation and structural effects.

On the theory front literature reveals several critical factors influencing RuO₂ magnetic predictions:

1. **Electron Correlation:** The stabilization of a magnetic state typically requires a Hubbard U , suggesting that magnetism in RuO₂ is not a simple one-electron phenomenon but depends on correlation effects in the Ru $4d$ orbitals.
2. **Lattice Parameters and Strain:** The magnetic state is highly sensitive to lattice parameters, with strain in thin films potentially inducing antiferromagnetism without large U .
3. **Exchange-Correlation Functional:** The choice of functional (e.g., GGA, LDA) and the inclusion of U significantly affect outcomes, with standard functionals favouring non-magnetic states.
4. **Computational Details:** Variations in k -point sampling, pseudopotentials, and convergence criteria can tip the balance between magnetic and non-magnetic states, indicating a system near a magnetic instability.

The multifaceted theoretical landscape underscores the challenges in modelling RuO₂, a material poised at the edge of a magnetic instability. The reliance on empirical U values, sensitivity to structural parameters, and discrepancies with experimental data highlight the need for alternative approaches. Our work, employing the parameter-free Atomic Self-Interaction Correction (ASIC) method [61], aims to address these challenges by minimizing reliance on tunable parameters, offering a novel perspective on the role of electron localization and self-interaction in RuO₂'s magnetic properties.

It is instructive to consider how results from more advanced exchange-correlation functionals compare to the DFT+ U and ASIC approaches discussed here. Hybrid functionals such as HSE06, which incorporate a fraction of screened non-local Hartree-Fock exchange,

generally provide more accurate band gaps and magnetic moments for transition metal oxides. For RuO₂, one would expect hybrid calculations to open a larger gap in the minority-spin channel and to yield a more accurate spin splitting relative to ARPES data. However, as detailed in Section 2.3.3, hybrid functionals destroy the block-tridiagonal sparsity of the Hamiltonian in a localized orbital basis, making them computationally intractable for NEGF-DFT transport calculations of the size required here. Meta-GGA functionals (e.g., SCAN) are another promising direction, satisfying more exact constraints than GGA while remaining semi-local; yet in real-space codes like SIESTA, they suffer from severe grid sensitivity that currently precludes their routine use in large transport supercells [65]. The DFT+ U approach represents the standard compromise in the literature: it is computationally efficient and effective at stabilizing magnetic order, but is reliant on a material-specific parameter U whose value affects both the magnetic moment magnitude and the band-gap opening. The ASIC method adopted in this work occupies a complementary niche: it is parameter-free and purely local, making it compatible with the NEGF framework while providing a physically motivated alternative to the empirical U . The consistency between our ASIC results and the DFT+ U predictions of [18, 23] strengthens confidence in both approaches and demonstrates that the key physics is robust to the specific choice of correlation correction.

3.4.4 Inducing Magnetism: The Role of Electron Correlation and Self-Interaction Correction

Given that standard DFT fails to stabilize a robust magnetic moment, we must consider more advanced methods that account for physical effects beyond the LDA/GGA level. Two such approaches are the inclusion of an on-site Hubbard U term (DFT+ U) to treat strong electron correlation, and the use of a self-interaction correction (SIC) to remedy the inherent delocalisation error of standard functionals.

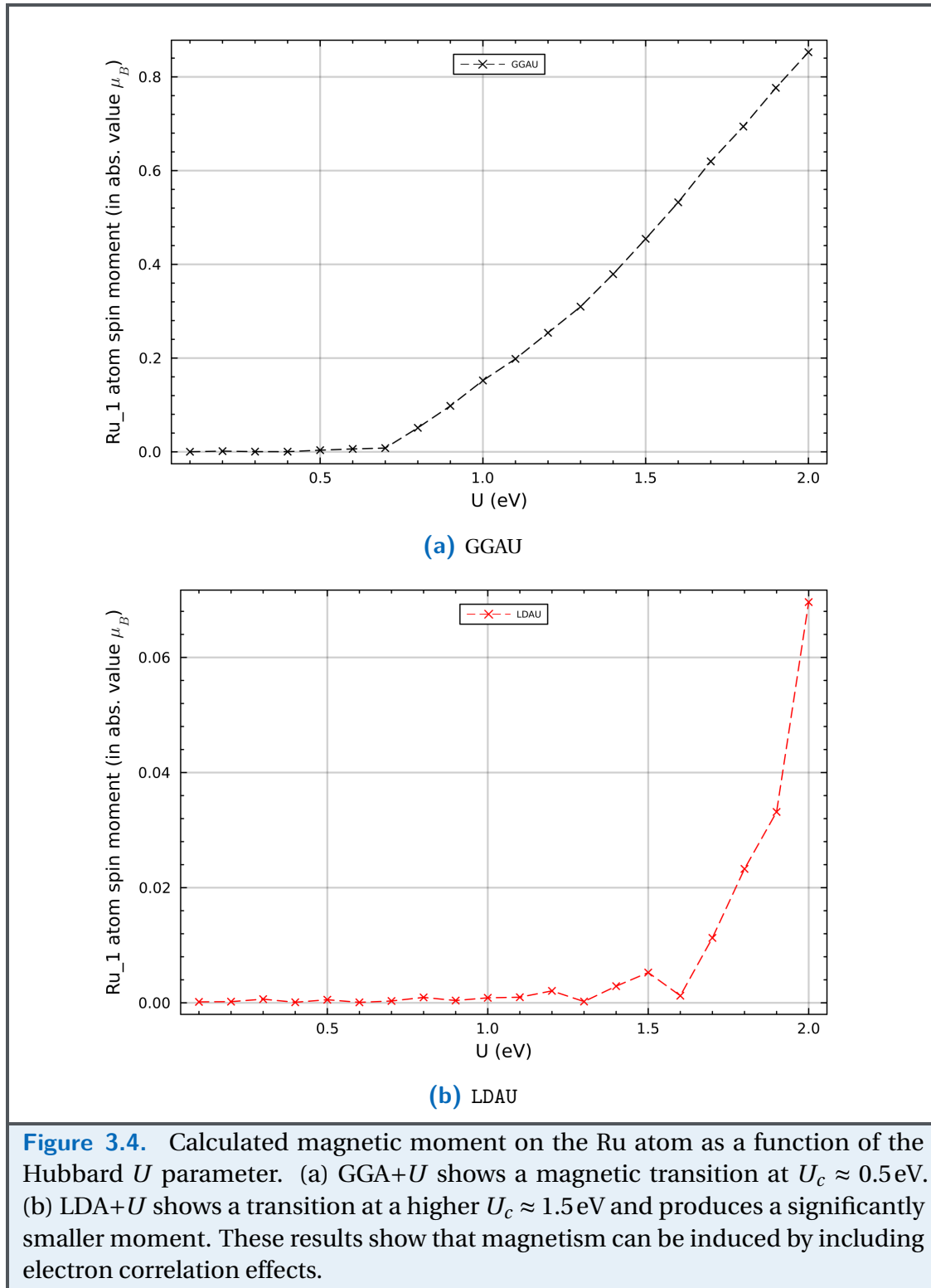
DFT+ U : A Parameter-Dependent Pathway to Magnetism

The DFT+ U method presented in Section 2.3.1 is widely used to improve the description of localized d or f electrons in transition metal oxides. By adding a penalty term (U) for non-integer orbital occupation, it can correctly capture insulating gaps and stabilize magnetic order where standard DFT fails. We investigated the effect of U on the Ru $4d$ orbitals, with the results shown in Fig. 3.4. A classic Stoner-like instability is observed: the magnetic moment is zero for small U and emerges only above a critical value, U_c . In our

GGA+ U calculation in Fig. 3.4, this transition occurs around $U \approx 0.5$ eV after which the moment grows steadily reaching $\approx 0.85 \mu_B$ at $U = 2.0$ eV. The LDA+ U calculation shows a similar trend but requires a larger critical U of ≈ 1.5 eV and results in a much smaller moment of $\approx 0.07 \mu_B$ at $U = 2.0$ eV.

These results are crucial for interpreting the experimental controversy. They demonstrate that a moment of the order reported in [23] ($\approx 0.05 \mu_B$) can indeed be stabilized, but only if one assumes the presence of significant electron correlation effects, parametrised by a U of approximately 1.5 – 2.0 eV for GGA. While such values are considered physically reasonable for 4d transition metals, the very necessity of invoking a tunable parameter like U reinforces the idea that magnetism in RuO₂ is not an intrinsic feature of its one-electron band structure but rather an emergent phenomenon driven by correlation. From a numerical precision standpoint, the individual moment values at each U carry an uncertainty of $\approx \pm 0.01 \mu_B$, arising from k -point discretization and basis-set completeness (see Section 2.5); the dominant source of quantitative uncertainty in the DFT+ U framework is therefore parametric rather than numerical, with the choice of U introducing a systematic

variation of order $0.2 \mu_B$ per eV in the altermagnetic regime.



ASIC: A Parameter-Free Correction for the Altermagnetic Band Structure

As an alternative to the parametric DFT+ U method, we employ the atomic self-interaction correction (ASIC) presented in Section 2.3.2 based on Ref. [61]. This method provides a more fundamental correction, aiming to cancel the unphysical interaction of an electron

with itself, a known deficiency of LDA and GGA which particularly affects localized states like the Ru $4d$ orbitals. This study employs the LDA with ASIC, a method that accurately reproduces experimental band gaps in ionic semiconductors and is well-suited for quantum transport simulations [61].

A central requirement for any spintronic application is that the theory must predict a significant spin polarisation; a bonus would be the spin-splitting of the band structure predicted by altermagnetism theory. We therefore investigated the band structure of RuO₂ under several computational configurations, as shown in Fig. 3.6. The key observation is that the nature of the spin-splitting is highly dependent on how the ASIC is applied. While standard LDA (initialized with a magnetic moment and antiferromagnetic spin arrangement) shows some splitting, it does not conform to the specific topology predicted for altermagnetism, particularly around the Fermi level. The crucial finding of our investigation is that the necessary spin-splitting signature of altermagnetism, namely large splitting between the $M \rightarrow \Gamma$ and $A \rightarrow Z$ high-symmetry points at Fermi level, is most accurately reproduced only when the ASIC is applied selectively to the Ruthenium atoms while the Oxygen atoms are treated with standard LDA. This physically motivated approach recognizes that the self-interaction error is most pronounced for the localized $4d$ electrons of the Ru cation. The resulting band structure (Fig. 3.6d) aligns well with the theoretical predictions for d -wave altermagnetism and is consistent with recent experimental ARPES measurements presented in Section 3.2. This agreement gives us confidence that the LDA-ASIC (Ru-only, the full ASIC seems to over-correct towards opening a gap) method provides the closest match to GGA+ U method for studying the quantum transport properties of a hypothetical altermagnetic RuO₂ phase. Moreover, it's also mimicking the effect of GGA+ U applied by previous computational studies reported in Section 3.4.3 on the same material and reported in Fig. 3.5. This model will therefore be used for the device simulations presented in the following chapter. The Ru magnetic moment predicted by LDA-ASIC ($\lambda = 0.5$, applied to Ru only) is $0.987 \mu_B$, with a numerical precision of $\approx \pm 0.01 \mu_B$

from the k -mesh and basis-set convergence criteria documented in Section 2.5.

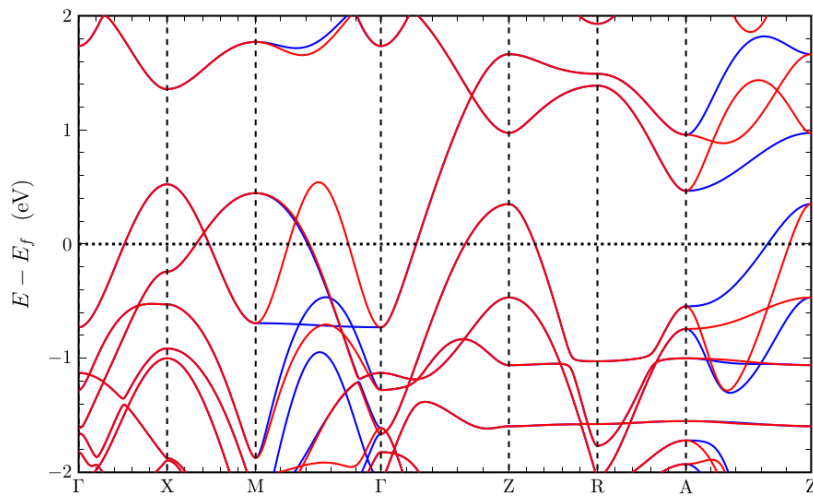
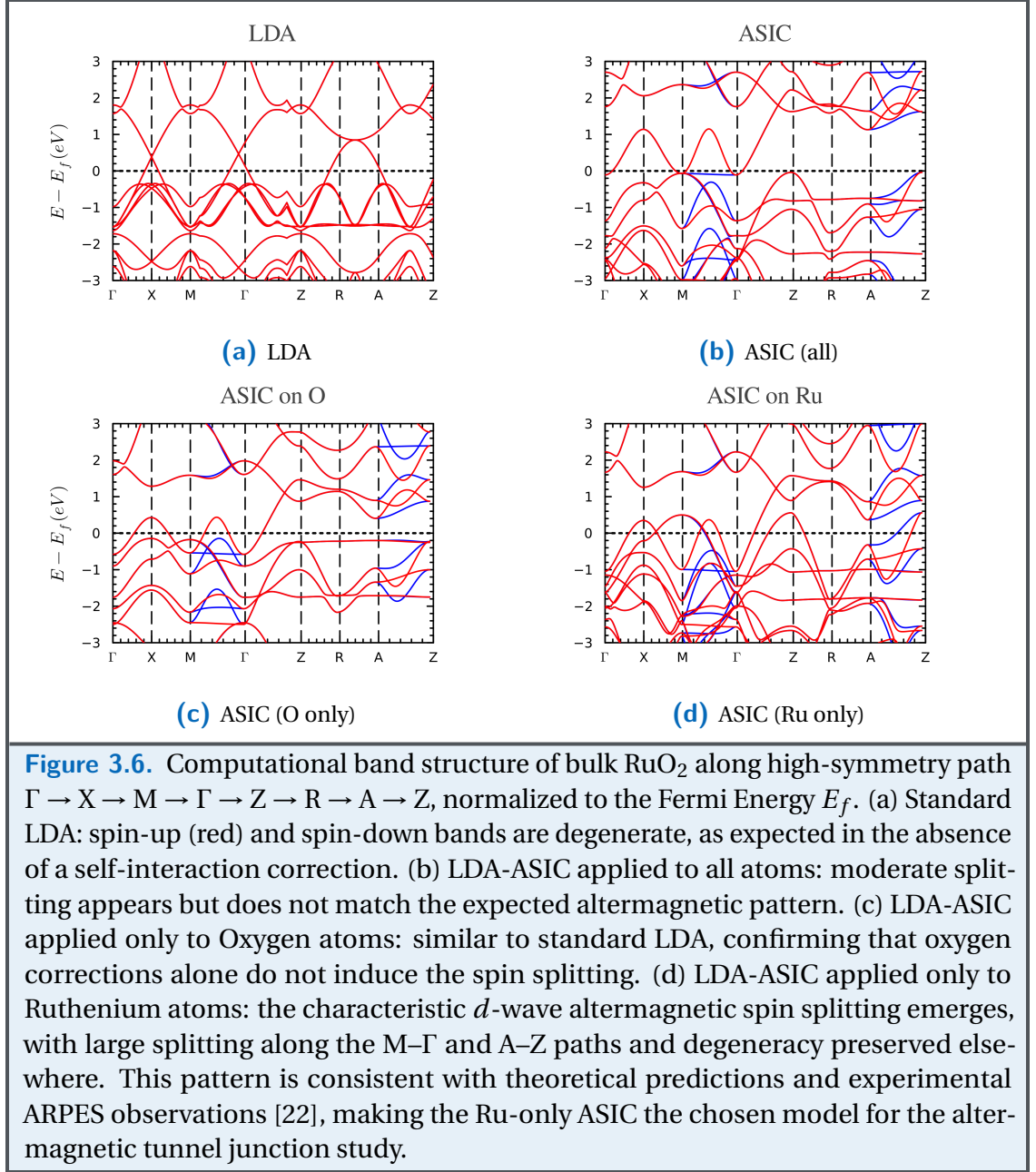


Figure 3.5. Electronic band structure of bulk RuO₂ along the high-symmetry path $\Gamma \rightarrow X \rightarrow M \rightarrow \Gamma \rightarrow Z \rightarrow R \rightarrow A \rightarrow Z$, computed with GGA+ U ($U = 2.0$ eV) and referenced to the Fermi level E_F . This calculation reproduces the benchmark result of Ref. [18] and illustrates the characteristic d -wave antiferromagnetic spin splitting: spin-up (blue) and spin-down (red) bands are strongly split along the $M \rightarrow \Gamma$ and $A \rightarrow Z$ paths while remaining nearly degenerate along $\Gamma \rightarrow X$ and $Z \rightarrow R$. This serves as the qualitative reference against which the ASIC band structure in Fig. 3.6 is evaluated.



3.5 Hall Effects as a Probe of the RuO₂ Ground State

The experimental observation of a large Anomalous Hall Effect (AHE) in RuO₂ thin films [19] stands as one of the most compelling pieces of evidence for its potential altermagnetism. In conventional ferromagnets, the AHE is proportional to the net magnetization. Its appearance in a compensated magnet like RuO₂ is therefore highly non-trivial and is theoretically permitted by the unique symmetries of the predicted altermagnetic phase,

which break crystal time-reversal symmetry [43]. The origin of this effect, however, remains intrinsically linked to the central controversy over the material's magnetic ground state.

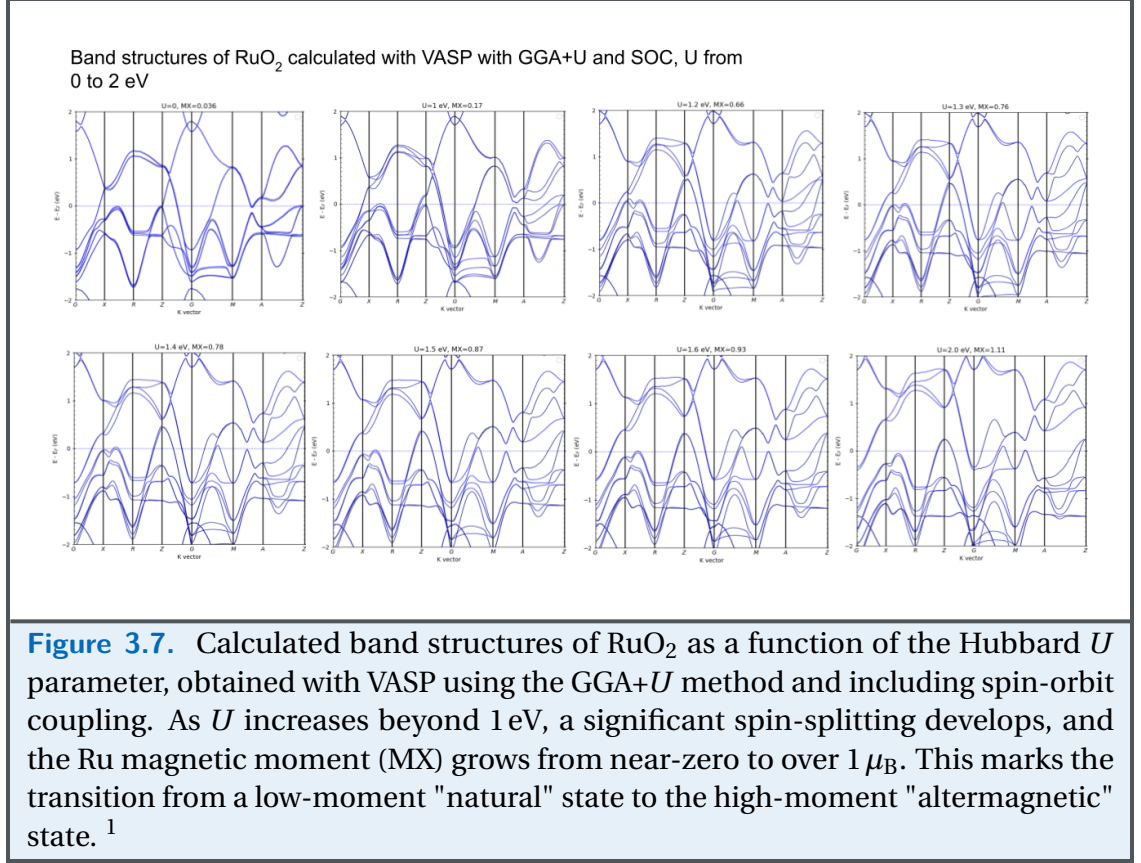
In this section, we employ first-principles calculations to investigate the intrinsic Hall conductivities of RuO₂, addressing the crucial question: under what conditions can a significant Anomalous Hall Conductivity (AHC) arise? To do this, we compute both the AHC and the Spin Hall Conductivity (SHC) from a unified theoretical framework. The calculations rely on the construction of a maximally localized Wannier function basis from the underlying Bloch states, a powerful and widely-used method for accurately evaluating Berry curvature-related transport phenomena in the Brillouin zone [110, 111]. By using the SHC calculation to validate our computational approach against established literature [88], we can provide a robust theoretical verdict on the link between the magnetic state of RuO₂ and its anomalous Hall response, offering a direct bridge between our computational models and experimental reality. A comprehensive review of the physics of spin Hall effects can be seen in [112].

3.5.1 The Anomalous Hall Conductivity: A Signature Dependent on Magnetic Order

Our primary investigation focuses on how the AHC depends on the magnetic state of the RuO₂ crystal. As established in Section 3.4.4, a magnetic moment is not a stable feature of standard DFT but can be induced by including electron correlation effects via a Hubbard U parameter. This allows us to computationally model the two distinct regimes at the heart of the controversy: a "natural" low-moment state consistent with experiments on pristine crystals, and an "altermagnetic" high-moment state that represents the theoretically predicted phase.

The transition between these states is illustrated in Fig. 3.7, which shows the calculated band structures and corresponding Ru magnetic moments (MX) as a function of U . For $U < 1.0$ eV, the moment remains small ($< 0.1 \mu_B$). However, above this threshold, a Stoner-like instability drives the system into a high-moment state, reaching over $1 \mu_B$ at $U = 2.0$ eV. This high-moment state is characterized by a large spin-splitting of the bands, consistent

with the altermagnetic picture.



With these two regimes clearly defined, we present the central result of this investigation in Fig. 3.8. The figure displays the calculated AHC as a function of energy for several magnetic moments, corresponding to different values of U . The conclusion is unambiguous. In the low-moment states ($MX < 0.1 \mu_B$, corresponding to $U < 1.0$ eV), the AHC is practically zero at the Fermi energy ($E - E_F = 0$). A significant AHC only emerges in the high-moment altermagnetic phase ($MX > 0.5 \mu_B$), where it develops a pronounced peak structure around the Fermi level.

This result provides a powerful theoretical explanation for the experimental dichotomy. The absence of AHC in the low-moment state is consistent with the lack of magnetism observed in pristine bulk crystals. Conversely, the large AHC predicted for the high-moment altermagnetic state aligns perfectly with the experimental AHE reported in thin films [19], where factors like epitaxial strain or defects may be responsible for stabilizing the required magnetic order. Our calculations therefore establish a direct theoretical link: the anomalous Hall effect in RuO₂ is not an intrinsic property of its non-magnetic state but appears to be a direct consequence and signature of the altermagnetic phase. This finding

¹Figure courtesy of Dr. Maria Stamenova.

is conceptually similar to recent work on Cr-doped RuO₂, where the AHE is also shown to arise from a symmetry breaking induced by the magnetic configuration [48].

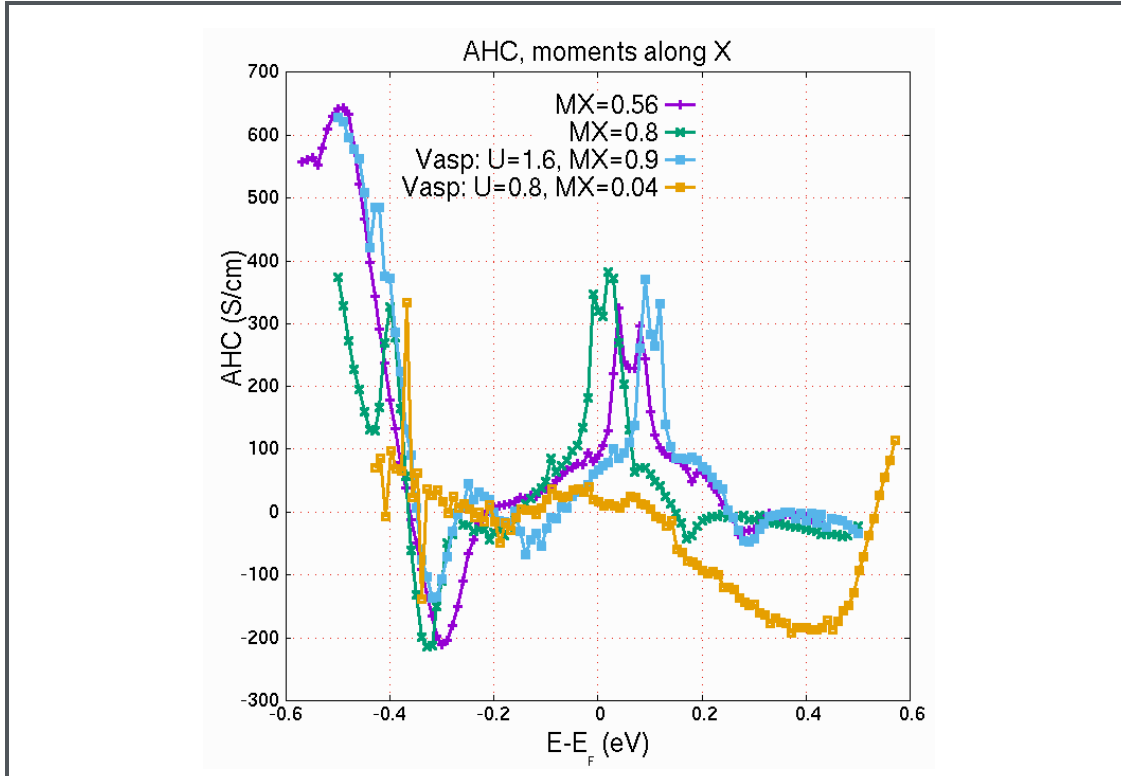


Figure 3.8. Calculated Anomalous Hall Conductivity (AHC) as a function of energy for different Ru magnetic moments stabilized by varying U or applying constraints. In the low-moment states (e.g., VASP $U = 0.8$, $MX = 0.04 \mu_B$), the AHC is negligible at the Fermi energy. A significant AHC only emerges in the high-moment altermagnetic phase (e.g., $MX = 0.56 \mu_B$, $0.8 \mu_B$, $0.9 \mu_B$). This result provides a direct theoretical link between the altermagnetic order of RuO₂ and the experimentally observed anomalous Hall effect reported in thin-film experiments [19].²

3.5.2 Computational Challenges: The Sensitivity of Wannier Interpolation

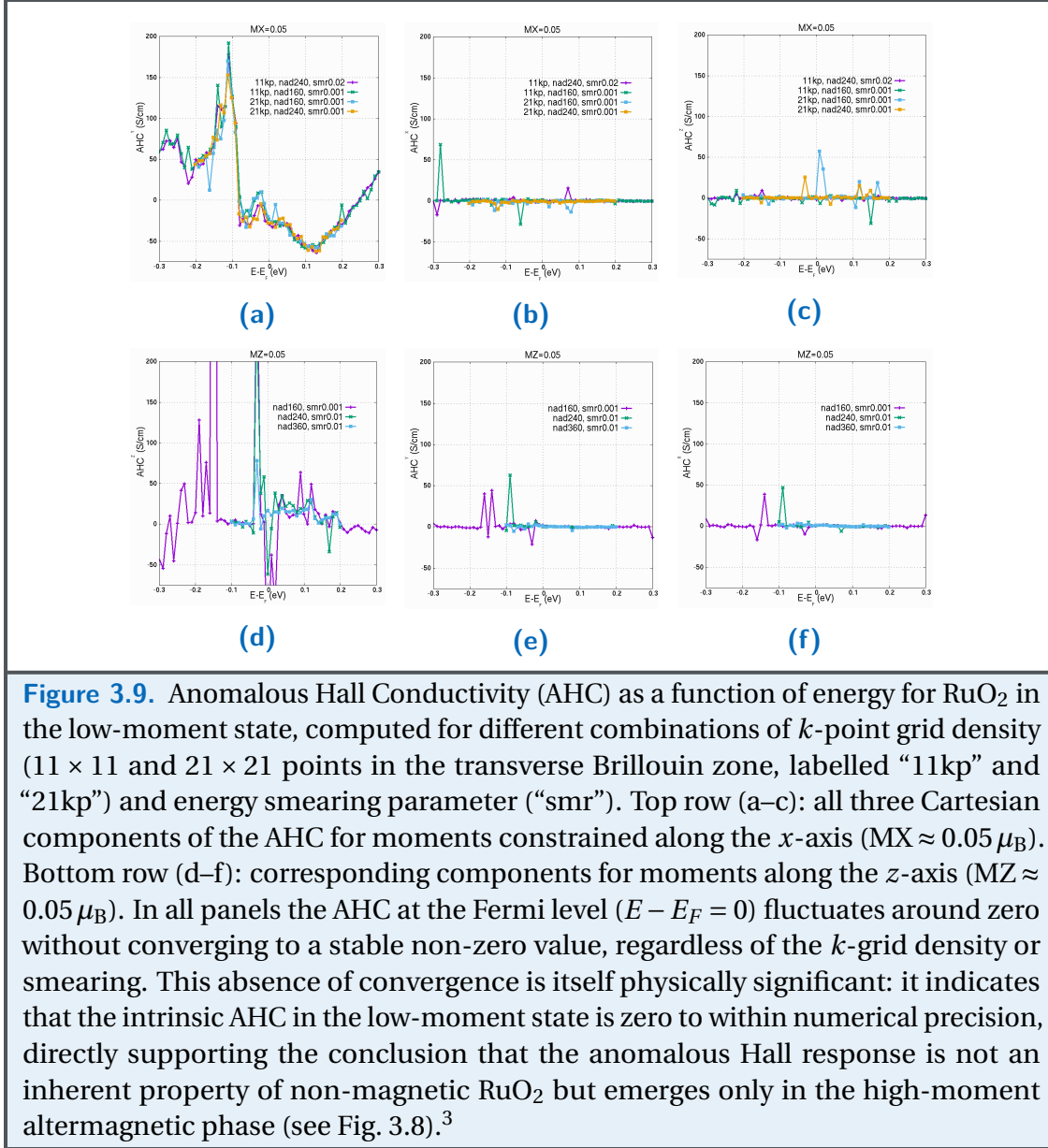
To provide full transparency and context for the results presented above, it is essential to address the computational complexities inherent in calculating Hall conductivities. The intrinsic AHC is determined by integrating the Berry curvature of the occupied bands over the entire Brillouin zone. Performing this integration accurately requires an exceptionally dense k -point grid, often orders of magnitude denser than what is feasible for a direct self-consistent DFT calculation. The standard and most powerful technique to overcome this limitation is to first construct a set of maximally localized Wannier functions (MLWFs) that perfectly reproduce the DFT band structure within a chosen energy window. This compact, analytic basis is then used to perform the Berry curvature integration on a virtually limitless k -point mesh [110, 111].

²Figure courtesy of Dr. Maria Stamenova.

However, the process of generating a high-fidelity MLWF representation is not always straightforward and can be highly sensitive to the input parameters. The quality of the Wannier interpolation depends on factors such as the number of bands included, the initial projections chosen for the localization procedure, and the energy windows used for disentanglement. Our investigation revealed that the AHC calculation in RuO₂ is a prime example of this sensitivity, but in a way that provides further physical insight.

The challenge is most pronounced in the "natural" low-moment state. As shown in Fig. 3.9, the calculated AHC in this regime is highly unstable with respect to the density of the underlying DFT k -point grid and the energy smearing used. While all calculations show a value fluctuating around zero at the Fermi level, there is no smooth convergence to a single, definitive value. This numerical instability is, in itself, an important result for the characterization of RuO₂. If the low-moment state possessed a robust, intrinsic AHC, the calculation would be expected to converge cleanly. The observed fluctuations are therefore best interpreted as numerical noise around a true physical value of zero, reinforcing the

conclusion that there is no intrinsic AHC in this state.



Even in the high-moment "altermagnetic" state, where the AHC is large, the quantitative result depends on the specifics of the Wannierisation. Figure 3.10 illustrates this by comparing three different Wannier interpolations for the same physical system (GGA+ U with $U = 1.6$ eV). Although all three attempts produce a good fit to the DFT bands, they yield quantitatively different AHC spectra. This highlights the existence of an inherent numerical uncertainty in the calculation. However, the crucial physical observation is that despite these quantitative variations, all three calculations consistently predict a large, non-zero AHC with a qualitatively similar shape. The physical signal is robust, even if its precise magnitude is subject to the details of the interpolation. This stands in sharp

³Figure courtesy of Dr. Maria Stamenova.

contrast to the low-moment case, where the signal itself is unstable. This detailed analysis underscores the care that must be taken in such calculations and strengthens our primary conclusion: a robust AHC is a feature of the altermagnetic phase only.

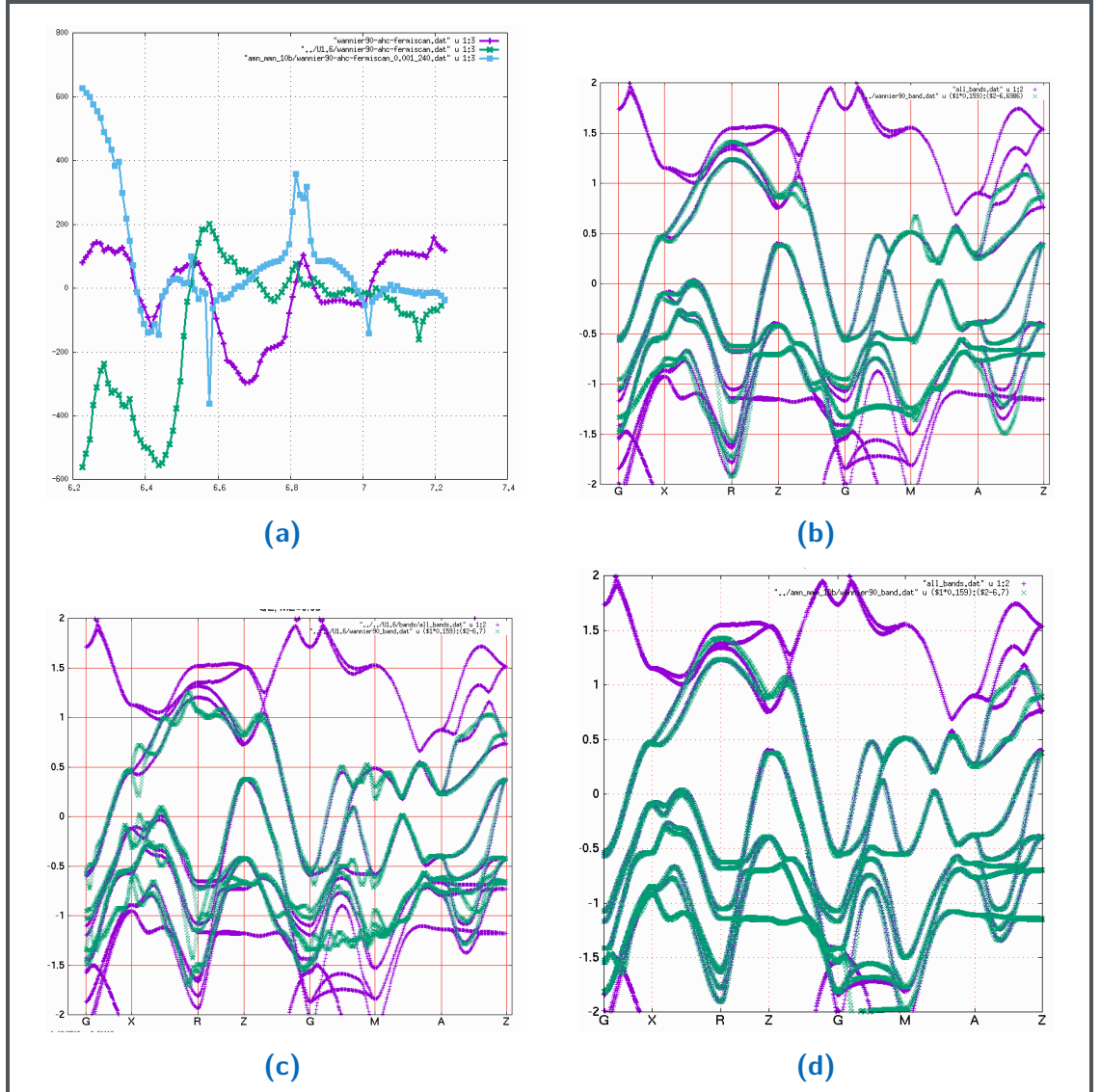


Figure 3.10. Sensitivity of the calculated Anomalous Hall Conductivity (AHC) to the choice of Wannier interpolation procedure, for RuO₂ in the high-moment altermagnetic state (GGA+ U , $U = 1.6$ eV, $M_X \approx 0.9 \mu_B$). (a) AHC versus energy for three independent Wannier constructions, each using 10 maximally localized Wannier functions (MLWFs) but with different initial projections and disentangling energy windows. (b–d) Quality of each corresponding Wannier fit: interpolated bands (green) overlaid on the DFT reference bands (purple). Although the quantitative AHC values differ between interpolations, all three consistently predict a large non-zero AHC centred near the Fermi level. This robustness of the physical signal, despite the procedural sensitivity, underpins the reliability of the central conclusion: a large anomalous Hall response is an intrinsic feature of the altermagnetic high-moment phase of RuO₂.⁴

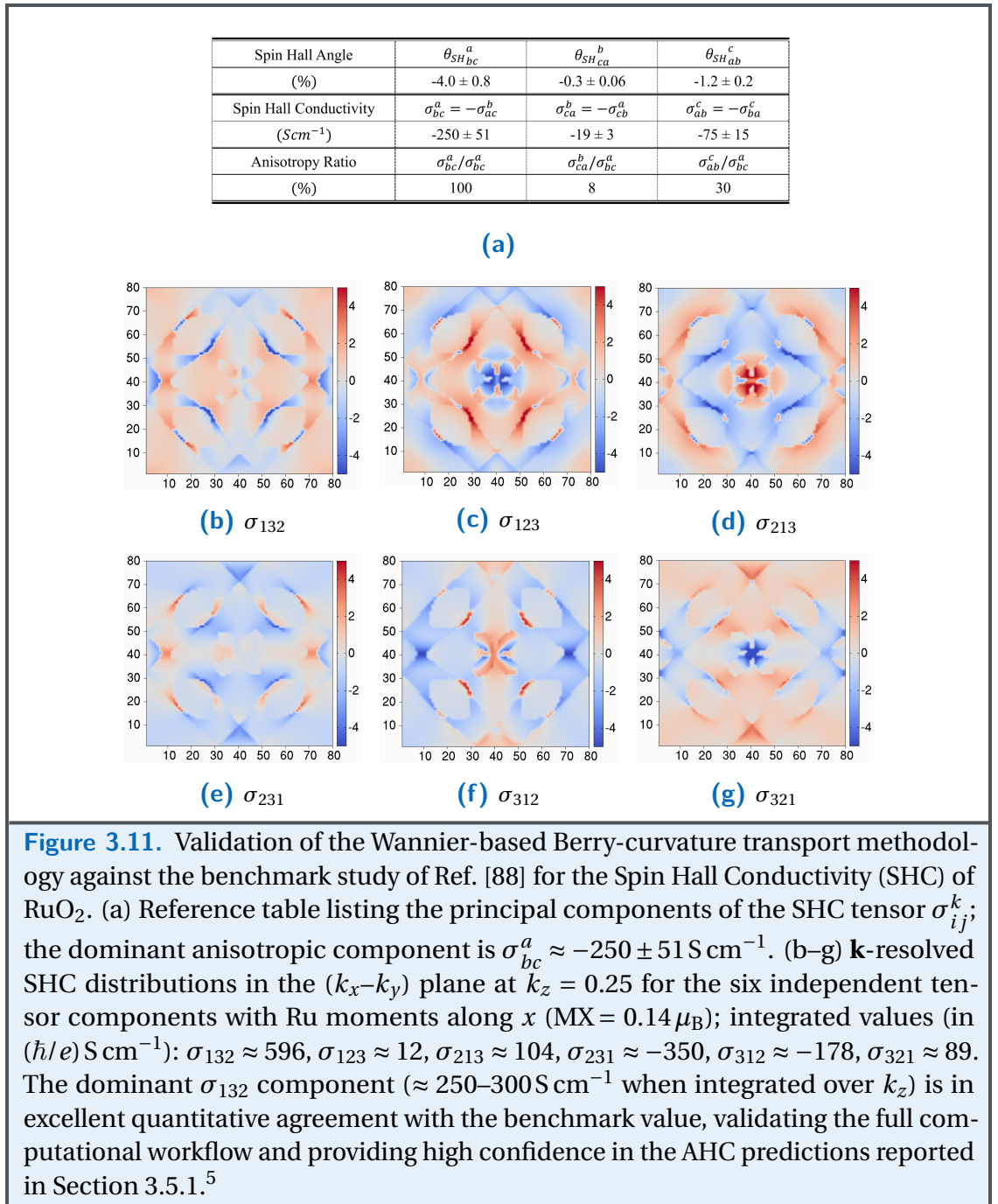
⁴Figure courtesy of Dr. Maria Stamenova.

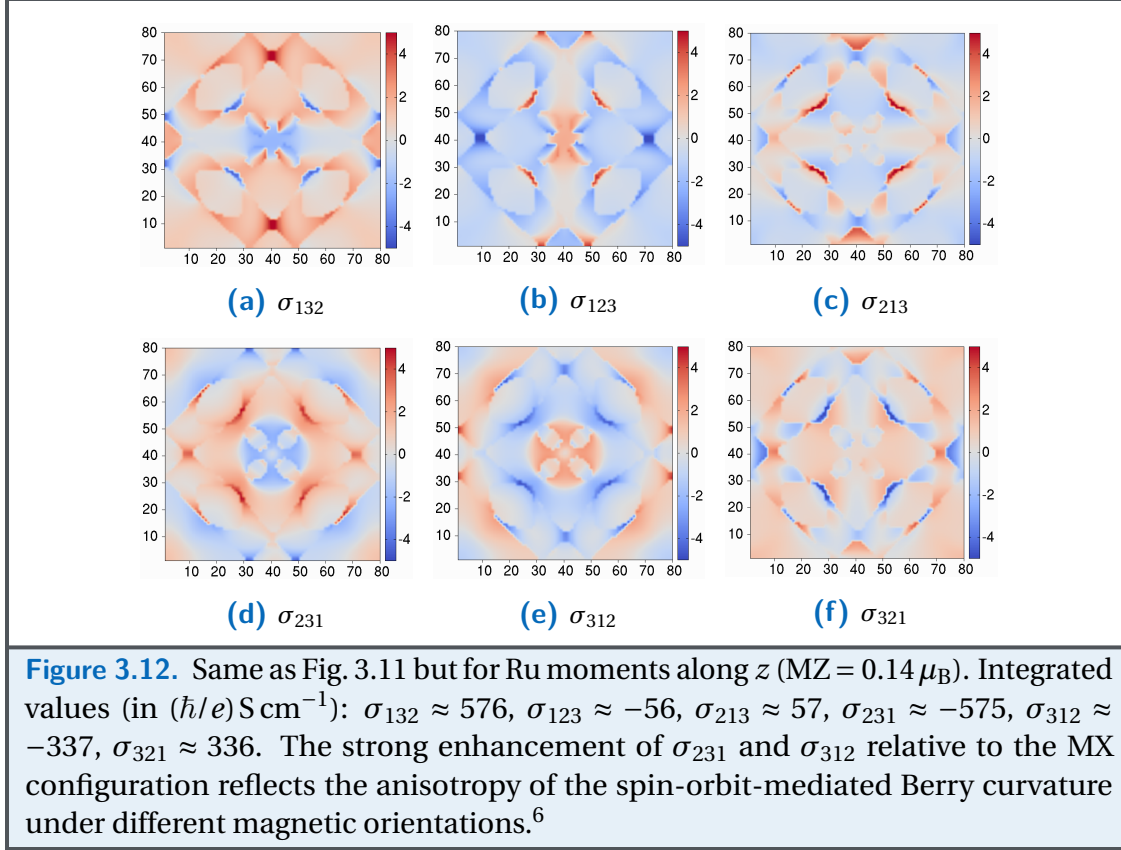
3.5.3 Methodological Validation: The Spin Hall Conductivity

Given the computational sensitivities discussed in the previous subsection, a critical validation of our Wannier-based methodology is necessary to substantiate our claims regarding the AHC. We achieve this by calculating the intrinsic Spin Hall Conductivity (SHC). Unlike the AHC, the SHC is symmetry-allowed in the centrosymmetric, non-magnetic phase of RuO₂ and is known to be significant due to the strong spin-orbit coupling of the Ruthenium atom [112]. This provides an ideal, non-zero benchmark against which to test the accuracy of our method. We compare our results directly with established theoretical and experimental work [88] which provides a robust characterization of the anisotropic Spin Hall Effect in rutile RuO₂.

Our first-principles calculations of the full SHC tensor successfully reproduce the key findings of this reference work with excellent quantitative agreement. This is demonstrated in Figs. 3.11 and 3.12. These figures present the benchmark table from Wang et al. alongside our own calculations of the k -resolved SHC components in the Brillouin zone. By integrating our calculated k -space distributions, we can extract the total conductivity values for each component. Our calculations yield a value for the dominant anisotropic component, σ_{bc}^a , of approximately $250 - 300 \text{ Scm}^{-1}$, which is in excellent quantitative agreement with the reported benchmark value of $-250 \pm 51 \text{ Scm}^{-1}$ (the sign difference is a matter of convention). The other, smaller components are also found to be of the correct order of magnitude, confirming that our method accurately captures both the scale and

the strong anisotropy of the Spin Hall Effect in this material.



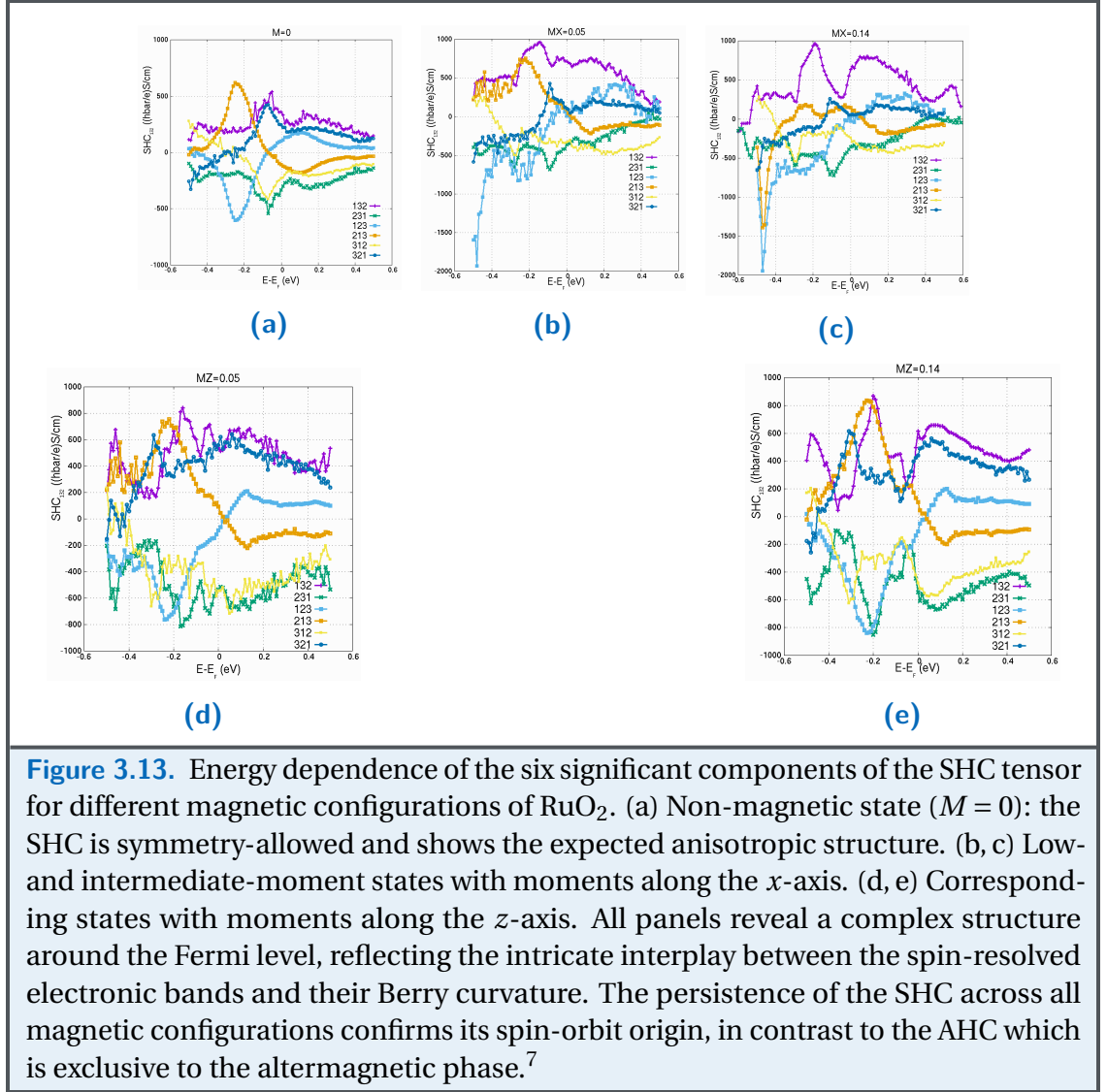


Beyond the integrated values at the Fermi level, our calculations also capture the rich energy dependence of the SHC tensor, as shown in Fig. 3.13. The various components exhibit complex structures around the Fermi level, reflecting the intricate details of the underlying spin-resolved electronic bands and their Berry curvature. The ability to reproduce this behaviour provides further evidence of the high fidelity of our computational

⁵Figure courtesy of Dr. Maria Stamenova.

⁶Figure courtesy of Dr. Maria Stamenova.

model.



The agreement demonstrated in Fig. 3.11 is the key finding of this subsection. It serves as a "sanity check" confirming that our computational workflow, from the underlying DFT calculations to the delicate Wannier interpolation and the final Berry curvature integration, is capable of accurately capturing the complex transport physics of RuO₂. This successful validation of our methodology provides high confidence in the central conclusion of this section: that the calculated AHC presented in Section 3.5.1 is a reliable physical prediction. The finding that a robust AHC is exclusively a feature of the high-moment state is therefore not a computational artifact, but reflects a genuine physical link between the altermagnetic order in RuO₂ and its anomalous Hall effect.

⁷Figure courtesy of Dr. Maria Stamenova.

4 Design and Quantum Transport in Altermagnetic Tunnel Junctions

Having established the theoretical foundations of altermagnetism in Chapter 1 and explored the complex and controversial case of its prototypical material RuO_2 for spintronics applications in Chapter 3, we now turn to the central application that motivates much of this research: the prospect of a novel altermagnetic tunnel junction (ALMTJ). The unique electronic structure of the proposed altermagnets that possesses a spin-split band structure without a net magnetic moment presents a unique opportunity to engineer spintronic devices. Combining the best attributes of both ferromagnets and antiferromagnets could be realisable in such devices. An ALMTJ, in principle, could offer the large magnetoresistive effects necessary for data readout while retaining the intrinsic advantages of compensated magnets, such as robustness against external fields and ultrafast dynamics.

This chapter addresses two fundamental questions that bridge the gap between theoretical concept and practical implementation. First, how can we move beyond single-material case studies to systematically design novel, experimentally feasible and viable ALMTJs from the vast library of known materials? We will develop and apply a set of physically grounded "golden rules" for this purpose. Second, what are the intrinsic quantum transport properties of a prototypical ALMTJ? What new physics emerges, beyond linear response conduction, under the application of a finite bias voltage? By performing a deep, first-principles-based dive into the $\text{RuO}_2 / \text{TiO}_2 / \text{RuO}_2$ system, where TiO_2 is chosen as a barrier because of lattice match and reasonable band gap of 3.1 eV, we aim to provide a theoretical benchmark for the performance of such devices and uncover intriguing non-equilibrium phenomena, including a giant inversion of the tunnelling

magnetoresistance.

4.1 The Promise of Altermagnetic Tunnelling

The cornerstone of modern spintronic memory and sensor technology is the magnetic tunnel junction (MTJ), where the tunnelling magnetoresistance (TMR) effect provides a robust method for reading a magnetic state. In a conventional ferromagnetic MTJ, the TMR is fundamentally tied to the presence of a global net spin polarization in the density of states at the Fermi level. The discovery of altermagnetism opens a pathway to realize a TMR effect in a device with zero net magnetization, based on a profoundly different physical mechanism. In an altermagnetic tunnel junction (ALMTJ), the electrodes are not ferromagnetic but altermagnetic. While these materials lack a global spin polarization, their electronic bands are spin-split in a momentum-dependent manner, as detailed in Chapter 1. This creates a rich texture of spin polarization across the Brillouin zone. One proposition on the topic [45], adapted the definition of Jullière TMR to a novel form of TMR rooted in the concept of spin-momentum-locked valleys. The altermagnetic spin splitting effectively divides the Fermi surface into distinct regions, or "valleys", each possessing a near-100% spin polarization. For instance, a region of the Brillouin zone around one high-symmetry point may be populated exclusively by spin-up electrons, while another region is populated exclusively by spin-down electrons. The TMR mechanism in an ALMTJ is then governed by the alignment of these spin valleys across the insulating barrier, as illustrated schematically in Fig. 4.1.

This mechanism acts as a perfect "spin-valve" in momentum space. Tunnelling is permitted or forbidden on a k -point-by- k -point basis depending on the relative Néel vector orientation. The resulting TMR is therefore not dependent on the total density of states, but on the precise overlap of these highly spin-polarized valleys. In principle, if the spin-valley polarization is complete, this effect could lead to a near-infinite TMR. This "spin-valleytronics" mechanism is fundamentally distinct from both the Jullière model in conventional ferromagnets and the orbital-based symmetry filtering in Fe/MgO/Fe junctions. It offers a pathway to giant magnetoresistance in devices that are simultaneously robust against external magnetic fields and capable of operating at the terahertz frequencies intrinsic to antiferromagnetic dynamics. The goal of this work is to computationally realize and

explore from first principles this powerful theoretical concept in a realistic device.

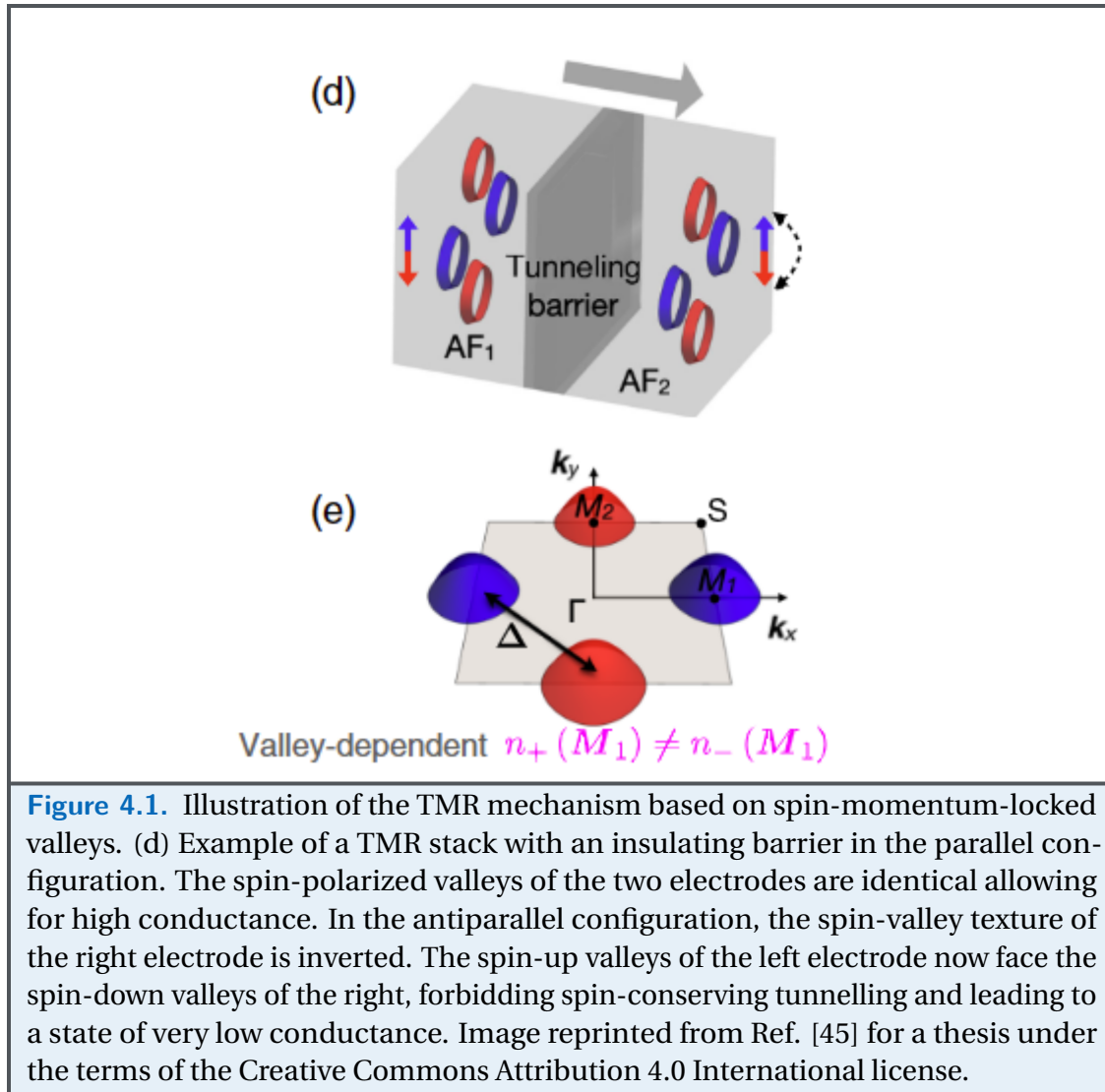


Figure 4.1. Illustration of the TMR mechanism based on spin-momentum-locked valleys. (d) Example of a TMR stack with an insulating barrier in the parallel configuration. The spin-polarized valleys of the two electrodes are identical allowing for high conductance. In the antiparallel configuration, the spin-valley texture of the right electrode is inverted. The spin-up valleys of the left electrode now face the spin-down valleys of the right, forbidding spin-conserving tunnelling and leading to a state of very low conductance. Image reprinted from Ref. [45] for a thesis under the terms of the Creative Commons Attribution 4.0 International license.

4.2 A "Golden Rule" Framework for

Antiferromagnetic Tunnel Junction Design

The theoretical promise of antiferromagnetic / altermagnetic tunnelling is vast, but its practical realization hinges on identifying and synthesizing suitable material heterostructures. The challenge lies in navigating the immense landscape of known magnetic compounds to find the rare combinations that satisfy the stringent physical and structural requirements for a high-performance AFMTJ-ALMTJ. To address this, we have developed a systematic, multi-step screening methodology based on a set of physically-grounded "golden rules". This framework allows us to move from abstract symmetry classifications to a concrete

list of promising, experimentally-viable material candidates. By experimentally-viable, we mean ensuring the materials are non-toxic, stable, and compatible with epitaxial growth techniques, allowing safe handling, high-quality synthesis, and practical use in laboratory settings with standard equipment. Taking out the experimentally-viable criteria, the number of candidates without this rule is skyrocketing.

Our search begins by focusing on the specific crystal symmetries that permit the desired form of magnetism. As discussed in Chapter 1, the classification scheme based on relativistic magnetic space groups (MSGs) [15, 113] provides a systematic way to identify materials capable of hosting non-relativistic spin splitting. Our primary interest is in materials belonging to the Spin-Splitting Type 4 (SST-4) class. This category encompasses all centrosymmetric and non-centrosymmetric crystals where a momentum-dependent spin splitting is allowed by symmetry in a compensated magnetic state, forming the very definition of a spin-split antiferromagnet (SS-AFM). Moreover, our workflow combines symmetry analysis with materials database mining. Firstly, we identify all MSGs corresponding to the SST-4 classification. Secondly, we query the MAGNDATA database [114], a comprehensive repository of experimentally determined magnetic structures, for all real material prototypes that crystallize in one of these SST-4 compatible space groups. This process yields a foundational list of potential SS-AFM electrodes. The final, most critical step is to apply our set of golden rules to this list to screen for viability and to identify suitable insulating barrier materials from the Crystallographic Open Database (COD) [115, 116].

4.2.1 The Golden Rules for AFMTJ / ALMTJ Design

1. **Rule 1: The Electrode Must Be a Metallic Spin-Split Antiferromagnet / Altermagnet.** The fundamental requirement for the electrodes is that they must be metallic to support a current and must exhibit bandstructure spin-splitting as SS-AFM and altermagnets. The metallic nature ensures a sufficient density of states at the Fermi level to act as a source and drain for tunnelling electrons. The altermagnetic order provides the crucial momentum-dependent spin splitting that enables the TMR effect.
2. **Rule 2: The Néel Temperature is at least Room Temperature.** For any practical application in spintronic memory or logic, the device must operate reliably at and above room temperature (300 K). This necessitates that the magnetic order of the

altermagnetic electrodes be robust, with a Néel temperature (T_N) significantly higher than the operating temperature. Applying this filter to the initial list of SST-4 candidates from MAGNDATA is a critical screening step. A complete list of altermagnetic candidates is provided in Chapter A for completeness, our primary focus for device design will be on the subset of materials with reported $T_N \geq 300$ K.

3. **Rule 3: Structural and Electronic Compatibility of the Insulator.** The insulating barrier is not a passive component; its properties are as crucial as those of the electrodes. The selection of a suitable insulator is governed by three strict criteria related to its compatibility with the chosen altermagnetic metal:

- **Crystallographic Symmetry Matching:** For coherent quantum mechanical tunnelling, which is the basis of giant TMR, the atomic structure at the electrode-insulator interface must be as close to perfect as possible. This requires that the insulator shares the same crystallographic (non-magnetic) space group as the SS-AFM / altermagnetic electrode. For instance, for an altermagnet with a rutile structure as RuO_2 ($P4_2/mnm$), the ideal insulator should also crystallize in a rutile or a closely related tetragonal structure.
- **Minimal Lattice Mismatch:** High-quality, single-crystal heterostructures are grown via epitaxy, where the crystal lattice of one material is grown in atomic layer-by-layer fashion. This process is only possible if the lattice parameters of the two materials are very closely matched. A lattice mismatch greater than a few percent introduces dislocations and other defects at the interface. These structural imperfections, alongside potential challenges like chemical intermixing or thermodynamic instability act as scattering centres that destroy the coherent tunnelling required for a large TMR. While a strict experimental preference for high-quality epitaxy is often a mismatch of $< 2\%$, we relax this constraint in our theoretical search to a broader range of $3 - 4\%$ to identify a larger pool of potential candidates that might be realized with advanced growth techniques.
- **Sufficiently Large Band Gap:** The barrier material must be a robust electrical insulator. A key requirement is a wide band gap, which we define as $E_g > 3$ eV. If the band gap is too small, electrons can be thermally excited over the barrier (thermionic emission) or tunnel through a much lower effective barrier, creating

a large leakage current. This leakage current is not spin-dependent and acts as a shunt, masking the pure spin-polarized tunnelling current and drastically reducing the observed TMR.

Candidate Material	T_N (K)	MSG	Altermagnetic Class	Class
RuO ₂	> 300 (disputed)	$P4_2'/mnm'$	d -wave (xy)	SST-4 & Spin-Symmetry
CrSb	≈ 700	$P6_3'/m'm'c$	g -wave	SST-4 & Spin-Symmetry
CrSe	≈ 300	$P\bar{3}1m'$	Not classified	SST-4

Table 4.1. A summary of promising metallic altermagnetic candidates identified through both the relativistic Magnetic Space Group (MSG) classification and altermagnetic spin symmetries screening approaches. The table lists the known Néel temperature (T_N), the MSG (where applicable), and the predicted altermagnetic class (e.g., d -wave). The final column indicates the theoretical framework(s) that predicted the material's potential.

Going beyond the three primary rules for electrodes identification, an ideal ALMTJ would employ an insulator that is, itself, an altermagnet. While seemingly counter-intuitive, an altermagnetic insulator does not conduct charge but possesses a spin-split electronic structure, including its evanescent states within the band gap. These evanescent states are the quantum mechanical wavefunctions that decay exponentially inside the barrier, mediating the tunnelling process. The physics of coherent tunnelling in MTJs is governed by symmetry filtering. A giant TMR is achieved when the propagating states in the electrodes have a specific symmetry (e.g., the Δ_1 symmetry in Fe / MgO / Fe) that matches the symmetry of the most slowly decaying evanescent state in the barrier. In an ALMTJ, this principle is extended to both orbital and spin symmetry. If the insulator were also an altermagnet with the same spin symmetry as the electrodes, it would create a near-perfect spin and symmetry filter. It would selectively allow the tunnelling of electrons with a specific spin and k -space symmetry while strongly blocking all others. This perfect matching would, in principle, lead to an exceptionally high TMR, potentially far exceeding what is possible with a conventional non-magnetic insulator. While finding a material that satisfies all golden rules and is an altermagnetic insulator is a formidable challenge, it represents the ultimate goal in rational ALMTJ design.

In conclusion, these systematic screening strategies provide a powerful roadmap for the discovery of new spintronic devices. On one hand, the relativistic magnetic space

group approach is highly effective for mining existing databases of known magnets staying in the traditional two magnetic phases framework. On the other hand, the non-relativistic spin-symmetry approach offers a predictive power that can uncover altermagnetism in previously overlooked materials. The identification of several high- T_N candidates with suitable crystal structures confirms that the design of novel ALMTJs is not merely a theoretical exercise but a tangible goal for experimental materials science. Having established these design principles, the remainder of this chapter will focus on the design and the detailed computational analysis of the prototypical RuO_2 / TiO_2 / RuO_2 system.

— 4.3 Candidate Materials and Heterostructure Design —

Having established a systematic design framework, we now apply our golden rules to the candidates identified from the SST-4 symmetry class and the broader altermagnetism literature. This screening process allows us to transition from a list of promising individual materials to concrete, viable heterojunctions, forming the blueprint for a new generation of spintronic devices. Our analysis focuses on the three most promising room-temperature metallic altermagnets identified in Section 4.2: RuO_2 , CrSb , and CrSe . For each candidate, we search for a suitable insulating barrier, paying close attention to structural, electronic, and symmetry compatibility to make the most viable AFMTJs and ALMTJs.

4.3.1 The RuO_2 / TiO_2 / RuO_2 System: A Prototypical Case Study

The RuO_2 system is arguably the most compelling candidate for an initial deep-dive analysis. As the archetypal d -wave altermagnet, it has been the subject of intense theoretical and experimental study as we have seen in Chapter 3. The search for a suitable insulating barrier naturally leads to titanium dioxide (TiO_2), another common rutile-structured oxide.

Let us evaluate this pairing against our golden rules:

- **Symmetry Matching:** Both RuO_2 and TiO_2 (in its rutile phase) share the same non-magnetic parent space group, $P4_2/mnm$. This identical crystallographic symmetry is the ideal scenario, as it allows for a seamless structural transition at the interface, minimizing disruptions to the electronic wavefunctions that mediate quantum

tunnelling.

- **Lattice Mismatch:** The lattice parameters for RuO_2 ($a = 4.492 \text{ \AA}$, $c = 3.106 \text{ \AA}$) and TiO_2 ($a = 4.594 \text{ \AA}$, $c = 2.959 \text{ \AA}$) are reasonably well-matched. The in-plane mismatch along the a -axis is approximately +2.3%, which falls within the bounds for high-quality epitaxial growth. The mismatch along the c -axis is larger, at approximately $-4.9 - 5\%$. Despite this, the stacking direction along the longitudinal c -axis is not problematic. While this is outside the ideal experimental window, it can often be accommodated in thin-film growth through strain engineering.
- **Insulator Band Gap:** Rutile TiO_2 has a wide and experimentally verified band gap of approximately 3.1 eV [117], comfortably satisfying our $> 3 \text{ eV}$ criterion to prevent leakage currents and ensure transport is dominated by tunnelling.

Based on these criteria, the $\text{RuO}_2 / \text{TiO}_2 / \text{RuO}_2$ heterostructure emerges as an exceptionally strong candidate. The existing proposal of this junction in the literature [18], which predicted a giant TMR of $\approx 500\%$, provides a valuable point of reference. However, our investigation approaches the system from a different theoretical framework. Whereas the prior work relied on the parametric DFT+ U method, our study will employ the parameter-free ASIC approach, as justified in Chapter 3, to provide a novel perspective on the intrinsic transport properties of this junction. The combination of a strong theoretical precedent, excellent structural compatibility, and the opportunity for direct comparison makes the $\text{RuO}_2 / \text{TiO}_2 / \text{RuO}_2$ system the ideal prototype for the detailed analysis in the remainder of this chapter. The potential insulators are summarized in Tab. 4.2.

Insulator Candidate for RuO_2	Band Gap (eV)	Lattice Mismatch (a)
TiO_2	3.1 [117]	+2.3%
BaO	3.8 [118]	+2.2%

Table 4.2. Potential insulating barriers for RuO_2 . TiO_2 is an excellent candidate due to its identical crystal structure and reasonable lattice match. BaO, while having a large gap, possesses a different crystal structure (rock-salt) making epitaxial growth challenging since BaO needs to be grown slightly tilted with respect to RuO_2 .

4.3.2 The CrSb/ NiTe System: A High-Temperature Candidate

Beyond RuO₂, chromium antimonide (CrSb) stands out as a highly robust candidate due to its exceptionally high Néel temperature of ≈ 700 K. For this hexagonal altermagnet, our screening identified nickel telluride (NiTe) as a promising insulating barrier. NiTe is a non-magnetic insulator with a large band gap of 3.59 eV [119]. The structural match is also compelling. CrSb has a hexagonal lattice with parameters $a = 4.103$ Å and $c = 5.463$ Å. NiTe crystallizing in a similar hexagonal structure, has parameters $a = 3.964$ Å and $c = 5.351$ Å. This corresponds to an in-plane lattice mismatch of -3.4% , which is within our relaxed screening window, and a c -axis mismatch of -2.1% , which is excellent. Given its high thermal stability and good structural compatibility, the CrSb/NiTe system represents a very promising avenue for future experimental and theoretical exploration.

Insulator Candidate for CrSb	Band Gap (eV)	Lattice Mismatch (a/c)
NiTe	3.59 [119]	$-3.4\% / -2.1\%$

Table 4.3. A promising insulator candidate for CrSb. NiTe offers a large band gap and good lattice compatibility, making the heterostructure a strong candidate for high-temperature altermagnetic devices.

4.3.3 The Case of SST-4 CrSe: Challenges in Insulator Matching

Chromium selenide (CrSe) with a Néel temperature of ≈ 300 K sits just at the threshold for room-temperature applications. However, applying our golden rules to this material reveals a significant challenge: finding a suitable insulating barrier. Our database search identified ruthenium bromide (RuBr₃) as a potential structural match, but as shown in Tab. 4.4, its electronic properties are unsuitable. With a band gap of only 0.70 eV [120], RuBr₃ is a semiconductor, not a wide-gap insulator. It would permit significant leakage current, making it impossible to build a functional tunnel junction. This finding is, in itself, an important result of our systematic screening. It demonstrates that not every promising SST-4 in this case or an altermagnet can be easily integrated into a tunnel junction. The lack of a readily available, wide-gap, lattice-matched insulator currently makes CrSe a less viable candidate for ALMTJ applications compared to RuO₂ or CrSb, highlighting the critical importance of considering the entire heterostructure, not just the electrode, in

materials design.

Insulator Candidate for CrSe	Band Gap (eV)	Notes
RuBr ₃	0.70 [120]	Non-viable; band gap too small for a tunnel barrier.

Table 4.4. Screening results for CrSe. The identified structurally similar material, RuBr₃, fails the electronic requirements of our golden rules due to its small band gap.

Having systematically evaluated the top candidates, our path forward is clear. The RuO₂/TiO₂ system provides the most compelling combination of factors for an in-depth computational study. The following sections will therefore focus entirely on a first-principles analysis of the quantum transport properties of this prototypical altermagnetic tunnel junction.

— 4.4 The RuO₂ / TiO₂ / RuO₂ Model System

Based on the screening process in the previous section, which identified the RuO₂ / TiO₂ / RuO₂ ALMTJ as a uniquely promising candidate, we now proceed to construct a detailed computational model of this system. This model is built under the working hypothesis that the altermagnetic phase of RuO₂ can be stabilized (e.g. in a thin-film geometry), allowing us to provide a definitive theoretical benchmark for the performance of such a device and to explore the rich underlying transport physics. Our investigation will be conducted using the SIESTA package [65] for obtaining a self-consistent electronic structure coupled with the SMEAGOL code [64], which implements the non-equilibrium Green's function (NEGF) formalism to simulate quantum transport.

4.4.1 Device Geometry and Magnetic States

The model altermagnetic tunnel junction is constructed to be periodic in the transverse (x, y) directions and open for transport along the longitudinal (z) direction. The device consists of three main parts: two semi-infinite RuO₂(001) electrodes acting as the source and drain, and a central scattering region. This scattering region contains the insulating TiO₂ (001) barrier as well as several "buffer" layers of the RuO₂ electrodes to ensure

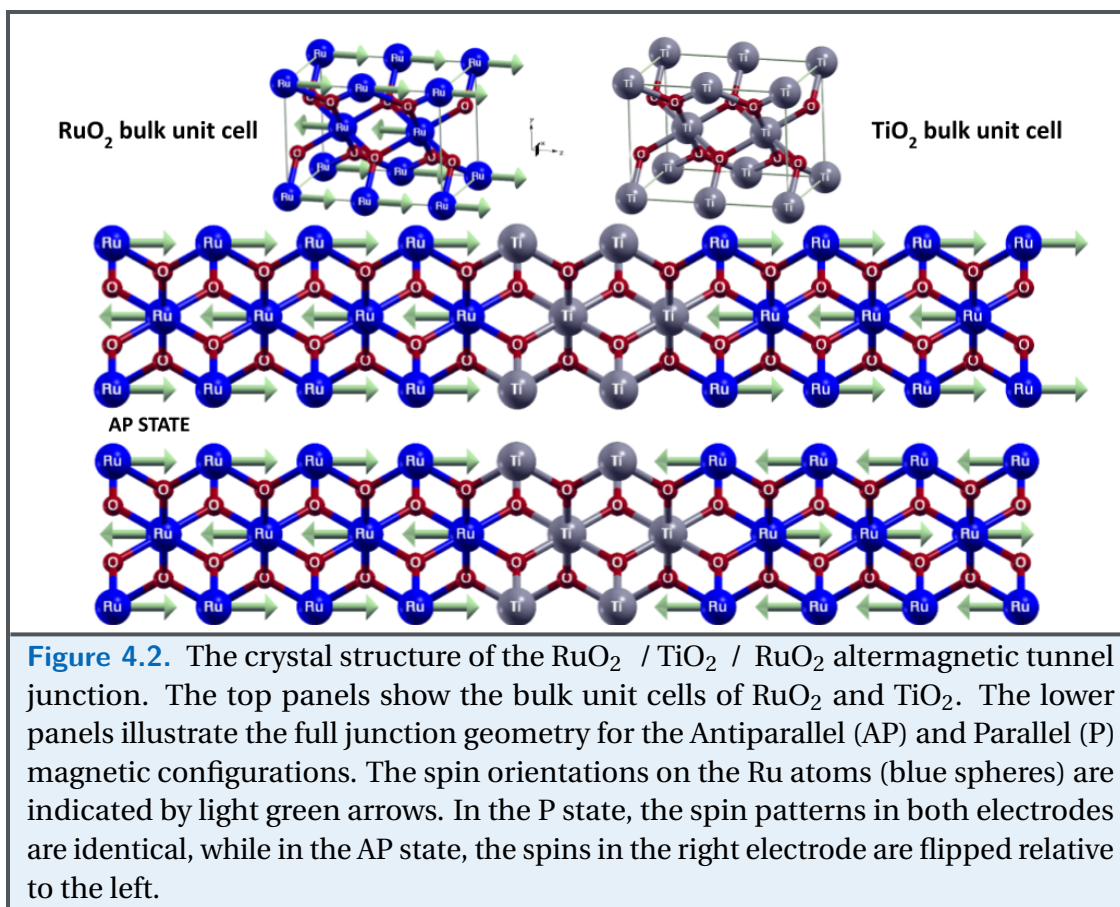
screening of the Friedel oscillations and a smooth transition of the electronic potential from the bulk leads into the barrier. Our primary model consists of a scattering region with a total of 60 atoms, comprising 8 monolayers (ML) of RuO_2 on each side of a 4 ML TiO_2 barrier. To investigate the effect of barrier thickness, a second model with an 8 ML TiO_2 barrier (for a total of 72 atoms) was also constructed. The interface between the materials is atomically sharp, created by stacking the bulk-like unit cells without any specific structural reconstruction. The complete geometry of the junction, along with the bulk unit cells of RuO_2 and TiO_2 , is illustrated in Fig. 4.2.

A key requirement for describing the TMR effect is an unambiguous definition of the magnetic states of the junction. For the altermagnetic system, we define two distinct configurations, also depicted in Fig. 4.2:

- **In the Parallel (P) configuration**, the Néel vectors of the two altermagnetic electrodes are aligned. In this configuration, the local spin arrangement on the Ru sublattices is identical in both the left and right electrodes. This means their spin-valley textures in momentum space are identical. A spin-up electron approaching the barrier from a specific k -point valley in the left electrode finds a corresponding spin-up valley at the same k -point in the right electrode. Since spin is conserved during the quantum mechanical tunnelling process (no SOC), this channel is open, leading to a high conductance state.
- **In the Antiparallel (AP) configuration**, the Néel vector of one electrode is flipped relative to the other. This operation has the crucial effect of reversing the spin label of every valley in that electrode's Brillouin zone. Now, the spin-up electron from the left electrode encounters a spin-down valley at the same k -point in the right electrode. As the spin-flip process is strongly suppressed in the tunnelling barrier, this channel is effectively closed.

The TMR effect arises from the difference in quantum tunnelling conductance between

these two well-defined magnetic states.

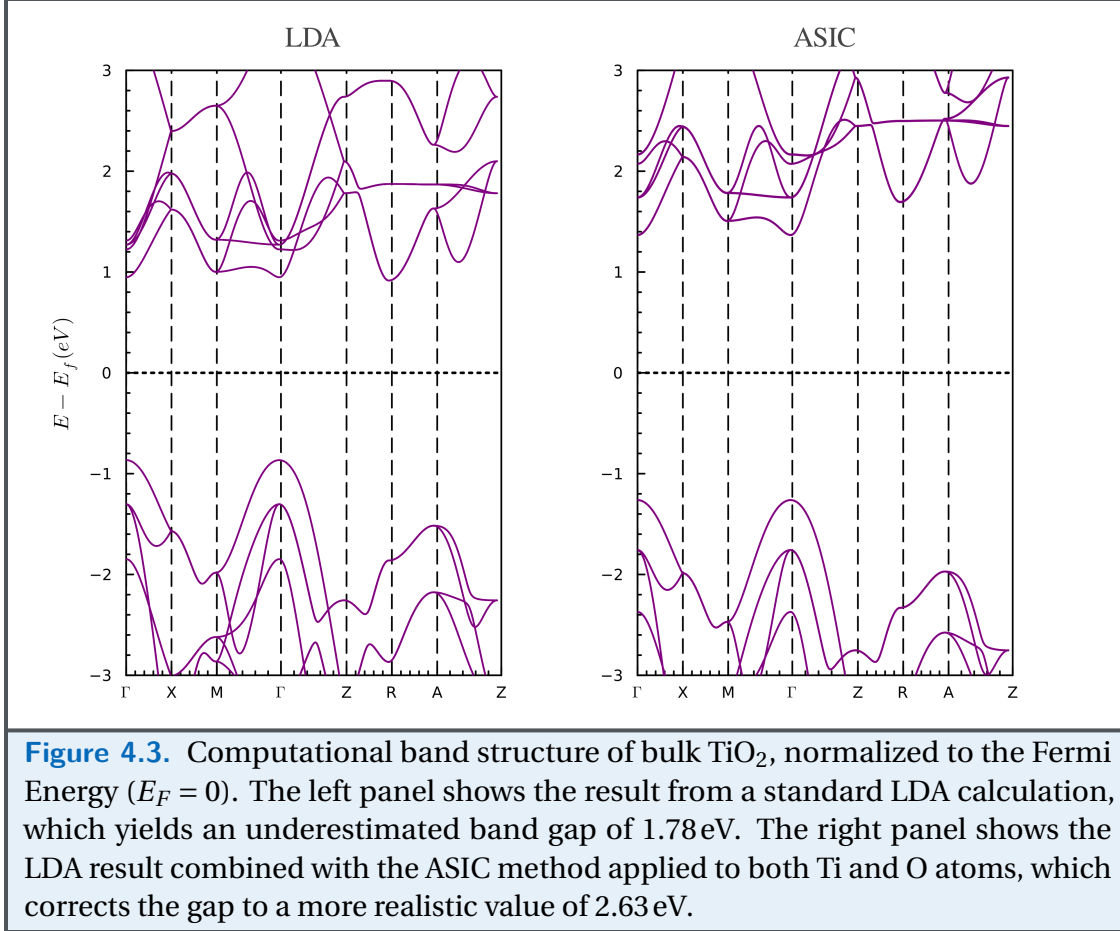


4.4.2 Computational Methodology for Quantum Transport

The predictive power of our transport simulations depends critically on the accuracy of the underlying DFT model. As established in Chapter 3, standard LDA or GGA functionals are insufficient to describe the altermagnetic state of RuO₂. We therefore employ a sophisticated **hybrid ASIC approach** tailored to the physics of each component of the junction.

- **For the RuO₂ Electrodes:** We use the LDA-ASIC(Ru) method. As demonstrated in Chapter 3, applying the atomic self-interaction correction only to the Ru atoms, while treating the electrode Oxygen atoms with standard LDA, yields a band structure that is most consistent with the predictions of altermagnetism theory.
- **For the TiO₂ Barrier:** We apply the LDA-ASIC correction to both the Ti and O atoms. The primary goal here is to obtain the most physically accurate insulating band gap, which governs the decay of the electronic wavefunctions and thus the magnitude of the tunnelling current.

Figure 4.3 illustrates the necessity of this correction for the barrier. Standard LDA severely underestimates the band gap of TiO_2 , yielding a value of only 1.78 eV. The application of ASIC opens this gap significantly to 2.63 eV. While this is still smaller than the experimental value of ≈ 3.1 eV, it represents a crucial, parameter-free improvement over LDA and provides a much more realistic description of the tunnel barrier.



The transport calculations follow a rigorous two-step process to ensure both accuracy and computational efficiency. First, a self-consistent charge density for the entire junction (open boundaries) at equilibrium was obtained using SMEAGOL. These SCF calculations were performed with a Double-Zeta Polarized (DZP) basis set and a $15 \times 15 \times 1$ Monkhorst-Pack k -point grid until the density matrix converged to a tolerance of 10^{-4} . Subsequently, non-self-consistent transmission coefficient calculations were performed using SMEAGOL on the previously converged Hamiltonian and Density Matrix. For this step, a much denser $60 \times 60 \times 1$ k -point grid for zero bias, transmission coefficient calculation and $40 \times 40 \times 1$ k -point grid for finite bias were used to accurately resolve the fine features of the transmission spectrum across the Brillouin zone. All calculations were performed with a real-space mesh cutoff of 600 Ry and a fully spin-polarized formalism. The adequacy of the $60 \times 60 \times 1$

zero-bias transmission grid was verified by comparison with a $80 \times 80 \times 1$ grid: the peak TMR at the Fermi level changes by less than 5%, and the transmission hot-spot positions in the Brillouin zone are unchanged. The estimated numerical uncertainty on the TMR values reported below is $\pm 10\text{--}20\%$ of the quoted percentage (e.g., $427\% \pm \sim 40\%$), while the predicted inversion voltage $V_{inv} \approx 0.35\text{ V}$ carries an uncertainty of $\pm 0.05\text{ V}$ from the finite bias-step resolution. A comprehensive account of all convergence criteria is given in Section 2.5.

— 4.5 Zero-Bias Transport Properties and Tunnelling Magnetoresistance —

Before exploring the complexities of finite-bias operation, it is essential to first characterize the intrinsic transport properties of the $\text{RuO}_2 / \text{TiO}_2 / \text{RuO}_2$ device in the zero-bias limit under the altermagnetic hypothesis. This idealized scenario provides a direct probe of the system's electronic structure around the Fermi level and allows for a clear, fundamental understanding of the TMR effect. The transport properties are calculated using the Landauer formula, adapted to ALMTJ definition of P and AP state where the zero-bias conductance (G) for a given spin channel is proportional to the transmission coefficient, $T(E)$, evaluated at the Fermi energy (E_F).

For a two-terminal device, the total TMR is defined by the conductance in the Parallel (P) and Antiparallel (AP) magnetic states. At zero bias, this can be expressed directly in terms of the total transmission coefficients:

$$TMR(E) = \frac{T_P(E) - T_{AP}(E)}{T_{AP}(E)} \quad (1)$$

where $T_{P/AP}(E) = T_{P/AP,\uparrow}(E) + T_{P/AP,\downarrow}(E)$ are the respective transmission coefficient for the parallel and antiparallel state of the MTJ. We now analyse the results for our three primary computational models: the ASIC model with a 4 ML and 8 ML barrier, and the standard LDA model with an 8 ML barrier.

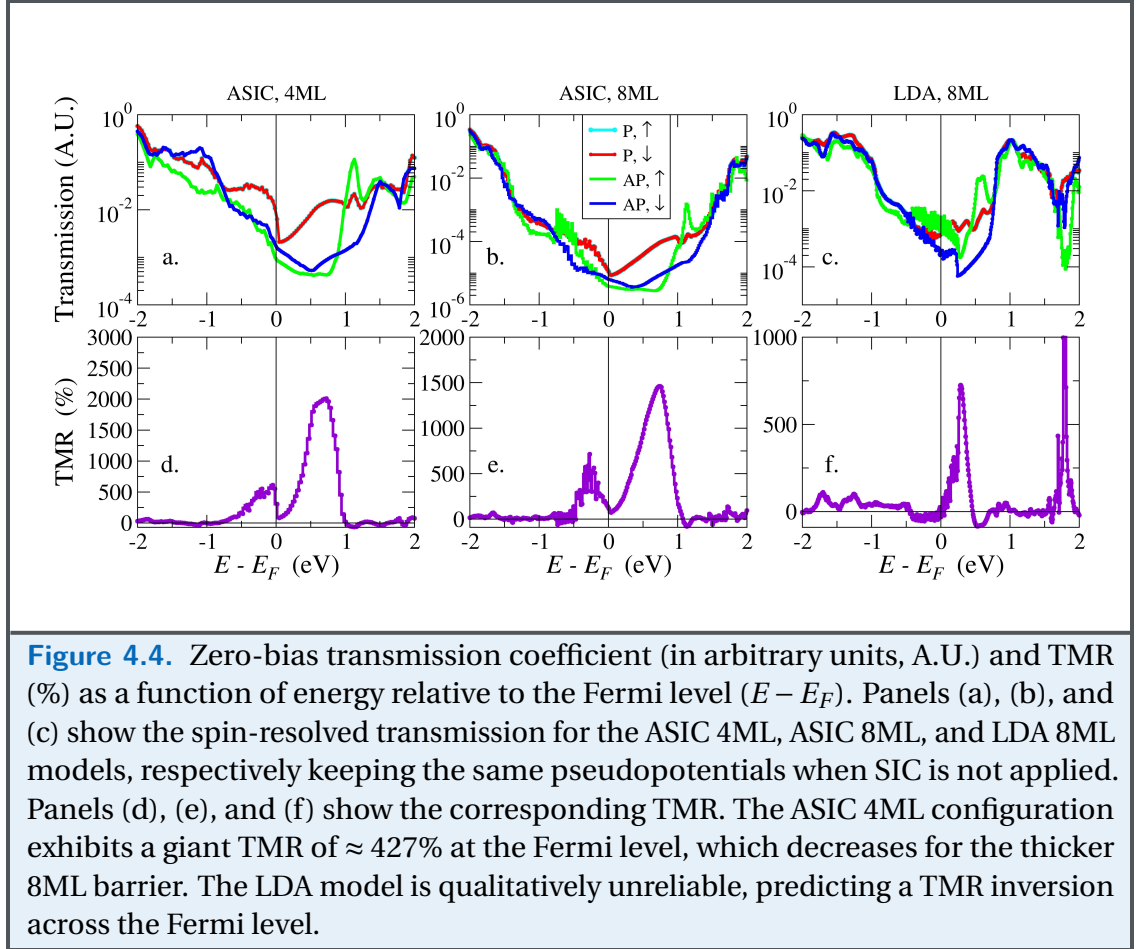
4.5.1 Giant Tunnelling Magnetoresistance verification in the ASIC Model

The central results of our zero-bias calculations are presented in Fig. 4.4. The top panels display the spin-resolved transmission coefficients on a logarithmic scale, while the

bottom panels show the corresponding energy-resolved TMR. Let us first examine the most ideal case: the ASIC model with a thinner 4 ML TiO_2 barrier, shown in Fig. 4.4 a). The transmission for the P state (cyan and red curves) is significantly higher than that for the AP state (green and blue curves) in the vicinity of the Fermi level ($E_F = 0$). This large gap between the P and AP transmission, arising from the efficient symmetry matching of wavefunctions in the P state, is the microscopic origin of the magnetoresistance. It is crucial to note that for the P state, the total device possesses time-reversal symmetry, which dictates that the transmission must be identical for both spin channels ($T_{P,\uparrow} = T_{P,\downarrow}$) in the SST-4 framework. In the altermagnetism framework, the junction preserves a symmetry operation with a lattice rotation in the crystallographic space group forming a protective symmetry. This forces the bands to be Kramers-degenerate dictating that the transmission must be identical for both spin channels ($T_{P,\uparrow} = T_{P,\downarrow}$) in both SST-4 and altermagnetism framework. This degeneracy is lifted in the AP state, where this combined symmetry is broken. This large transmission gap translates directly into a giant tunnelling magnetoresistance, which, as shown in Fig. 4.4 d. reaches a peak value of approximately 427% precisely at the Fermi level. In fact, this result is highly significant for two reasons. First, it demonstrates that an altermagnetic tunnel junction can, in principle, achieve a TMR ratio that is highly competitive with state-of-the-art ferromagnetic MTJs. Second, our calculation was performed using the parameter-free ASIC method. The fact that our result is in excellent agreement with the $\approx 500\%$ TMR predicted in [18] using the parametric DFT+ U method provides powerful, independent validation. It strongly suggests that the giant TMR effect in the $\text{RuO}_2 / \text{TiO}_2 / \text{RuO}_2$ system is a robust physical phenomenon, not an artifact of a specific computational approach, e.g. in the case of ASIC, that proved its theoretical reliability before [61]. The predicted Ru moment for ASIC calculation is $M_{\text{Ru}} = 0.987 \mu_B$ whereas the classic LDA moment is $M_{\text{Ru}} = 1.33 \times 10^{-4} \mu_B$.

The quantitative agreement between our parameter-free ASIC result (427%) and the DFT+ U prediction ($\approx 500\%$) from [18] is not coincidental: both calculations capture the same underlying spin-valley filtering mechanism, differing only in the level of the electron-correlation correction applied to the Ru 4d shell. This robustness is further corroborated at the model-Hamiltonian level. Šmejkal et al. derived a general analytical framework for valley-dependent TMR in altermagnets using a spin-valley tight-binding model of the form $\hat{H}_{\text{AM}}(\mathbf{k}) = \varepsilon_0(\mathbf{k})\hat{\sigma}_0 + J_{\text{AM}}(\mathbf{k})\hat{\sigma}_z$, where for the rutile d -wave symmetry $J_{\text{AM}}(\mathbf{k}) = 2t_{\text{AM}}[\cos(k_x a) - \cos(k_y a)]$ [45]. This minimal model reproduces the key qualita-

tive features of the first-principles band structure: alternating spin-polarized valleys at the Fermi surface separated by nodal planes along the $\langle 110 \rangle$ directions. The model prediction of a giant TMR in this symmetry class provides an independent, analytically tractable validation of the mechanisms observed in our full NEGF-DFT calculation. Ab-initio results that are simultaneously supported by such model calculations carry substantially greater interpretive weight, as they demonstrate that the effect is not a numerical artefact but a direct consequence of the symmetry of the altermagnetic phase.



4.5.2 Effect of Barrier Thickness and the Failure of LDA

Comparing the 4 ML ASIC device (Fig. 4.4 a,d) with the 8 ML ASIC device (Fig. 4.4 b,e) reveals the expected impact of the barrier thickness. As the TiO_2 barrier is increased from 4 ML to 8 ML, the overall magnitude of the transmission coefficient drops by approximately two orders of magnitude due to the exponential decay of the wavefunctions inside the insulating gap. This reduction in conductance is accompanied by a significant decrease in the peak TMR, which falls from 427% to a still substantial 126%. Expectedly, this behaviour is characteristic of coherent tunnelling devices. Thicker barriers increase the probability of inelastic scattering and dephasing events that disrupt the phase coherence of the tunnelling

electron, thereby diminishing the magnetoresistive effect. This result underscores the critical importance of fabricating atomically precise, high-quality, and ultra-thin barriers for achieving optimal device performance. Finally, we turn to the results from the standard LDA functional for the 8 ML barrier, shown in Fig. 4.4 c,f). The outcome is strikingly different. The gap between the P and AP transmission curves is much smaller and less defined. Most importantly, the calculated TMR exhibits a sharp inversion across the Fermi level, swinging from positive values ($\approx +20\%$) just below E_F to negative values ($\approx -30\%$) just above it. This demonstrates that the standard LDA model is not merely quantitatively inaccurate but is qualitatively unreliable for this system, as it fails to even predict the correct sign of the magnetoresistance depending on the precise energy. This finding reinforces the conclusions from Chapter 3: that standard local functionals are insufficient to capture the essential physics of altermagnetism in this system, making the use of advanced, physically-motivated correction schemes like ASIC indispensable.

These promising zero-bias results, which establish the potential for a giant TMR effect in the $\text{RuO}_2 / \text{TiO}_2 / \text{RuO}_2$ system, provide strong motivation to investigate the device performance under more realistic finite-bias conditions. Moving beyond the idealized zero-bias scenario is essential, as any practical electronic device must operate with an applied voltage. This investigation will therefore be critical for assessing the device's true capabilities by characterizing the robustness of the TMR effect against bias. In fact, evolution under finite bias is a key factor for the technological viability of ALMTJs and will provide a deeper understanding of the underlying physics at play in a functional device.

— 4.6 Quantum Transport under Finite Bias —

The analysis of the zero-bias transport properties in the preceding section revealed the significant potential of the $\text{RuO}_2 / \text{TiO}_2 / \text{RuO}_2$ system under the altermagnetic hypothesis. We have predicted a giant TMR effect shown in Section 4.5 competitive with mature ferromagnetic technologies. However, real-world spintronic devices operate under an applied finite bias voltage. It is therefore of high importance to investigate whether this promising TMR survives under operational conditions and to uncover what new physical phenomena may emerge. This section presents the core results of our investigation into

the non-equilibrium transport properties of the ALMTJ, built upon the strong foundation of our zero-bias findings.

4.6.1 Theoretical Framework for Finite-Bias Transport

When a finite bias voltage (V) is applied across the tunnel junction, the electrochemical potentials of the left (μ_L) and right (μ_R) electrodes are shifted relative to each other, with $eV = \mu_L - \mu_R$. In this non-equilibrium scenario, the simple zero-bias TMR definition around Fermi level becomes insufficient. Instead, the electrical current (I) must be calculated directly by integrating the energy and voltage dependent transmission coefficient, $T(E, V)$, over the energy window defined by the two electrochemical potentials. As detailed in Chapter 2 of this thesis, the current is given by the Landauer-Büttiker formula:

$$I(V) = \frac{2e}{h} \int_{-\infty}^{\infty} T(E, V) [f(E - \mu_L) - f(E - \mu_R)] dE \quad (2)$$

where $f(E)$ is the Fermi-Dirac distribution. Consequently, the TMR at a finite bias voltage V must be redefined in terms of the calculated currents flowing in the Parallel (I_P) and Antiparallel (I_{AP}) magnetic states:

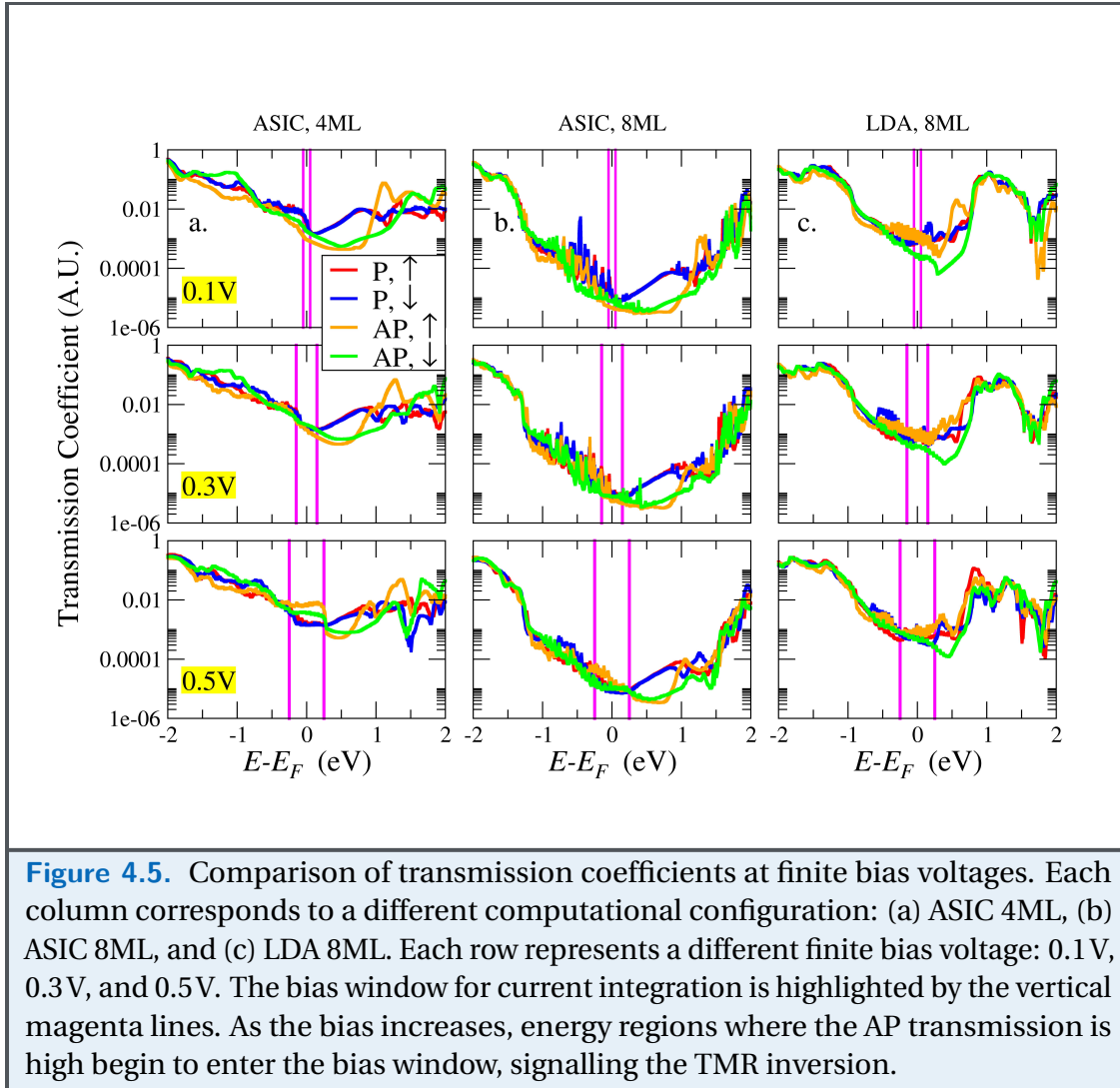
$$TMR(V) = \frac{I_P(V) - I_{AP}(V)}{I_{AP}(V)} \quad (3)$$

This current-based definition allows for a complete characterization of the device's magnetoresistive response as a function of the operating voltage. The physical explanation of TMR as a spin-valley dependent phenomena has been given in Section 4.1.

4.6.2 Transmission Spectra under Finite Bias

Before analysing the calculated $I - V$ characteristics, it is instructive to examine how the transmission spectrum itself evolves under an applied bias. Figure 4.5 displays the transmission coefficients for our three models at bias voltages of 0.1 V, 0.3 V, and 0.5 V. The magenta lines indicate the energy window over which the transmission is integrated to calculate the current. At a low bias of 0.1 V (Fig. 4.5, top row), the situation is qualitatively similar to the zero-bias case. For the ASIC model, the transmission for the P state remains significantly higher than for the AP state within this narrow energy window close to Fermi energy, suggesting the TMR will remain positive and large. However, as the bias voltage increases to 0.3 V and 0.5 V, a critical new feature emerges. The bias window widens

and it begins to include energy regions where the transmission of the AP state becomes comparable to, or even exceeds, the P state one. This is particularly noticeable for the spin-down channel of the AP state ($AP, \downarrow$, blue curve), which develops prominent peaks at positive energies. Since the total current is an integral of these curves over the bias window, the growing contribution from these highly conductive AP channels indicates a complex evolution of the TMR. This suggests that the simple picture of P-state dominance may break down at higher biases, a point that will be conclusively demonstrated by the $I - V$ curves and will be explained in detail through a $\mathbf{k}_\perp$ -resolved analysis in the following section.

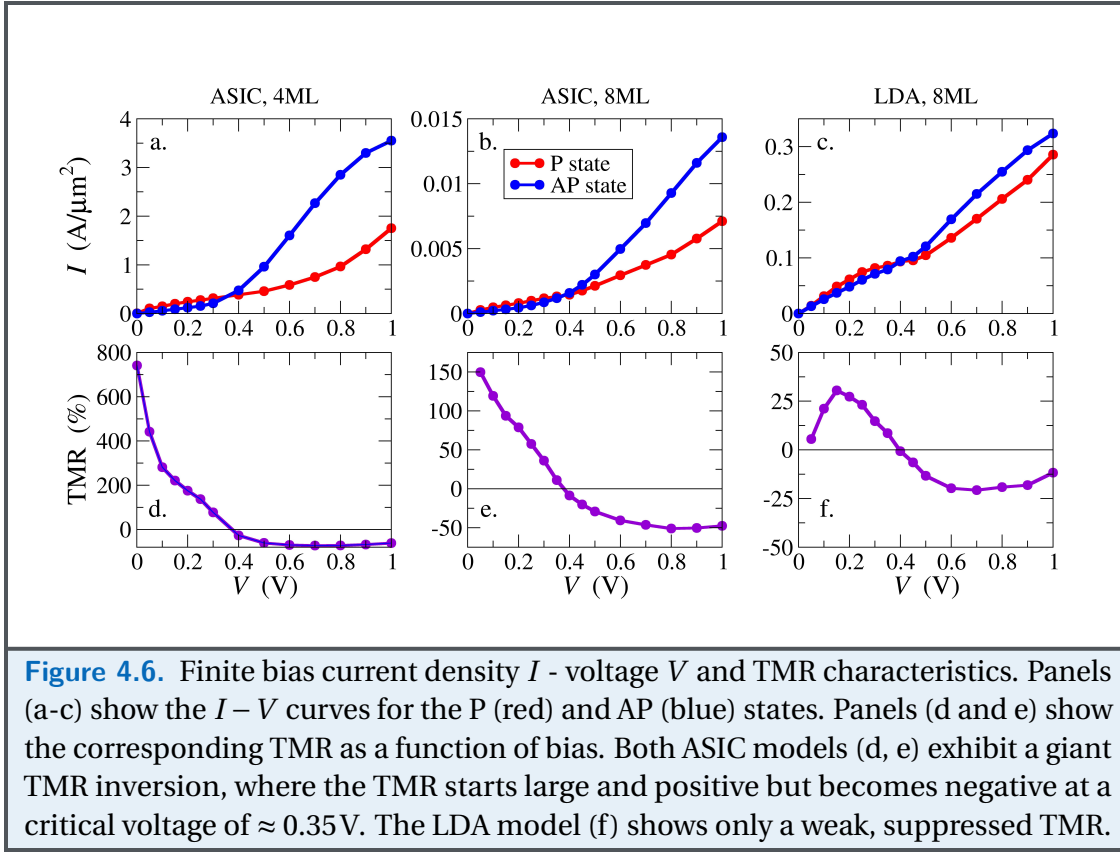


4.6.3 Current-Voltage Characteristics and Giant TMR Inversion

The final, decisive results of our transport calculations are the current-voltage ($I - V$) characteristics and the resulting bias-dependent TMR, presented in Fig. 4.6. These plots reveal a rich and non-trivial behaviour that constitutes one of the major findings of this thesis.

Let us first analyse the ASIC 4ML model (Fig. 4.6 a,d), which represents the most favourable computational scenario. At low bias, the current in the P state (I_P) is substantially larger than in the AP state (I_{AP}), as expected from the zero-bias results. This leads to a very large TMR, which is still over 300% at 0.1 V for 4ML ALMTJ. However, as the voltage increases, the TMR decays rapidly. The $I - V$ curves in panel (a) clearly show that I_{AP} increases more steeply with voltage than I_P , leading to a crossover at approximately $V_{inv} = 0.35$ V. Beyond this point, the AP state becomes more conductive than the P state. This results in a dramatic TMR inversion, where the TMR passes through zero and becomes negative, reaching approximately -75% at higher biases for 4ML ALMTJ. A remarkably similar trend is observed for the thicker ASIC 8ML barrier (Fig. 4.6 b,e). While the absolute current magnitudes are much lower due to the thicker barrier, the qualitative physics remains the same. The TMR starts at a positive value ($\approx 150\%$ at 0.1 V) and inverts at a nearly identical voltage of $V_{inv} \approx 0.36$ V. The robust prediction of this giant TMR inversion across different barrier thicknesses in our parameter-free ASIC model suggests it is an intrinsic and fundamental property of the $\text{RuO}_2 / \text{TiO}_2 / \text{RuO}_2$ system. The inversion voltage $V_{inv} \approx 0.35$ V corresponds to an energy scale of ≈ 350 meV, roughly 14 times larger than the thermal energy $k_B T \approx 25$ meV at room temperature. This large ratio implies that the TMR inversion is a robust, thermally stable feature of the device, not subject to smearing by thermal fluctuations at operating temperature. Finally, the LDA 8ML model (Fig. 4.6 c, f) serves as a crucial control. Here, the $I - V$ curves for the P and AP states are much closer together, and the resulting TMR is heavily suppressed across the entire bias range. While a small oscillation around zero is visible, it lacks the clear, high-magnitude, and systematic inversion seen in the ASIC models. Once again, this confirms that standard LDA fails to capture the essential transport physics of this altermagnetic junction, further validating

the necessity of our advanced computational approach.



4.7 Microscopic Origin of the Bias-dependent TMR

The giant TMR inversion predicted by ASIC, where the junction switches from a low-conductance to a high-conductance state in the AP configuration under bias, is a remarkable phenomenon. While the $I - V$ characteristics presented in Section 4.6 conclusively demonstrate this effect, they do not reveal its underlying physical cause. To uncover the microscopic mechanism, we must move beyond the integrated current and analyse how the $\mathbf{k}_\perp$ -resolved transmission probability is distributed within the two-dimensional Brillouin zone. This section provides a detailed $\mathbf{k}_\perp$ -resolved analysis of the transmission channels. We will first establish the origin of the giant TMR at zero bias by examining the spin-dependent transmission. We will then show that the TMR inversion is driven by the bias-induced opening of highly efficient conduction pathways in a specific spin channel of the AP state, a mechanism rooted in the principles of symmetry filtering specific to the altermagnet symmetry.

4.7.1 $\mathbf{k}_\perp$ -resolved Transmission Analysis at Zero and Finite Bias

To uncover the origin of the bias-dependent TMR inversion, we first examine the $\mathbf{k}_\perp$ -resolved transmission maps, which provide crucial insights into the momentum-space contributions to the total conductance at a given energy. These maps visualize the transmission probability $T(\mathbf{k}, E)$ at specific energy points within the two-dimensional Brillouin zone (2D BZ) transverse to the transport direction, thereby identifying "hot spots" of high electronic transmission. The total transmission is an integral of the spin- and energy-dependent transmission coefficient, $T_\sigma(E, \mathbf{k}_\perp)$, over all parallel wavevectors $\mathbf{k}_\perp$ and both spin channels σ shown in Eq. (22). By visualizing these quantities, we can pinpoint the origin of high and low conduction.

Figure 4.7 compares the $\mathbf{k}_\perp$ -resolved transmission coefficients for the ASIC 8ML model as a function of energy and spin to the corresponding available conduction channels in bulk RuO_2 (top panels). At zero bias, the distinct behaviour of the P and AP states, which leads to the giant positive TMR, is quite straightforward. As shown in Fig. 4.7 (row "TRC P" and "TRC AP", 0 eV column for ASIC) and similarly for LDA in Fig. 4.9, on one hand for the P configuration, transmission is generally high and broadly distributed across several regions of the 2D BZ. Characteristic diagonal patterns and central cross-like features due to C_2 rotation axis in plane for the altermagnet RuO_2 indicate strong momentum-matched transmission channels. On the other hand, the P spin-down channel and the AP configuration (spin up and spin down) exhibits significantly suppressed transmission at 0 eV. This smaller value at 0 eV is explained by the bandgap of TiO_2 compared to ± 1 eV. Its maps are dominated by low transmission values (blue regions), particularly at and around the Γ point, with only isolated, weak transmission pathways visible. This profound difference at zero bias, where the P state offers better conductive channels while the AP state is strongly attenuated, directly underpins the large positive TMR observed at zero bias we've seen in Section 4.5. The highly transmissive majority-spin (up) channel in the P state faces a near-total blockade in the AP state due to the mismatch of the spin-split bands and the altermagnet lattice symmetry of spin-momentum-locked valley. Consequently, this strong suppression in the AP state at zero bias can be largely attributed to the spin-momentum locking of the altermagnetic electrodes, leading to a severe mismatch between the spin-polarized states across the interface when aligned antiparallel. Furthermore, Fig. 4.7 provides a more detailed spin-resolved picture for the ASIC case. While the "up" spin channel for the AP state (bottom row, "Transmission - AP

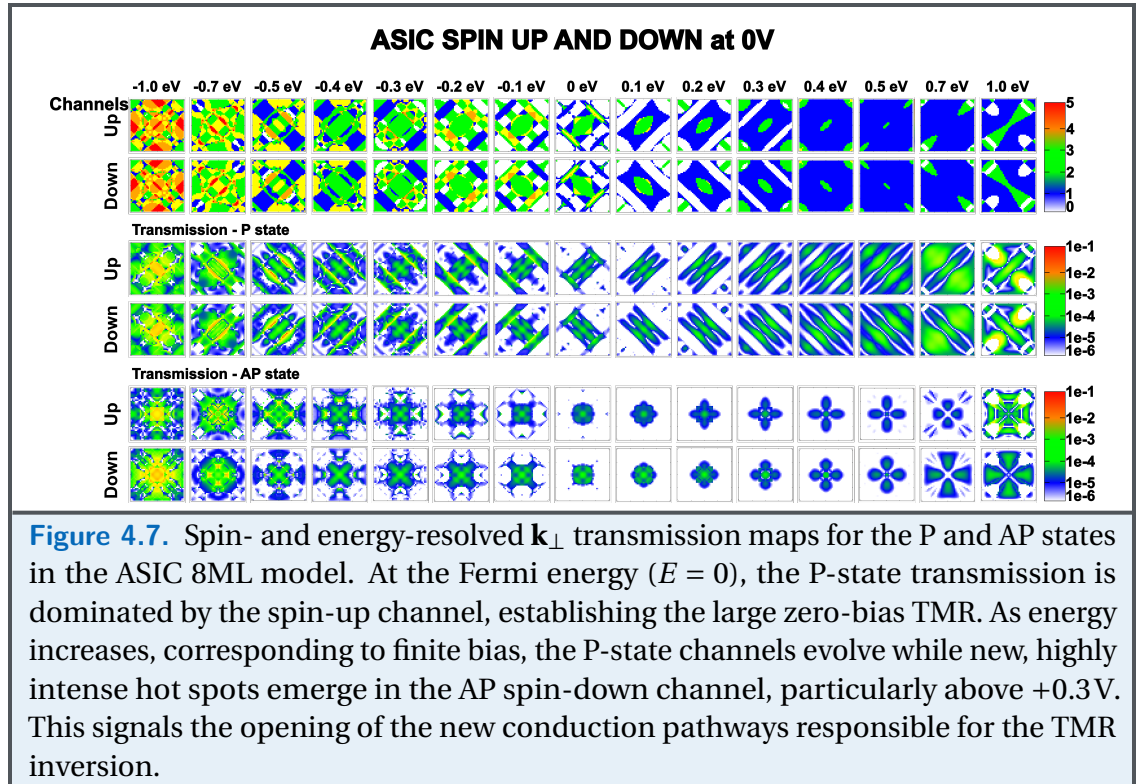
state Up") shows some emerging transmission at higher energies (e.g., 0.5V to 1.0V), the dramatic increase in the total AP transmission is particularly pronounced for the spin-down channel, which shows strong activation at positive energies (not explicitly shown in the provided "ASIC, spin up plotted" figure but suggested by the overall rise of AP transmission in Section 4.6 Fig. 4.5). This highlights the importance of considering both spin channels, as the altermagnetic nature introduces a complex interplay between spin and momentum. The opening of new channels for the AP state, especially for one spin component, is a direct consequence of the bias-induced energy shifts. Moreover, the LDA results in Fig. 4.9 and Fig. 4.8 (right panels) serve as a crucial counterpoint. Here, the zero-bias transmission difference between P and AP is much less pronounced. Critically, at finite bias, the AP state does not develop the same robust, highly conductive hot spots observed in the ASIC case. The $\mathbf{k}_\perp$ -resolved maps for LDA show consistently lower transmission magnitudes and less distinct features for both P and AP states across the energy range, reinforcing why the TMR inversion is suppressed or absent in this model. Overall, this zero-bias picture is the foundation for understanding the finite-bias behaviour. As a positive bias is applied, the integration window in Eq. (2) extends to positive energies in these maps. Examining the energy evolution in Fig. 4.7 reveals a critical trend:

- **P-State:** The high-transmission spin-up channel that dominates at Fermi energy gradually weakens and changes shape at higher energies.
- **AP-State:** While the spin-up channel remains resistive, the spin-down channel undergoes a dramatic transformation. Starting from around +0.2 V, new and extremely intense hot spots begin to emerge. By +0.5 V, these new features dominate the landscape, indicating the opening of incidental enhanced conduction channels that were completely unavailable at zero bias due to spin-momentum-locked valley channel matching at different energies specific to altermagnets.

This effect is summarized and directly visualized in Fig. 4.8, which compares the total transmission at low bias (0.1 V) and high bias (0.5 V). Indeed, the situation changes dramatically at finite bias, leading to the observed TMR inversion. Consider the $\mathbf{k}_\perp$ -resolved transmission maps at a higher bias voltage, for instance, 0.5 V, as presented in Fig. 4.8 (bottom row, panels "TRC P up" and "TRC AP up" for ASIC, spin up plotted). When a bias is applied, the energy window for current integration shifts, allowing access to different electronic states. At 0.5 V, a crucial development occurs in the AP state: new, highly con-

ductive transmission channels open up. For the ASIC case, specifically, the AP state (e.g., AP, $\downarrow$ channel if we consider combined spin channels, or the majority spin channel shown in the figure for the AP state) develops prominent hot spots that were absent or severely suppressed at zero bias. These new channels often appear as clear, bright regions (red / yellow) in the $\mathbf{k}_{\perp}$ -resolved maps. A dramatic increase in transmission efficiency is induced. Concurrently, while the P-state channels may still exhibit significant transmission, their relative dominance diminishes.

In some cases, the original P-state channels that were highly conductive at zero bias may become suppressed or shift out of the relevant bias window, or new P-state channels that are less efficient than the emerging AP-state channels may dominate. This differential evolution, namely the emergence of highly conductive channels in the AP state combined with a relative decline in P-state dominance, is the direct cause of the current crossover and subsequent TMR inversion. For example, comparing the "TRC AP up" maps in Fig. 4.8 at 0.1 V (top left) versus 0.5 V (bottom left) for the ASIC case clearly illustrates this. At 0.1 V, the AP transmission (red / yellow hot spots) is minimal. However, at 0.5 V, distinct and intense hot spots emerge, particularly in the corners and along certain diagonals, signifying newly activated high-transmission pathways. This is in contrast to the P-state (top left vs bottom left "TRC P up") which while still having some transmission, does not show the same relative enhancement.



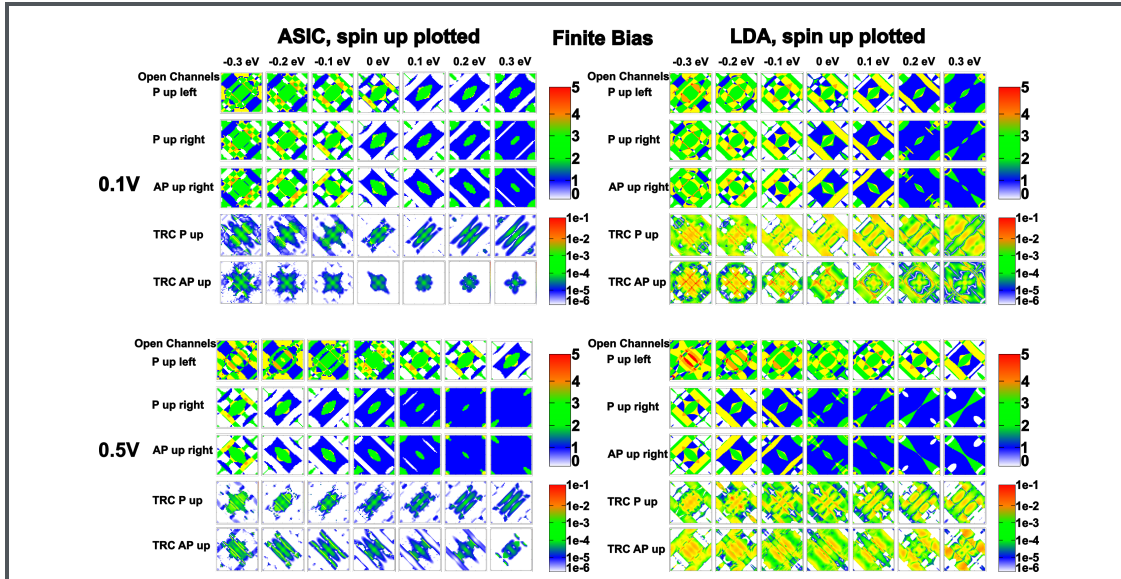


Figure 4.8. Microscopic origin of the TMR inversion in the ASIC model. The figure compares the $k_{\perp}$ -resolved transmission coefficients (*TRC*) for the ASIC (left) and LDA (right) models at low bias (0.1 V, top) and high bias (0.5 V, bottom). At 0.1 V, the P state in the ASIC model shows distinct transmission hot spots while the AP state transmission is negligible. At 0.5 V, after the inversion, new and intense transmission hot spots emerge for the AP state. These bias-activated channels make the AP state more conductive than the P state, causing the TMR to become negative.

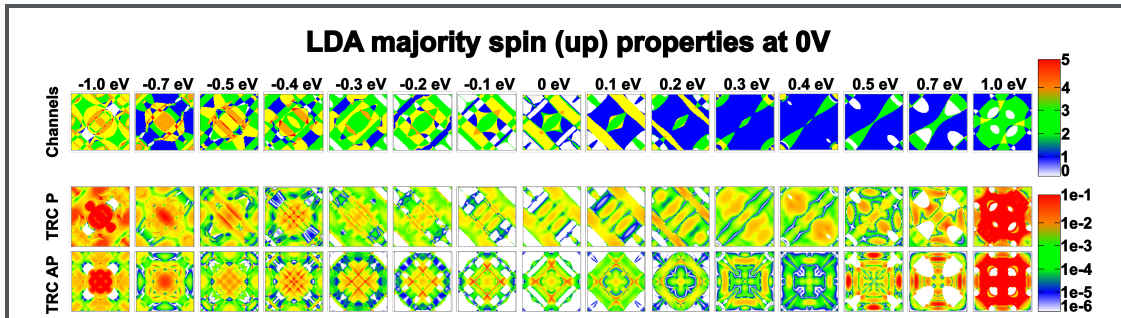


Figure 4.9. Energy-resolved $k_{\perp}$ transmission maps for the P and AP states in the LDA 8ML model, serving as a critical comparison to the ASIC results in Fig. 4.7. In contrast to the ASIC calculations, the LDA model shows a much smaller difference between P and AP transmission at the Fermi level ($E = 0$). More importantly, as energy increases, there is no emergence of new, intense transmission hot spots in the AP state. The transmission features remain diffuse and of low magnitude across the energy range, which microscopically explains why the LDA model fails to predict both a large zero-bias TMR and the giant TMR inversion observed with the more accurate ASIC method.

4.7.2 Bias-Induced Symmetry Filtering

The emergence of these spin-specific transmission hot spots is governed by the principle of symmetry filtering derived in foundational studies [121, 122]. The insulating TiO_2 barrier is not a simple potential wall; its evanescent states (the decaying wavefunctions

within the band gap) possess specific symmetries. For an electron to tunnel efficiently, its Bloch state in the electrode must match the symmetry of the most slowly decaying evanescent state in the barrier at that particular $\mathbf{k}_\perp$. Our analysis reveals a nuanced, bias-driven contribution to this effect. In altermagnets, the spin-splitting in the bulk electronic structure is momentum-dependent, leading to complex spin-momentum textures. At zero bias, the giant TMR arises because the spin-up states of the RuO_2 electrodes in the P configuration satisfy this symmetry-matching condition, creating the "P up" hot spots. In the AP state, neither spin channel at the Fermi level has the correct symmetry to couple to these efficient channels, leading to poor conduction. An applied finite bias V , alters the energy alignment between the electrodes. For the AP configuration, this has a spin-specific consequence. It shifts the energy bands of the emitting electrode relative to the collecting electrode. At the inversion voltage V_{inv} , the bias brings the minority band of the emitter into alignment with majority spin states in the collector that possess the favourable symmetry in $\mathbf{k}$ -space valleys different from the bulk regions of open channels required for efficient coupling to the barrier high-transmission channels. This bias-induced alignment of energy, momentum, and wavefunction symmetry unlocks the previously forbidden "AP down" tunnelling pathways. In essence, the voltage acts as a spin-selective switch. It activates a dormant, highly conductive spin-down channel in the AP configuration, causing its conductance to surge past the P-state conductance. This mechanism where a specific spin channel is toggled "on" by the bias voltage, is the fundamental driver of the giant TMR inversion. It represents a sophisticated interplay between altermagnetism, quantum tunnelling, and symmetry, highlighting how an electric field can be used to dramatically reconfigure the spin-dependent transport properties of these novel materials. The fact that this non-equilibrium channel-opening effect can be reproduced qualitatively within the minimal spin-valley tight-binding model of [45], where the Keldysh energy shift under bias shifts the emitter minority band into alignment with collector majority-spin valleys, provides a further layer of analytical validation for the first-principles result.

— 4.8 Limitations: The Pristine Interface Assumption —

The quantum transport results presented in this chapter were obtained within the framework of a ballistic, pristine lattice model. Both the structural model and the NEGF-DFT

formalism employed here assume perfectly ordered, atomically sharp interfaces with full translational periodicity in the transverse (x, y) plane. Under this assumption, the transverse crystal momentum $\mathbf{k}_\perp$ is a conserved quantum number throughout the device, and the transmission function factorizes as a sum over independent $\mathbf{k}_\perp$ channels. This is the foundational premise that enables the $\mathbf{k}_\perp$ -resolved analysis presented in Section 4.7.1 and, ultimately, the prediction of the giant TMR and its inversion. While the pristine model is the appropriate starting point for establishing a theoretical benchmark, it is essential to explicitly acknowledge the limitations that arise from the neglect of disorder, as this constitutes an important and potentially significant caveat.

The significance of this limitation is qualitatively different for the present system compared to more extensively studied junctions such as Fe/MgO/Fe. In the Fe/MgO case, the dominant contribution to the TMR arises from the highly symmetric Δ_1 band of Fe, whose evanescent state in MgO decays most slowly at and near the Γ point ($\mathbf{k}_\perp = 0$) [121, 122]. Because the Γ point corresponds to long-wavelength electronic states, a degree of self-averaging over short-range interface roughness naturally occurs: short-range disorder cannot efficiently scatter states with $\mathbf{k}_\perp \approx 0$, as this would require a large momentum transfer. Consequently, the giant TMR in Fe/MgO/Fe junctions is relatively robust against interface disorder.

The present $\text{RuO}_2 / \text{TiO}_2 / \text{RuO}_2$ system operates in a fundamentally different regime. As demonstrated by the $\mathbf{k}_\perp$ -resolved analysis in Section 4.7.1, the dominant transmission hot spots in the altermagnetic configuration do *not* reside near the Γ point. Instead, they are located at or near the Brillouin-zone boundaries, a direct consequence of the d -wave spin splitting in RuO_2 , which places the spin-polarized valleys at finite $\mathbf{k}_\perp$. States near a zone boundary correspond to short-wavelength Bloch waves that are highly sensitive to atomic-scale perturbations. Interface roughness, oxygen vacancies, or Ru/Ti intermixing, which is unavoidable in any realistic junction fabrication, can scatter electrons between these valleys via large- $\mathbf{k}_\perp$ transfers, directly disrupting the momentum-matched spin-filtering channels that underpin the predicted TMR. This represents a genuine and potentially significant limitation of the present pristine-interface model that must be acknowledged explicitly.

Quantifying these disorder effects rigorously would require going beyond the current computational framework. The most direct approach would be a supercell NEGF-DFT

calculation with a large lateral supercell (e.g., 10×10 or 20×20 primitive cells in the transverse plane), in which random disorder configurations are explicitly included and thermodynamically averaged over multiple realizations. An alternative is the coherent potential approximation (CPA) [123], which treats disorder in a mean-field fashion and can be integrated naturally into the Green's function formalism. Both approaches are computationally demanding but represent natural and necessary extensions of the present work. Until such calculations are performed, the predictions presented in this chapter should be understood as upper bounds on the achievable TMR, valid in the limit of a perfect single-crystalline device, a target that may be approached in high-quality epitaxially grown junctions but is not yet routinely achievable in experiment.

— 4.9 Limitations: The Non-Relativistic Approximation and Spin-Orbit Coupling —

All transport calculations in this chapter were performed within a fully non-relativistic NEGF-DFT framework, in which spin-orbit coupling (SOC) is neglected. This choice is physically motivated and scientifically justified, but it constitutes a second important limitation that must be stated explicitly.

The altermagnetic phase itself is a non-relativistic phenomenon. As discussed in Chapter 1, the momentum-dependent spin splitting that underpins the TMR mechanism arises entirely from the crystal-field potential and the rotational symmetry connecting opposite-spin sublattices [14]. SOC is a relativistic perturbation on top of this non-relativistic order. As noted in Section 1.4, the hierarchy of energy scales is clear: the primary non-relativistic spin splitting is of order hundreds of meV to 1 eV, while SOC-driven effects such as the Dzyaloshinskii-Moriya interaction are typically of order meV in most transition-metal oxides [9]. The non-relativistic approximation is therefore appropriate for capturing the dominant physics.

However, Ruthenium Ru with its 4d electrons is a moderately heavy element and the SOC in Ru, while smaller than the primary altermagnetic splitting, is not entirely negligible. Its practical importance for the $\text{RuO}_2 / \text{TiO}_2 / \text{RuO}_2$ junction can be assessed qualitatively. At the single-particle level, SOC introduces spin-mixing: states that are purely spin-up or

spin-down in the non-relativistic limit acquire a small admixture of the opposite spin. In the context of tunnelling transport, this spin-mixing would slightly reduce the spin-valley polarization of the RuO_2 Fermi surface and introduce small off-diagonal elements in the spin-resolved transmission matrix. The qualitative effect on the TMR would be a partial reduction of the peak value (since spin-mixing reduces the spin-polarization contrast between the P and AP transmission channels) and a possible shift of the inversion voltage V_{inv} , because the bias-induced alignment condition that activates the AP spin-down channel depends on the spin character of the emitter bands. The spin-filtering mechanism, rooted in the d -wave rotational symmetry and operative at the eV energy scale, would not be destroyed by SOC, but its efficiency would be quantitatively reduced.

Importantly, the SIESTA/SMEAGOL framework employed in this thesis is in principle capable of including SOC, as it supports a fully non-collinear spinor formalism in which the spin-orbit interaction is included via the relativistic pseudopotential [64, 65]. Such non-collinear NEGF-DFT calculations would allow for a direct, quantitative assessment of the SOC correction to the TMR and the inversion voltage. This represents a natural and important future extension of the present work. The non-relativistic predictions presented here should therefore be understood as the leading-order result in an expansion where SOC is treated perturbatively; this is a well-justified starting point for a system where the non-relativistic altermagnetic physics is the primary driver of the magnetoresistive response.

Outlook, Future work and Conclusion

Part IV

5 Outlook and Conclusion

5.1 Summary of Key Findings

This thesis has navigated a path from fundamental magnetic theory to applied device physics, yielding several key findings that collectively advance our understanding of altermagnetic spintronics. The primary results from each stage of this research are summarized below.

Firstly, this work established a rigorous theoretical foundation for altermagnetism as the third fundamental magnetic phase, as detailed in Chapter 1. By moving beyond the traditional framework of relativistic magnetic space groups and employing the more general theory of spin symmetry groups (see Section 1.2), this research clarified how altermagnets fundamentally differ from conventional antiferromagnets. The key distinction was identified in the nature of the symmetry operation connecting the opposite spin sublattices: a crystal rotation in altermagnets versus a time-reversal-partnered operation in antiferromagnets (Section 1.3). This was shown to be the origin of the primary signature of altermagnetism: a large, non-relativistic, momentum-dependent spin splitting of the electronic bands in a globally compensated magnet.

Secondly, the thesis confronted the timely and significant scientific controversy surrounding the intrinsic magnetism of RuO_2 , the prototypical altermagnet candidate. The extensive review and analysis in Chapter 3 highlighted the conflict between initial reports of magnetism and subsequent high-precision experiments finding none (Section 3.2).

Our own first-principles calculations demonstrated that standard DFT functionals fail to stabilize a magnetic moment, confirming the fragility of this state (Section 3.4.2). The crucial contribution here was the identification of a parameter-free Atomic Self-Interaction Correction (ASIC) scheme, applied selectively to the Ru atoms, as the closest method for modelling the altermagnetic phase (Section 3.4.4). This advanced computational model successfully reproduced the key features of the altermagnetic band structure, thereby providing a robust and justifiable foundation for the subsequent quantum transport investigations.

Thirdly, to bridge the gap between abstract theory and practical device engineering, this work developed and applied a systematic screening methodology based on a set of "golden rules" for designing viable altermagnetic tunnel junctions (Section 4.2). This framework, which combines symmetry analysis with materials database mining under strict physical and structural constraints, provided a rational pathway for identifying promising heterostructures. The application of these rules successfully validated the $\text{RuO}_2 / \text{TiO}_2 / \text{RuO}_2$ system as an excellent prototypical candidate and identified the high-temperature CrSb / NiTe junction as a compelling target for future research, while simultaneously demonstrating the non-viability of other candidates like CrSe due to a lack of suitable insulators (Section 4.3).

Finally, the quantum transport investigation of the $\text{RuO}_2 / \text{TiO}_2 / \text{RuO}_2$ ALMTJ, presented in Chapter 4, yielded the central and most novel findings of this thesis. At zero bias, our parameter-free ASIC calculations predicted a giant tunnelling magnetoresistance (TMR) reaching over 400%, independently confirming the potential of this system for high-performance memory applications (Section 4.5). More significantly, the investigation under operational conditions revealed a remarkable non-equilibrium phenomenon: a giant, bias-induced TMR inversion (Section 4.6). The TMR was shown to systematically switch from large positive to large negative values at a critical voltage of approximately 0.35 V. The microscopic origin of this effect was uncovered through a $\mathbf{k}_\perp$ -resolved analysis and attributed to a bias-induced, spin-selective activation of a dormant tunnelling channel, a sophisticated manifestation of symmetry filtering in a non-equilibrium altermagnetic system (Section 4.7).

5.2 Outlook and Future Research Directions

The findings presented in this thesis, while providing resolution to specific theoretical questions, simultaneously open several new and compelling avenues for future research. The confirmation of a giant TMR effect and the prediction of its inversion in an ideal altermagnetic junction underscore the rich physics available in these systems, motivating further exploration on both the experimental and theoretical fronts. This section outlines the most promising of these future directions, which build directly upon the foundations laid herein.

5.2.1 Experimental Directions

The most pressing challenge highlighted by this research is bridging the gap between the theoretical promise of altermagnetic RuO_2 and its contested experimental reality. The fragility of its magnetic ground state, explored in Chapter 3, suggests that future experimental work must move from characterizing pristine bulk crystals to actively engineering the altermagnetic phase in thin films. A targeted investigation into the role of epitaxial strain, as suggested by the reconciliation hypotheses in Section 3.2.4, is a primary research direction. By growing high-quality RuO_2 thin films on a variety of substrates designed to induce specific compressive or tensile strains, it may be possible to stabilize the altermagnetic order.

Should such stable altermagnetic films be realized, the direct experimental verification of the transport phenomena predicted in Chapter 4 becomes the subsequent critical goal. The fabrication of $\text{RuO}_2 / \text{TiO}_2 / \text{RuO}_2$ devices, following the structural blueprint laid out in Section 4.4, would be paramount. Low-temperature quantum transport measurements on these devices would serve two purposes. First, to search for the giant positive TMR predicted at low bias (Section 4.5). Second, and more compellingly, to perform a voltage-dependent sweep to search for the characteristic TMR inversion signature at the predicted critical voltage of approximately 0.35 V (Section 4.6). The experimental observation of this inversion would provide powerful validation for the advanced ASIC-based transport model and the underlying physics of bias-induced symmetry filtering detailed in this work.

5.2.2 Theoretical Directions

The computational framework established in this thesis can be extended to explore several important unanswered questions. A natural and immediate next step is to leverage the "golden rules" methodology from Section 4.2 and apply the same in-depth NEGF-DFT analysis to other promising candidates. The high-Néel-temperature CrSb / NiTe system, identified in Section 4.3, is a particularly attractive target. Such a study would reveal whether giant TMR and its inversion are general features of ALMTJs or specific quirks of the RuO₂ system, thereby broadening the fundamental understanding of this device class.

Furthermore, this thesis focused exclusively on the "read" mechanism (TMR) of the spintronic device. A complete assessment requires an understanding of the "write" mechanism, which is governed by spin-transfer torques (STT). The existing NEGF-DFT framework can be extended to calculate the bias-dependent field-like and damping-like components of the STT within the RuO₂ / TiO₂ / RuO₂. This would allow for an estimation of the critical current density required to switch the Néel vector, providing a crucial metric for the device's potential in MRAM applications.

Finally, the models investigated herein assumed perfect, atomically sharp interfaces. Future theoretical work should incorporate more realistic features, such as the effects of interfacial disorder, atomic intermixing, and the presence of point defects like oxygen vacancies, whose potential importance was highlighted in Section 3.2.4 and Section 4.8. Such simulations would provide a more complete picture of device performance and robustness in scenarios closer to experimental reality.

An open frontier that merits dedicated investigation is the role of dimensionality in the altermagnetic phase. This thesis has exclusively studied three-dimensional bulk-like electrodes. However, a compelling question arises: can altermagnetism be realized and exploited in two-dimensional (2D) and quasi-2D material systems? In three dimensions, the altermagnetic phase is defined by a specific set of magnetic space groups where opposite-spin sublattices are connected by crystal rotations (Section 1.2). Upon reducing to a single layer or a few-layer structure, the relevant space group changes: the bulk inversion center is eliminated along with the third dimension, fundamentally altering the set of allowed crystal symmetries. Moreover, the Mermin-Wagner theorem [124] forbids spontaneous long-range magnetic order in two-dimensional isotropic systems with short-range interac-

tions, imposing a stringent thermodynamic constraint on any 2D altermagnetic candidate. This dimensional reduction may either suppress the altermagnetic phase (if the relevant rotation symmetry becomes impermissible in 2D) or potentially stabilize it (if the reduced dimensionality enhances the separation of opposite-spin sublattices by lifting degeneracies present in the bulk). The interplay of confinement and magnetic fragility, already discussed for RuO₂ thin films in Section 3.2.4, suggests that dimensionality is a powerful tuning parameter for the altermagnetic phase [28]. Beyond bulk-like thin films, a growing family of van der Waals layered magnets can be exfoliated to the single-layer limit [42, 125]. Identifying which among these layered materials satisfy the spin-group requirements for altermagnetism in their monolayer form, and predicting the spin splitting and transport properties thereof, represents a natural next step in the theoretical exploration of this field. The prospect of integrating 2D altermagnets into van der Waals heterostructures with atomically sharp interfaces, which are inherently clean and disorder-free in a way that epitaxial 3D junctions are not, is particularly attractive, as it could avoid some of the disorder limitations outlined in Section 4.8.

5.2.3 Long-Term Vision

Looking beyond the immediate next steps, the unique constellation of properties offered by altermagnets positions them as a potentially transformative platform for information technology. The ability to generate and utilize highly spin-polarized currents without the drawback of stray magnetic fields, as explored in Chapter 1, combined with the intrinsic terahertz dynamics of compensated magnets, addresses several of the key limitations of conventional ferromagnet-based spintronics.

If the materials science and fabrication challenges, such as those detailed for RuO₂, can be surmounted, the design principles and novel physical phenomena explored in this thesis could become foundational. The bias-induced TMR inversion, for instance, hints at the possibility of creating multi-level memory cells or reconfigurable logic-in-memory devices where the device function is controlled by the operating voltage. Ultimately, altermagnetic spintronics represents a tangible and promising pathway toward a new generation of non-volatile, high-speed, and ultra-dense memory and logic technologies, contributing to a future that moves beyond the scaling limits of conventional electronics.

An intriguing speculative direction opened by the altermagnetic framework is the possible existence of a phenomenon one might term *alterplasmonics*. Plasmons are collective,

quantized oscillations of the conduction electron density [126]. In a conventional metal, plasmons are charge-symmetric: both spin channels contribute equally, and the plasmon dispersion is determined by the total electron density. However, in an altermagnet, the conduction electrons occupy spin-split, momentum-dependent valleys. By analogy with the spin-valley filtering described throughout this thesis, where the alternating spin polarization of the Fermi surface enables spin-selective tunnelling, one may ask whether collective charge excitations (plasmons) in an altermagnetic material could acquire a valley-selective, momentum-dependent character. Such "altermagnetic plasmons" would be distinct from both spin plasmons in ferromagnets (which are split by the uniform exchange field) and from conventional metal plasmons (which are spin-degenerate). Their dispersion could be sensitive to the relative Néel vector orientation in the same way that the TMR is, opening the possibility of magnetically tunable plasmonic devices operating in the terahertz regime, the natural dynamics scale of antiferromagnets. Whether alterplasmonics constitutes a robust new physical effect or remains a weak perturbation to conventional plasmonic behaviour is an open question for future theoretical and experimental investigation. Nevertheless, the analogy between spin-momentum locking in the single-particle band structure and momentum-dependent collective oscillations in the many-body spectrum represents a compelling frontier at the intersection of altermagnetism and plasmonics.

A List of 2D and 3D Altermagnet Candidates

Table A.1: A comprehensive list of candidate materials predicted or observed to host an altermagnetic phase, compiled from the literature [42]. The table includes the non-magnetic space group (NSG), the magnetic space group (MSG) where available, and the reported Néel temperature. RT denotes room temperature. Image reprinted with permission from Ref. [42] under Wiley advanced license number 6086980370462.

No.	Compound	NSG	MSG	Néel temp. [K]
1	KV ₂ Se ₂ O	P4/mmm (123)	P4'/mm'm	≥ RT
2	Rb _{1-δ} V ₂ Te ₂ O	P4/mmm (123)	P4'/mm'm	≥ 307
3	RuO ₂	P4 ₂ /mnm (136)	P4' ₂ /mnm'	> 300
4	Mn ₅ Si ₃	P6 ₃ /mcm (193)	P $\bar{1}$	240
5	CrSb	P6 ₃ /mmc (194)	P6' ₃ /m'm'm'c	> 600
6	MnTe	P6 ₃ /mmc (194)	Cm'c'm	310
7	LiFeP ₂ O ₇	P2 ₁ (4)	P2 ₁	22
8	CaLaFeAgO ₆	Pc (7)	—	—
9	Mn ₄ Nb ₂ O ₉	Cc (9)	Cc	52.1(1)
10	CaFe ₅ O ₇	P2 ₁ /m (11)	P2' ₁ /m'	125
11	Fe ₃ F ₈ (H ₂ O) ₂	C2/m (12)	C2'/m'	157

Continued on next page

Table A.1 – continued from previous page

No.	Compound	NSG	MSG	Néel temp. [K]
12	RbMnF ₄	P2 ₁ /a (14)	P $\bar{1}$	3.7(1)
13	Li ₂ Co(SO ₄) ₂	P2 ₁ /c (14)	P2 ₁ '/c'	7–8
14	LiFe(SO ₄) ₂	P2 ₁ /c (14)	P2 ₁ /c	35–39
15	Li ₂ Mn(SO ₄) ₂	P2 ₁ /c (14)	P2 ₁ /c	6
16	Fe ₃ (PO ₄) ₂ (OH) ₂	P2 ₁ /c (14)	P2 ₁ /c	160
17	CuF ₂	P2 ₁ /c (14)	—	69
18	Li ₂ Ni(SO ₄) ₂	P2 ₁ /c (14)	P2 ₁ /c	15
19	K ₂ ReI ₆	P2 ₁ /n (14)	P2 ₁ /c	24
20	Sr ₂ CoTeO ₆	P2 ₁ /n (14)	P2 ₁ /c	15–19
21	Sr ₂ ScOsO ₆	P2 ₁ /n (14)	P2 ₁ /c	92
22	Sr ₂ YRuO ₆	P2 ₁ /n (14)	P2 ₁ /c	26
23	Sr ₂ TbIrO ₆	P2 ₁ /n (14)	P2 ₁ /c	51
24	Sr ₂ LuRuO ₆	P2 ₁ /n (14)	P2 ₁ /c	30(1)
25	Sr ₂ YbRuO ₆	P2 ₁ /n (14)	P2 ₁ /c	36
26	Sr ₂ TmRuO ₆	P2 ₁ /n (14)	P2 ₁ /c	36
27	Sr ₂ TbRuO ₆	P2 ₁ /n (14)	P2 ₁ /c	41
28	Sr ₂ HoRuO ₆	P2 ₁ /n (14)	P2 ₁ /c	36
29	Sr ₂ DyRuO ₆	P2 ₁ /n (14)	P2 ₁ /c	39.5
30	Mn ₂ ScSbO ₆	P2 ₁ /n (14)	P2 ₁ /c	22.3
31	CuAg(SO ₄) ₂	P2 ₁ /n (14)	—	—
32	La ₂ LiRuO ₆	P2 ₁ /n (14)	P2 ₁ /c	30
33	Sr ₂ Co _{0.9} Mg _{0.1} TeO ₆	P2 ₁ /n (14)	P2 ₁ /c	13
34	Sr ₂ CoOsO ₆	B2/c (15)	C2/c	108
35	Ba ₃ CoIr ₂ O ₉	C2/c (15)	C2/c	107
36	FeSO ₄ F	C2/c (15)	C2'/c'	100
37	FeOHSO ₄	C2/c (15)	C2'/c'	125
38	BiCrO ₃	C2/c (15)	C2/c	—
39	BaCrF ₅	P2 ₁ 2 ₁ 2 ₁ (19)	P2 ₁ '2 ₁ '2 ₁	3.4
40	FeHO ₂	Pmn2 ₁ (31)	—	> RT
41	CaLaCr ₂ O ₆	Pmn2 ₁ (31)	—	—

Continued on next page

Table A.1 – continued from previous page

No.	Compound	NSG	MSG	Néel temp. [K]
42	Y ₂ Cu ₂ O ₅	Pna2 ₁ (33)	Pna2 ₁	13
43	NaFeO ₂	Pna2 ₁ (33)	—	—
44	[C(ND ₂) ₃]Cu(DCOO) ₃	Pna2 ₁ (33)	Pn'a'2 ₁	—
45	[C(ND ₂) ₃]Cu(DCOO) ₃	Pna2 ₁ (33)	Pna2 ₁	—
46	Ca ₃ Mn ₂ O ₇	Cmc2 ₁ (36)	Cm'c2 ₁ '	115
47	Ca ₃ Cr ₂ O ₇	Cmc2 ₁ (36)	—	—
48	ErGe _{1.83}	Cmc2 ₁ (36)	Cmc2 ₁	6
49	Cu ₂ V ₂ O ₇	Fdd2 (43)	Fd'd'2	33.4(1)
50	ZnFeF ₅ (H ₂ O) ₂	Imm2 (44)	Imm2	9(2)
51	Sr ₂ MnGaO ₅	Ima2 (46)	Im'a2'	180
52	BiFe _{0.5} Sc _{0.5} O ₃	Ima2 (46)	Im'a2'	≈220
53	[C(ND ₂) ₃]Co(DCOO) ₃	Pnna (52)	Pn'na'	14.04(1)
54	[C(ND ₂) ₃]Mn(DCOO) ₃	Pnna (52)	Pn'n'a	8.73(2)
55	Fe _{1.5} Mn _{1.5} BO ₅	Pbam (55)	Pbam	100
56	Mn(N(CN) ₂) ₂	Pnnm (58)	Pnn'm'	16
57	FeSb ₂	Pnnm (58)	Pnn'm'	—
58	CrSb ₂	Pnnm (58)	—	273
59	Eu ₃ In ₂ As ₄	Pnnm (58)	—	—
60	Fe ₂ WO ₆	Pbcn (60)	Pbc'n'	—
61	Ca ₂ RuO ₄	Pbca (61)	—	—
62	CaCrO ₃	Pbnm (62)	Pb'n'm	90
63	YCrO ₃	Pbnm (62)	Pn'ma'	141
64	TbCrO ₃	Pbnm (62)	Pn'm'a	158
65	DyCrO ₃	Pbnm (62)	Pn'm'a	—
66	ErCrO ₃	Pbnm (62)	Pn'ma'	133
67	CeFeO ₃	Pbnm (62)	Pnma	220
68	CeFeO ₃	Pbnm (62)	Pn'ma'	720
69	SmFeO ₃	Pbm _n (62)	Pn'm'a	480
70	SmFeO ₃	Pnma (62)	Pn'ma'	675
71	TbFeO ₃	Pbnm (62)	Pn'ma'	681

Continued on next page

Table A.1 – continued from previous page

No.	Compound	NSG	MSG	Néel temp. [K]
72	DyFeO ₃	Pbnm (62)	Pnma	73
73	DyFeO ₃	Pbnm (62)	Pn'ma'	—
74	ErVO ₃	Pbnm (62)	P2 ₁ /c	56
75	NdCoO ₃	Pbnm (62)	Pnma	1.20(1)
76	TbCr _{0.5} Mn _{0.5} O ₃	Pbnm (62)	Pn'ma'	≈84
77	κ-Cl	Pnma (62)	Pn'ma'	23
78	YVO ₃	Pnma (62)	Pn'm'a	77
79	NdVO ₃	Pnma (62)	P2 ₁ '/m'	135
80	LaCrO ₃	Pnma (62)	Pnma	289
81	LaCrO ₃	Pnma (62)	Pn'ma'	—
82	ScCrO ₃	Pnma (62)	Pnma	73
83	InCrO ₃	Pnma (62)	Pnma	93
84	TlCrO ₃	Pnma (62)	Pnma	89
85	ZrCrO ₃	Pnma (62)	—	—
86	YRuO ₃	Pnma (62)	Pn'ma'	97
87	LaMnO ₃	Pnma (62)	Pn'ma'	139.5
88	CaMnO ₃	Pnma (62)	—	120
89	Ca _{1-x} Ce _x MnO ₃	Pnma (62)	—	—
90	NdMnO ₃	Pnma (62)	P2 ₁ /m	85
91	PrMnO ₃	Pnma (62)	Pn'ma'	91
92	LaFeO ₃	Pnma (62)	Pn'ma'	750
93	NdFeO ₃	Pnma (62)	Pn'ma'	760
94	NaOsO ₃	Pnma (62)	Pn'ma'	411.(2)
95	Fe ₂ PO ₅	Pnma (62)	Pnma	250
96	NiFePO ₅	Pnma (62)	Pnma	178(5)
97	CuFePO ₅	Pnma (62)	Pnma	195(5)
98	Mn ₂ SeO ₃ F ₂	Pnma (62)	Pn'ma'	26
99	MnO ₂	Pnma (62)	—	—
100	KMnF ₃	Pnma (62)	Pn'ma'	75
101	Ca ₂ MnGaO ₅	Pnma (62)	Pnm'a'	160

Continued on next page

Table A.1 – continued from previous page

No.	Compound	NSG	MSG	Néel temp. [K]
102	$\text{Ca}_2\text{PrCr}_2\text{TaO}_9$	Pnma (62)	$\text{Pn}'\text{m}'\text{a}$	130
103	$\text{Ca}_2\text{PrCr}_2\text{NbO}_9$	Pnma (62)	$\text{Pn}'\text{m}'\text{a}$	110
104	$\text{Ho}_{0.2}\text{Bi}_{0.8}\text{FeO}_3$	Pnma (62)	$\text{Pn}'\text{ma}'$	—
105	$\text{Ho}_{0.15}\text{Bi}_{0.85}\text{FeO}_3$	Pnma (62)	$\text{Pn}'\text{ma}'$	—
106	$\text{La}_{0.95}\text{Ba}_{0.05}\text{Mn}_{0.95}\text{Ti}_{0.05}\text{O}_3$	Pnma (62)	$\text{Pn}'\text{ma}'$	—
107	$\text{La}_{0.75}\text{Bi}_{0.25}\text{Fe}_{0.5}\text{Cr}_{0.5}\text{O}_3$	Pnma (62)	Pnma	350
108	$\text{La}_{0.5}\text{Sr}_{0.5}\text{FeO}_{2.5}\text{F}_{0.5}$	Pnma (62)	$\text{Pn}'\text{ma}'$	—
109	$\text{BiFe}_{0.5}\text{Sc}_{0.5}\text{O}_3$	Pnma (62)	$\text{Pn}'\text{m}'\text{a}$	≈ 220
110	$\text{Bi}_{0.85}\text{Ca}_{0.15}\text{Fe}_{0.55}\text{Mn}_{0.45}\text{O}_3$	Pnma (62)	$\text{Pn}'\text{m}'\text{a}$	—
111	$\text{Ca}_2\text{Fe}_{0.875}\text{Cr}_{0.125}\text{GaO}_5$	Pnma (62)	$\text{Pn}'\text{m}'\text{a}$	—
112	$\text{Pb}_2\text{Mn}_{0.6}\text{Co}_{0.4}\text{WO}_6$	Pmcn (62)	$\text{Pm}'\text{c}2_1'$	9
113	NiCrO_4	Cmcm (6 ₃)	Cmcm	—
114	$\text{Tb}_2\text{Ir}_3\text{Ga}_9$	Cmcm (6 ₃)	$\text{Cm}'\text{cm}'$	12.5
115	CaIrO_3	Cmcm (6 ₃)	$\text{Cm}'\text{cm}'$	115
116	CoFe_3O_5	Cmcm (6 ₃)	$\text{Cm}'\text{cm}'$	≈ 300
117	Nd_2PdGe_6	Cmce (64)	$\text{Cm}'\text{c}'\text{a}$	4
118	LaCaFeO_4	Cmce (64)	$\text{Cm}'\text{c}'\text{a}$	—
119	$\text{Sr}_4\text{Fe}_4\text{O}_{11}$	Cmmm (65)	$\text{Cmm}'\text{m}'$	232(4)
120	$\text{La}_2\text{NiO}_3\text{F}_2$	Cccm (66)	—	49
121	$\text{YBaMn}_2\text{O}_{5.5}$	Ibam (72)	$\text{C}2/\text{m}$	≈ 140
122	$\text{YBaMn}_2\text{O}_{5.5}$	Ibam (72)	$\text{Ib}'\text{a}'\text{m}$	≈ 120
123	$\text{Y}_2\text{SrCu}_{0.6}\text{Co}_{1.4}\text{O}_{6.5}$	Ibam (72)	$\text{Ib}'\text{a}'\text{m}$	385
124	$\text{Bi}_{0.8}\text{La}_{0.2}\text{Fe}_{0.5}\text{Mn}_{0.5}\text{O}_3$	Imma (74)	$\text{Imm}'\text{a}'$	≈ 240
125	$\alpha\text{-MnO}_2$	I4/m (87)	—	—
126	KRu_4O_8	I4/m (87)	—	—
127	$\text{K}_y\text{Fe}_{2-x}\text{Se}_2$	I4/m (87)	$\text{C}2'/\text{m}'$	—
128	$\text{Rb}_y\text{Fe}_{2-x}\text{Se}_2$	I4/m (87)	$\text{C}2'/\text{m}'$	—
129	NaFeO_2	P412 ₁ 2 (92)	—	—
130	GdAlSi	I41md (109)	$\text{I}41'\text{m}'\text{d}$	32
131	$\text{SrMn}_2\text{V}_2\text{O}_8$	I41cd (110)	$\text{Ib}'\text{a}2'$	42.2

Continued on next page

Table A.1 – continued from previous page

No.	Compound	NSG	MSG	Néel temp. [K]
132	Ba ₂ MnSi ₂ O ₇	P $\bar{4}$ 2 ₁ m (113)	P $\bar{4}$ '2 ₁ m'	2.95
133	Ba ₂ CoGe ₂ O ₇	P $\bar{4}$ 2 ₁ m (113)	Cm'm2'	6.7
134	Ca ₂ CoSi ₂ O ₇	P $\bar{4}$ 2 ₁ m (113)	P2 ₁ 2 ₁ '2'	5.7
135	CsCoF ₄	I $\bar{4}$ c2 (120)	I $\bar{4}$ 'c2'	54
136	CuFeS ₂	I $\bar{4}$ 2d (122)	I $\bar{4}$ 2d	> RT
137	UCr ₂ Si ₂ C	P4/mmm (123)	Pm'm'm	—
138	ZrMn ₂ Ge ₄ O ₁₂	P4/nbm (125)	P4'/nbm'	8
139	CeMn ₂ Ge ₄ O ₁₂	P4/nbm (125)	P4'/nbm'	7.6
140	CeMnCoGe ₄ O ₁₂	P4/nbm (125)	Pb'an'	5.8
141	Nb ₂ FeB ₂	P4/mbm (127)	P4/mb'm'	—
142	Ta ₂ FeB ₂	P4/mbm (127)	P4/mb'm'	—
143	NdB ₂ C ₂	P4/mbm (127)	—	8.8
144	Mg ₂ FeIr ₅ B ₂	P4/mbm (127)	—	—
145	Mg ₂ MnIr ₅ B ₂	P4/mbm (127)	—	—
146	Mg ₂ NiIr ₅ B ₂	P4/mbm (127)	—	—
147	Sc ₂ MnIr ₅ B ₂	P4/mbm (127)	—	—
148	MnF ₂	P4 ₂ /mnm (136)	P4' ₂ /mnm'	67.4
149	CoF ₂	P4 ₂ /mnm (136)	P4' ₂ /mnm'	37.7
150	NiF ₂	P4 ₂ /mnm (136)	Pnn'm'	73.2
151	ReO ₂	P4 ₂ /mnm (136)	P4' ₂ /mnm'	—
152	β -MnO ₂	P4 ₂ /mnm (136)	—	—
153	LiFe ₂ F ₆	P4 ₂ /mnm (136)	P4' ₂ /mnm'	105
154	La ₂ NiO ₄	P4 ₂ /ncm (138)	Pc'c'n	80
155	La ₂ O ₃ FeMnSe ₂	I4/mmm (139)	Im'm'm	76
156	Sr _{0.7} Tb _{0.3} CoO _{2.9}	I4/mmm (139)	I4'/mmm'	≈300
157	Sr _{0.7} Er _{0.3} CoO _{2.8}	I4/mmm (139)	I4'/mmm'	≈300
158	Sr _{0.7} Ho _{0.3} CoO _{2.7}	I4/mmm (139)	I4'/mmm'	≈300
159	KMnF ₃	I4/mcm (140)	I4/mcm	86.8
160	β -Fe ₂ (PO ₄)O	I41/amd (141)	I41'/am'd	408
161	Co ₂ (PO ₄)O	I41/amd (141)	I41'/am'd	—

Continued on next page

Table A.1 – continued from previous page

No.	Compound	NSG	MSG	Néel temp. [K]
162	Cr ₂ S ₃	R $\bar{3}$ (148)	P $\bar{1}$	—
163	MnTiO ₃	R3c (161)	Cc'	28
164	PbNiO ₃	R3c (161)	R3c	205
165	ZrMnO ₃	R3c (161)	—	—
166	GaFeO ₃	R3c (161)	Cc'	408
167	ScFeO ₃	R3c (161)	Cc'	360(5)
168	Ho _{0.1} Bi _{0.9} FeO ₃	R3c (161)	R3c	—
169	Ho _{0.05} Bi _{0.95} FeO ₃	R3c (161)	R3c	—
170	K _{1.62} Fe ₄ O _{6.62} (OH) _{0.38}	P $\bar{3}$ 1c (163)	P $\bar{3}$ 1c	—
171	MnCO ₃	R $\bar{3}$ c (167)	C2/c	31.5
172	FeBO ₃	R $\bar{3}$ c (167)	C2'/c'	348
173	FeCO ₃	R $\bar{3}$ c (167)	R $\bar{3}$ c	38
174	NiCO ₃	R $\bar{3}$ c (167)	C2/c	24.25
175	CoCO ₃	R $\bar{3}$ c (167)	C2/c	18.1
176	CoF ₃	R $\bar{3}$ c (167)	R $\bar{3}$ c	460
177	FeF ₃	R $\bar{3}$ c (167)	C2'/c'	394
178	CrF ₃	R $\bar{3}$ c (167)	—	—
179	NiF ₃	R $\bar{3}$ c (167)	—	—
180	VF ₃	R $\bar{3}$ c (167)	—	—
181	α -Fe ₂ O ₃	R $\bar{3}$ c (167)	—	259.1(2)
182	α -Fe ₂ O ₃	R $\bar{3}$ c (167)	C2'/c'	955
183	Ca ₃ LiRuO ₆	R $\bar{3}$ c (167)	C2'/c'	117.0(8)
184	Sr ₃ LiRuO ₆	R $\bar{3}$ c (167)	C2'/c'	90
185	Sr ₃ NaRuO ₆	R $\bar{3}$ c (167)	C2'/c'	70
186	Ca ₃ LiOsO ₆	R $\bar{3}$ c (167)	C2'/c'	117.1(9)
187	LaCrO ₃	R $\bar{3}$ c (167)	R $\bar{3}$ c	—
188	Ca ₃ Co _{2-x} Mn _x O ₆ ($x \approx 0.96$)	R $\bar{3}$ c (167)	R3c	16.5
189	La _{0.33} Sr _{0.67} FeO ₃	R $\bar{3}$ c (167)	C2/c	200
190	CoNb ₃ S ₆	P6 ₃ 22 (182)	C2'2'2 ₁	25
191	VNb ₃ Sb ₆	P6 ₃ 22 (182)	C2'2'2 ₁	50

Continued on next page

Table A.1 – continued from previous page

No.	Compound	NSG	MSG	Néel temp. [K]
192	Fe ₂ Mo ₃ O ₈	P6 ₃ mc (186)	P6 ₃ 'm'c	59(3)
193	Co ₂ Mo ₃ O ₈	P6 ₃ mc (186)	P6 ₃ 'm'c	42(3)
194	MnO	P6 ₃ mc (186)	—	—
195	MnSe	P6 ₃ mc (186)	—	—
196	UNiGa	P $\bar{6}$ 2m (189)	P $\bar{6}$ 2m'	38
197	FeS	P $\bar{6}$ 2c (190)	P $\bar{6}$ '2c'	—
198	CsCr ₃ Sb ₅	P6/mmm (191)	—	—
199	BaMnO ₃	P6 ₃ /mmc (194)	P6 ₃ '/m'cm'	2.3
200	CrNb ₄ S ₈	P6 ₃ /mmc (194)	P6 ₃ '/m'm'c	—
201	Ba ₃ NiRu ₂ O ₉	P6 ₃ /mmc (194)	P6 ₃ '/m'm'c	—
202	Ba ₅ Co ₅ ClO ₁₃	P6 ₃ /mmc (194)	P6 ₃ '/m'm'c	110
203	MnSe	P6 ₃ /mmc (194)	—	120(10)
204	CaMnN ₂	P6 ₃ /mmc (194)	—	—
205	EuIn ₂ As ₂	P6 ₃ /mmc (194)	—	—
206	CsNiCl ₃	P6 ₃ /mmc (194)	C22'2 ₁ '	4.85
207	CsCoCl ₃	P6 ₃ /mmc (194)	P6 ₃ '/m'cm'	20.82
208	RbCoBr ₃	P6 ₃ /mmc (194)	P6 ₃ '/m'cm'	36
209	CoNb ₄ Se ₈	P6 ₃ /mmc (194)	—	168
210	MnTeLi _{0.003}	P6 ₃ /mmc (194)	C2'/m'	307
211	MnSe ₂	Pa $\bar{3}$ (205)	Pca'	49
212	MnSe ₂	Pa $\bar{3}$ (205)	Pbca	—
213	Co ₂ Mo ₃ N	P4132 (213)	C22'2 ₁ '	—
214	Ce ₄ Sb ₃	I $\bar{4}$ 3d (220)	I $\bar{4}$ 2d'	—
215	Ce ₄ Ge ₃	I $\bar{4}$ 3d (220)	I $\bar{4}$ 2d	≈5
216	Tb ₂ C ₃	I $\bar{4}$ 3d (220)	Fd'd2'	33(4)
217	LiTi ₂ O ₄	Fd $\bar{3}$ m (227)	I41'/am'd	—
218	Bi ₂ RuMnO ₇	Fd $\bar{3}$ m (227)	Fd'd'd	20
219	Er ₂ Ru ₂ O ₇	Fd $\bar{3}$ m (227)	I41'/am'd	90
220	La ₂ NiO ₄	Bmab	—	770
221	La ₂ CuO ₄	Bmab	Pccn	316(2)

B Table of Spin-splitting Types 4 & 5 Magnetic Space Groups

[illegible]



Bibliography

- [1] A. Hirohata, K. Yamada, Y. Nakatani, I.-L. Prejbeanu, B. Diény, P. Pirro, and B. Hillebrands, “Review on spintronics: Principles and device applications,” *Journal of Magnetism and Magnetic Materials*, vol. 509, p. 166711, Sept. 2020.
- [2] M. N. Baibich, J. M. Broto, A. Fert, F. N. Van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friederich, and J. Chazelas, “Giant magnetoresistance of (001)fe/(001)cr magnetic superlattices,” *Phys. Rev. Lett.*, vol. 61, pp. 2472–2475, Nov 1988.
- [3] G. Binasch, P. Grünberg, F. Saurenbach, and W. Zinn, “Enhanced magnetoresistance in layered magnetic structures with antiferromagnetic interlayer exchange,” *Phys. Rev. B*, vol. 39, pp. 4828–4830, Mar 1989.
- [4] C. Chappert, A. Fert, and F. N. Van Dau, “The emergence of spin electronics in data storage,” *Nature Materials*, vol. 6, pp. 813–823, Nov. 2007. Number: 11 Publisher: Nature Publishing Group.
- [5] M. Julliere, “Tunneling between ferromagnetic films,” *Physics Letters A*, vol. 54, no. 3, pp. 225–226, 1975.
- [6] S. S. P. Parkin, C. Kaiser, A. Panchula, P. M. Rice, B. Hughes, M. Samant, and S.-H. Yang, “Giant tunnelling magnetoresistance at room temperature with mgo (100) tunnel barriers,” *Nature Materials*, vol. 3, pp. 862–867, Dec 2004.
- [7] S. Yuasa, T. Nagahama, A. Fukushima, Y. Suzuki, and K. Ando, “Giant room-temperature magnetoresistance in single-crystal fe/mgo/fe magnetic tunnel junctions,” *Nature materials*, vol. 3, pp. 868–71, 01 2005.
- [8] T. Jungwirth, X. Marti, P. Wadley, and J. Wunderlich, “Antiferromagnetic spintronics,” *Nature Nanotechnology*, vol. 11, pp. 231–241, Mar. 2016. Number: 3 Publisher: Nature Publishing Group.
- [9] V. Baltz, A. Manchon, M. Tsoi, T. Moriyama, T. Ono, and Y. Tserkovnyak, “Antiferromagnetic spintronics,” *Reviews of Modern Physics*, vol. 90, p. 015005, Feb. 2018. arXiv:1606.04284 [cond-mat].
- [10] T. Jungwirth, J. Sinova, A. Manchon,

- X. Marti, J. Wunderlich, and C. Felser, “The multiple directions of antiferromagnetic spintronics,” *Nature Physics*, vol. 14, pp. 200–203, Mar. 2018. Number: 3 Publisher: Nature Publishing Group.
- [11] B. G. Park, J. Wunderlich, X. Martí, V. Holý, Y. Kurosaki, M. Yamada, H. Yamamoto, A. Nishide, J. Hayakawa, H. Takahashi, A. B. Shick, and T. Jungwirth, “A spin-valve-like magnetoresistance of an antiferromagnet-based tunnel junction,” *Nature Materials*, vol. 10, pp. 347–351, May 2011. Number: 5 Publisher: Nature Publishing Group.
- [12] L. Šmejkal, J. Sinova, and T. Jungwirth, “Altermagnetism: spin-momentum locked phase protected by non-relativistic symmetries.” arXiv:2105.05820, Oct. 2021. arXiv:2105.05820. Published version: [14].
- [13] L. Šmejkal, J. Sinova, and T. Jungwirth, “Emerging research landscape of altermagnetism,” *Physical Review X*, vol. 12, p. 040501, Dec. 2022. Originally arXiv:2204.10844.
- [14] L. Šmejkal, J. Sinova, and T. Jungwirth, “Beyond Conventional Ferromagnetism and Antiferromagnetism: A Phase with Nonrelativistic Spin and Crystal Rotation Symmetry,” *Physical Review X*, vol. 12, p. 031042, Sept. 2022. Publisher: American Physical Society.
- [15] L.-D. Yuan, Z. Wang, J.-W. Luo, E. I. Rashba, and A. Zunger, “Giant momentum-dependent spin splitting in centrosymmetric low- z antiferromagnets,” *Phys. Rev. B*, vol. 102, p. 014422, Jul 2020.
- [16] L.-D. Yuan, Z. Wang, J.-W. Luo, and A. Zunger, “Prediction of low- Z collinear and noncollinear antiferromagnetic compounds having momentum-dependent spin splitting even without spin-orbit coupling,” *Physical Review Materials*, vol. 5, p. 014409, Jan. 2021. Publisher: American Physical Society.
- [17] R. González-Hernández, L. Šmejkal, K. Výborný, Y. Yahagi, J. Sinova, T. Jungwirth, and J. Železný, “Efficient Electrical Spin Splitter Based on Nonrelativistic Collinear Antiferromagnetism,” *Physical Review Letters*, vol. 126, p. 127701, Mar. 2021. Publisher: American Physical Society.
- [18] D.-F. Shao, S.-H. Zhang, M. Li, C.-B. Eom, and E. Y. Tsymbal, “Spin-neutral currents for spintronics,” *Nature Communications*, vol. 12, p. 7061, Dec 2021.
- [19] Z. Feng, X. Zhou, L. Šmejkal, L. Wu,

- Z. Zhu, H. Guo, R. González-Hernández, X. Wang, H. Yan, P. Qin, X. Zhang, H. Wu, H. Chen, Z. Meng, L. Liu, Z. Xia, J. Sinova, T. Jungwirth, and Z. Liu, “An anomalous Hall effect in altermagnetic ruthenium dioxide,” *Nature Electronics*, vol. 5, pp. 735–743, Nov. 2022. Publisher: Nature Publishing Group.
- [20] A. Bose, N. J. Schreiber, R. Jain, D.-F. Shao, H. P. Nair, J. Sun, X. S. Zhang, D. A. Muller, E. Y. Tsybal, D. G. Schlom, and D. C. Ralph, “Tilted spin current generated by the collinear antiferromagnet ruthenium dioxide,” *Nature Electronics*, pp. 1–8, May 2022. Publisher: Nature Publishing Group.
- [21] S. Karube, T. Tanaka, D. Sugawara, N. Kadoguchi, M. Kohda, and J. Nitta, “Observation of Spin-Splitter Torque in Collinear Antiferromagnetic RuO₂,” *Physical Review Letters*, vol. 129, p. 137201, Sept. 2022. Publisher: American Physical Society.
- [22] O. Fedchenko, J. Minár, A. Akashdeep, S. W. D’Souza, D. Vasilyev, O. Tkach, L. Odenbreit, Q. Nguyen, D. Kutnyakhov, N. Wind, L. Wenthau, M. Scholz, K. Rossnagel, M. Hoesch, M. Aeschlimann, B. Stadtmüller, M. Kläui, G. Schönhense, T. Jungwirth, A. B. Hellenes, G. Jakob, L. Šmejkal, J. Sinova, and H.-J. Elmers, “Observation of time-reversal symmetry breaking in the band structure of altermagnetic RuO₂,” *Science Advances*, vol. 10, p. eadj4883, Jan. 2024. Publisher: American Association for the Advancement of Science.
- [23] T. Berlijn, P. C. Snijders, O. Delaire, H.-D. Zhou, T. A. Maier, H.-B. Cao, S.-X. Chi, M. Matsuda, Y. Wang, M. R. Koehler, P. R. C. Kent, and H. H. Weitering, “Itinerant Antiferromagnetism in RuO₂,” *Physical Review Letters*, vol. 118, p. 077201, Feb. 2017. Publisher: American Physical Society.
- [24] M. Hiraishi, H. Okabe, A. Koda, R. Kadono, T. Muroi, D. Hirai, and Z. Hiroi, “Nonmagnetic ground state in RuO₂ revealed by muon spin rotation,” *Phys. Rev. Lett.*, vol. 132, p. 166702, Apr 2024.
- [25] P. Keßler, L. Garcia-Gassull, A. Suter, T. Prokscha, Z. Salman, D. Khalyavin, P. Manuel, F. Orlandi, I. I. Mazin, R. Valentí, and S. Moser, “Absence of magnetic order in RuO₂: insights from μ sr spectroscopy and neutron diffraction,” *npj Spintronics*, vol. 2, p. 50, 2024. Originally arXiv:2405.10820.
- [26] Z. Wu, M. Long, H. Chen, S. Paul, H. Matsuki, O. Zheliuk, U. Zeitler, G. Li, R. Zhou, Z. Zhu, D. Graf, T. I.

- Weinberger, F. M. Grosche, Y. Maeno, and A. G. Eaton, “The Fermi surface of RuO_2 measured by quantum oscillations,” *Physical Review X*, vol. 15, p. 031044, 2025. Originally arXiv:2503.20621.
- [27] A. Smolyanyuk, I. I. Mazin, L. Garcia-Gassull, and R. Valentí, “Fragility of the magnetic order in the prototypical altermagnet RuO_2 ,” *Phys. Rev. B*, vol. 109, p. 134424, Apr 2024.
- [28] S. Brahimi, D. P. Rai, and S. Lounis, “Confinement-induced altermagnetism in RuO_2 thin films,” *Journal of Physics: Condensed Matter*, vol. 37, p. 395801, Sept. 2025. Originally arXiv:2412.15377.
- [29] E. V. Gomonay and V. M. Loktev, “Spintronics of antiferromagnetic systems (Review Article),” *Low Temperature Physics*, vol. 40, pp. 17–35, Jan. 2014.
- [30] P. Wadley, B. Howells, J. Železný, C. Andrews, V. Hills, R. P. Campion, V. Novák, K. Olejník, F. Maccherozzi, S. S. Dhesi, S. Y. Martin, T. Wagner, J. Wunderlich, F. Freimuth, Y. Mokrousov, J. Kuneš, J. S. Chauhan, M. J. Grzybowski, A. W. Rushforth, K. W. Edmonds, B. L. Gallagher, and T. Jungwirth, “Electrical switching of an antiferromagnet,” *Science*, vol. 351, pp. 587–590, Feb. 2016.
- Publisher: American Association for the Advancement of Science.
- [31] S. Y. Bodnar, L. Šmejkal, I. Turek, T. Jungwirth, O. Gomonay, J. Sinova, A. A. Sapozhnik, H.-J. Elmers, M. Kläui, and M. Jourdan, “Writing and reading antiferromagnetic Mn_2Au by Néel spin-orbit torques and large anisotropic magnetoresistance,” *Nature Communications*, vol. 9, p. 348, Jan. 2018. Publisher: Nature Publishing Group.
- [32] R. Lebrun, A. Ross, O. Gomonay, S. A. Bender, L. Baldrati, F. Kronast, A. Qaiumzadeh, J. Sinova, A. Brataas, R. A. Duine, and M. Kläui, “Anisotropies and magnetic phase transitions in insulating antiferromagnets determined by a Spin-Hall magnetoresistance probe,” *Communications Physics*, vol. 2, p. 50, May 2019. Publisher: Nature Publishing Group.
- [33] M. Isasa, S. Vélez, E. Sagasta, A. Bedoya-Pinto, N. Dix, F. Sánchez, L. E. Hueso, J. Fontcuberta, and F. Casanova, “Spin Hall Magnetoresistance as a Probe for Surface Magnetization in $\text{Pt} / \text{CoFe}_2\text{O}_4$ Bilayers,” *Physical Review Applied*, vol. 6, p. 034007, Sept. 2016. Publisher: American Physical Society.

- [34] L. Han, X. Fu, R. Peng, X. Cheng, J. Dai, L. Liu, Y. Li, Y. Zhang, W. Zhu, H. Bai, Y. Zhou, S. Liang, C. Chen, Q. Wang, X. Chen, L. Yang, Y. Zhang, C. Song, J. Liu, and F. Pan, “Electrical 180° switching of Néel vector in spin-splitting antiferromagnet,” *Science Advances*, vol. 10, p. eadn0479, Jan. 2024. Publisher: American Association for the Advancement of Science.
- [35] R. Lebrun, A. Ross, S. A. Bender, A. Qaiumzadeh, L. Baldrati, J. Cramer, A. Brataas, R. A. Duine, and M. Kläui, “Tunable long-distance spin transport in a crystalline antiferromagnetic iron oxide,” *Nature*, vol. 561, pp. 222–225, Sept. 2018. Publisher: Nature Publishing Group.
- [36] H. Wang, C. Du, P. C. Hammel, and F. Yang, “Antiferromagnonic Spin Transport from $\text{Y}_3\text{Fe}_5\text{O}_{12}$ into NiO ,” *Physical Review Letters*, vol. 113, p. 097202, Aug. 2014. Publisher: American Physical Society.
- [37] S. M. Rezende, R. L. Rodríguez-Suárez, and A. Azevedo, “Theory of the spin Seebeck effect in antiferromagnets,” *Physical Review B*, vol. 93, p. 014425, Jan. 2016. Publisher: American Physical Society.
- [38] G. R. Hoogeboom, A. Aqeel, T. Kuschel, T. T. M. Palstra, and B. J. van Wees, “Negative spin Hall magnetoresistance of Pt on the bulk easy-plane antiferromagnet NiO ,” *Applied Physics Letters*, vol. 111, p. 052409, Aug. 2017.
- [39] R. Tamang, S. Gurung, D. P. Rai, S. Brahimi, and S. Lounis, “Newly discovered magnetic phase: A brief review on Altermagnets,” Dec. 2024. arXiv:2412.05377 [cond-mat].
- [40] T. Jungwirth, R. M. Fernandes, E. Fradkin, A. H. MacDonald, J. Sinova, and L. Šmejkal, “Altermagnetism: An unconventional spin-ordered phase of matter,” *Newton*, vol. 0, July 2025. Publisher: Elsevier.
- [41] K.-H. Ahn, A. Hariki, K.-W. Lee, and J. Kuneš, “Antiferromagnetism in RuO_2 as d-wave Pomeranchuk instability,” *Physical Review B*, vol. 99, p. 184432, May 2019. Publisher: American Physical Society.
- [42] L. Bai, W. Feng, S. Liu, L. Šmejkal, Y. Mokrousov, and Y. Yao, “Altermagnetism: Exploring New Frontiers in Magnetism and Spintronics,” *Advanced Functional Materials*, vol. 34, no. 49, p. 2409327, 2024.
- [43] L. Šmejkal, R. González-Hernández, T. Jungwirth, and J. Sinova, “Crystal time-reversal symmetry breaking and spontaneous Hall effect in collinear antiferromagnets,” *Science Advances*,

- vol. 6, p. eaaz8809, June 2020. Publisher: American Association for the Advancement of Science.
- [44] H. Bai, L. Han, X. Y. Feng, Y. J. Zhou, R. X. Su, Q. Wang, L. Y. Liao, W. X. Zhu, X. Z. Chen, F. Pan, X. L. Fan, and C. Song, “Observation of Spin Splitting Torque in a Collinear Antiferromagnet RuO_2 ,” *Physical Review Letters*, vol. 128, p. 197202, May 2022. Publisher: American Physical Society.
- [45] L. Šmejkal, A. B. Hellenes, R. González-Hernández, J. Sinova, and T. Jungwirth, “Giant and tunneling magnetoresistance effects from anisotropic and valley-dependent spin-momentum interactions in antiferromagnets,” *Physical Review X*, vol. 12, p. 011028, Feb. 2022. arXiv:2103.12664 [cond-mat].
- [46] L. Šmejkal, A. Marmodoro, K.-H. Ahn, R. González-Hernández, I. Turek, S. Mankovsky, H. Ebert, S. W. D’Souza, O. Šipr, J. Sinova, and T. Jungwirth, “Chiral Magnons in Altermagnetic RuO_2 ,” *Physical Review Letters*, vol. 131, p. 256703, Dec. 2023. Publisher: American Physical Society.
- [47] A. Smolyanyuk, I. I. Mazin, L. Garcia-Gassull, and R. Valentí, “Fragility of the magnetic order in the prototypical altermagnet RuO_2 ,” *Physical Review B*, vol. 109, p. 134424, Apr. 2024. Publisher: American Physical Society.
- [48] A. Smolyanyuk, L. Šmejkal, and I. I. Mazin, “Origin of anomalous hall effect in cr-doped RuO_2 ,” Nov. 2024. arXiv:2411.02507.
- [49] M. Born and R. Oppenheimer, “Zur Quantentheorie der Molekeln,” *Annalen der Physik*, vol. 389, no. 20, pp. 457–484, 1927.
- [50] P. Hohenberg and W. Kohn, “Inhomogeneous electron gas,” *Phys. Rev.*, vol. 136, pp. B864–B871, Nov 1964.
- [51] W. Kohn and L. J. Sham, “Self-consistent equations including exchange and correlation effects,” *Phys. Rev.*, vol. 140, pp. A1133–A1138, Nov 1965.
- [52] J. P. Perdew and A. Zunger, “Self-interaction correction to density-functional approximations for many-electron systems,” *Phys. Rev. B*, vol. 23, pp. 5048–5079, May 1981.
- [53] J. P. Perdew, J. A. Chevary, S. H. Vosko, K. A. Jackson, M. R. Pederson, D. J. Singh, and C. Fiolhais, “Atoms, molecules, solids, and surfaces: Applications of the generalized gradient approximation for exchange and correlation,” *Physical Review B*, vol. 46,

- pp. 6671–6687, Sept. 1992. Publisher: American Physical Society.
- [54] J. P. Perdew, K. Burke, and Y. Wang, “Generalized gradient approximation for the exchange-correlation hole of a many-electron system,” *Physical Review B*, vol. 54, pp. 16533–16539, Dec. 1996. Publisher: American Physical Society.
- [55] U. v. Barth and L. Hedin, “A local exchange-correlation potential for the spin polarized case. i,” *Journal of Physics C: Solid State Physics*, vol. 5, p. 1629, July 1972. Publisher: IOP Publishing.
- [56] J. Kubler, K.-H. Hock, J. Sticht, and A. R. Williams, “Density functional theory of non-collinear magnetism,” *Journal of Physics F: Metal Physics*, vol. 18, p. 469, Mar. 1988.
- [57] V. I. Anisimov, J. Zaanen, and O. K. Andersen, “Band theory and Mott insulators: Hubbard U instead of Stoner I,” *Physical Review B*, vol. 44, pp. 943–954, July 1991. Publisher: American Physical Society.
- [58] B. Himmetoglu, A. Floris, S. de Gironcoli, and M. Cococcioni, “Hubbard-corrected DFT energy functionals: The LDA+U description of correlated systems,” *International Journal of Quantum Chemistry*, vol. 114, no. 1, pp. 14–49, 2014.
- [59] M. Cococcioni and S. de Gironcoli, “Linear response approach to the calculation of the effective interaction parameters in the LDA+ u method,” *Physical Review B*, vol. 71, p. 035105, Jan. 2005. Publisher: American Physical Society.
- [60] A. Filippetti and N. A. Spaldin, “Self-interaction-corrected pseudopotential scheme for magnetic and strongly-correlated systems,” *Physical Review B*, vol. 67, p. 125109, Mar. 2003. Publisher: American Physical Society.
- [61] C. D. Pemmaraju, T. Archer, D. Sánchez-Portal, and S. Sanvito, “Atomic-orbital-based approximate self-interaction correction scheme for molecules and solids,” *Phys. Rev. B*, vol. 75, p. 045101, Jan 2007.
- [62] J. Heyd, G. E. Scuseria, and M. Ernzerhof, “Erratum: “Hybrid functionals based on a screened Coulomb potential” [*J. Chem. Phys.* **118**, 8207 (2003)],” *J. Chem. Phys.*, vol. 124, no. 21, p. 219906, 2006.
- [63] J. Sun, A. Ruzsinszky, and J. P. Perdew, “Strongly constrained and appropriately normed semilocal density functional,” *Phys. Rev. Lett.*, vol. 115, p. 036402, Jul 2015.

- [64] A. R. Rocha, V. M. García-Suárez, S. Bailey, C. Lambert, J. Ferrer, and S. Sanvito, “Spin and molecular electronics in atomically generated orbital landscapes,” *Phys. Rev. B*, vol. 73, p. 085414, Feb 2006.
- [65] J. M. Soler, E. Artacho, J. D. Gale, A. García, J. Junquera, P. Ordejón, and D. Sánchez-Portal, “The SIESTA method for ab-initio $\mathcal{O}(N)$ materials simulation,” *Journal of Physics: Condensed Matter*, vol. 14, pp. 2745–2779, Mar. 2002. Publisher: IOP Publishing.
- [66] S. Datta, *Electronic Transport in Mesoscopic Systems*. Cambridge Studies in Semiconductor Physics and Micro-electronic Engineering, Cambridge: Cambridge University Press, 1995.
- [67] S. Sanvito, “Electron transport theory for large systems,” *Computational Nanoscience*, vol. 4, p. 179, 2011.
- [68] D. S. Fisher and P. A. Lee, “Relation between conductivity and transmission matrix,” *Physical Review B*, vol. 23, pp. 6851–6854, June 1981. Publisher: American Physical Society.
- [69] S. Sanvito, “Ab-initio methods for spin-transport at the nanoscale level.” arXiv:cond-mat/0503445, 2005.
- [70] E. Artacho, E. Anglada, O. Diéguez, J. D. Gale, A. García, J. Junquera, R. M. Martin, P. Ordejón, J. M. Pruneda, D. Sánchez-Portal, and J. M. Soler, “The SIESTA method: developments and applicability,” *Journal of Physics: Condensed Matter*, vol. 20, p. 064208, Jan. 2008. Publisher: IOP Publishing.
- [71] A. García, N. Papior, A. Akhtar, E. Artacho, V. Blum, E. Bosoni, P. Brandimarte, M. Brandbyge, J. I. Cerdá, F. Corsetti, R. Cuadrado, V. Dikan, J. Ferrer, J. Gale, P. García-Fernández, V. M. García-Suárez, S. García, G. Huhs, S. Illera, R. Korytár, P. Koval, I. Lebedeva, L. Lin, P. López-Tarifa, S. G. Mayo, S. Mohr, P. Ordejón, A. Postnikov, Y. Pouillon, M. Pruneda, R. Robles, D. Sánchez-Portal, J. M. Soler, R. Ullah, V. W.-z. Yu, and J. Junquera, “Siesta: Recent developments and applications,” *The Journal of Chemical Physics*, vol. 152, p. 204108, May 2020. Publisher: American Institute of Physics.
- [72] C.-E. Boman, J. Danielsen, A. Haaland, B. Jerslev, C. E. Schäffer, E. Sunde, and N. A. Sørensen, “Refinement of the crystal structure of ruthenium dioxide,” *Acta Chem. Scand*, vol. 24, no. 1, pp. 116–122, 1970.
- [73] A. Jain, S. P. Ong, G. Hautier, W. Chen, W. D. Richards, S. Dacek, S. Cholia, D. Gunter, D. Skinner, G. Ceder, and

- K. A. Persson, "Commentary: The materials project: A materials genome approach to accelerating materials innovation," *APL Materials*, vol. 1, p. 011002, 07 2013.
- [74] The Materials Project, "Materials data on RuO₂ by materials project." Materials Project, Lawrence Berkeley National Laboratory, May 2020. Materials Project ID: mp-825.
- [75] H. Over, "Surface Chemistry of Ruthenium Dioxide in Heterogeneous Catalysis and Electrocatalysis: From Fundamental to Applied Research," *Chemical Reviews*, vol. 112, pp. 3356–3426, June 2012. Publisher: American Chemical Society.
- [76] W. Ryden, A. Lawson, and C. C. Sar-tain, "Electrical transport properties of IrO₂ and RuO₂," *Physical Review B*, vol. 1, no. 4, p. 1494, 1970.
- [77] J. M. Fletcher, W. E. Gardner, B. F. Greenfield, M. J. Holdoway, and M. H. Rand, "Magnetic and other studies of ruthenium dioxide and its hydrate," *Journal of the Chemical Society A: Inorganic, Physical, Theoretical*, pp. 653–657, Jan. 1968. Publisher: The Royal Society of Chemistry.
- [78] Z. Zhu, J. Strempfer, R. Rao, C. Occhialini, J. Pelliciari, Y. Choi, T. Kawaguchi, H. You, J. Mitchell, Y. Shao-Horn, and R. Comin, "Anomalous Antiferromagnetism in Metallic RuO₂ Determined by Resonant X-ray Scattering," *Physical Review Letters*, vol. 122, p. 017202, Jan. 2019. Publisher: American Physical Society.
- [79] I. H. Choi, S. G. Jeong, B. Jalan, and J. S. Lee, "Exploring altermagnetism in RuO₂: from conflicting experiments to emerging consensus," *Nano Convergence*, vol. 13, p. 1, 2026.
- [80] Z.-X. Li, H. Zhou, X. Wan, and W. Chen, "Diagnosing altermagnetic phases through quantum oscillations," *Physical Review B*, vol. 111, p. 125119, Mar. 2025. Publisher: American Physical Society.
- [81] M. Wenzel, E. Uykur, S. Rößler, M. Schmidt, O. Janson, A. Tiwari, M. Dressel, and A. A. Tsirlin, "Fermi-liquid behavior of nonaltermagnetic RuO₂," *Physical Review B*, vol. 111, p. L041115, Jan. 2025. Publisher: American Physical Society.
- [82] J. Liu, J. Zhan, T. Li, J. Liu, S. Cheng, Y. Shi, L. Deng, M. Zhang, C. Li, J. Ding, Q. Jiang, M. Ye, Z. Liu, Z. Jiang, S. Wang, Q. Li, Y. Xie, Y. Wang, S. Qiao, J. Wen, Y. Sun, and D. Shen, "Absence of Altermagnetic Spin Splitting Character in Rutile Oxide RuO₂," *Physical Review Letters*, vol. 133, p. 176401, Oct.

2024. Publisher: American Physical Society.
- [83] E. Brand, V. Rosendal, Y. Wu, T. Tran, A. Palliotto, I. V. Maznichenko, S. Ostannin, V. Esposito, A. Ernst, S. Zhou, D.-S. Park, and N. Pryds, “Defect-induced magnetic symmetry breaking in oxide materials,” *Applied Physics Reviews*, vol. 12, p. 011327, Mar. 2025.
- [84] S. G. Jeong, S. Lee, B. Lin, Z. Yang, I. H. Choi, J. Y. Oh, S. Song, S. W. Lee, S. Nair, R. Choudhary, J. Parikh, S. Park, W. S. Choi, J. S. Lee, J. M. LeBeau, T. Low, and B. Jalan, “Metallicity and anomalous Hall effect in epitaxially strained, atomically thin RuO₂ films,” *Proceedings of the National Academy of Sciences*, vol. 122, no. 27, p. e2500831122, 2025.
- [85] P. Quarterman, C. Sun, J. Garcia-Barriocanal, M. DC, Y. Lv, S. Manipatruni, D. E. Nikonov, I. A. Young, P. M. Voyles, and J.-P. Wang, “Demonstration of Ru as the 4th ferromagnetic element at room temperature,” *Nature Communications*, vol. 9, p. 2058, May 2018.
- [86] M. Alaei, N. Rezaei, I. Mikhailov, A. R. Oganov, and A. Qaiumzadeh, “Complex magnetic behavior in RuO₂ thin films driven by strain and substrate effects,” Feb. 2026. arXiv:2602.15126 [cond-mat].
- [87] Y. Kobayashi, S. Karube, I. Sugiura, H. Narita, R. Hisatomi, Y. Shiota, and T. Ono, “Detection of antiferromagnetic order in a RuO₂/Pt bilayer by spin Hall magnetoresistance,” *AIP Advances*, vol. 14, p. 115120, Nov. 2024.
- [88] Y.-C. Wang, Z.-Y. Shen, C.-H. Lin, W.-C. Hsu, Y.-Y. Chin, A. K. Singh, W.-L. Lee, S.-Y. Huang, and D. Qu, “Robust anisotropic spin hall effect in rutile RuO₂,” *Nano Letters*, 2025. Originally arXiv:2503.07985.
- [89] Y.-Y. Jiang, Z.-A. Wang, K. Samanta, S.-H. Zhang, R.-C. Xiao, W. J. Lu, Y. P. Sun, E. Y. Tsymbal, and D.-F. Shao, “Prediction of Giant Tunneling Magnetoresistance in RuO₂/TiO₂/RuO₂ (110) Antiferromagnetic Tunnel Junctions.” arXiv:2309.02634, Sept. 2023.
- [90] S. Noh, G.-H. Kim, J. Lee, H. Jung, U. Seo, G. So, J. Lee, S. Lee, M. Park, S. Yang, Y. S. Oh, H. Jin, C. Sohn, and J.-W. Yoo, “Tunneling Magnetoresistance in Altermagnetic RuO₂-Based Magnetic Tunnel Junctions,” *Physical Review Letters*, vol. 134, p. 246703, June 2025. Publisher: American Physical Society.
- [91] H. Chen, Z. Wang, P. Qin, Z. Meng, X. Zhou, X. Wang, L. Liu, G. Zhao,

- Z. Duan, T. Zhang, J. Liu, D. Shao, C. Jiang, and Z. Liu, "Spin-splitting magnetoresistance in altermagnetic RuO₂ thin films," *Advanced Materials*, vol. 37, p. 2507764, 2025.
- [92] N. Troullier and J. L. Martins, "Efficient pseudopotentials for plane-wave calculations," *Physical Review B*, vol. 43, pp. 1993–2006, Jan. 1991. Publisher: American Physical Society.
- [93] S. L. Dudarev, G. A. Botton, S. Y. Savrasov, C. J. Humphreys, and A. P. Sutton, "Electron-energy-loss spectra and the structural stability of nickel oxide: An LSDA+U study," *Physical Review B*, vol. 57, pp. 1505–1509, Jan. 1998. Publisher: American Physical Society.
- [94] M. Gell-Mann and K. A. Brueckner, "Correlation Energy of an Electron Gas at High Density," *Physical Review*, vol. 106, pp. 364–368, Apr. 1957. Publisher: American Physical Society.
- [95] D. M. Ceperley and B. J. Alder, "Ground State of the Electron Gas by a Stochastic Method," *Physical Review Letters*, vol. 45, pp. 566–569, Aug. 1980. Publisher: American Physical Society.
- [96] D. C. Langreth and J. P. Perdew, "Exchange-correlation energy of a metallic surface: Wave-vector analysis," *Physical Review B*, vol. 15, pp. 2884–2901, Mar. 1977. Publisher: American Physical Society.
- [97] D. C. Langreth and M. J. Mehl, "Beyond the local-density approximation in calculations of ground-state electronic properties," *Physical Review B*, vol. 28, pp. 1809–1834, Aug. 1983. Publisher: American Physical Society.
- [98] C. D. Hu and D. C. Langreth, "A Spin Dependent Version of the Langreth-Mehl Exchange-Correlation Functional," *Physica Scripta*, vol. 32, p. 391, Oct. 1985. Publisher: IOP Publishing.
- [99] G. Kresse and J. Furhmüller, "Software vasp, vienna (1999); kresse g. and hafner," *Phys. Rev. B*, vol. 47, no. R558, 1993.
- [100] G. Kresse and J. Furthmüller, "Efficiency of ab-initio total energy calculations for metals and semiconductors using a plane-wave basis set," *Computational materials science*, vol. 6, no. 1, pp. 15–50, 1996.
- [101] G. Kresse and J. Furthmüller, "Software vasp, vienna (1999)," *Phys. Rev. B*, vol. 54, no. 11, p. 169, 1996.
- [102] G. Kresse and D. Joubert, "From ultrasoft pseudopotentials to the projector augmented-wave method," *Physical review b*, vol. 59, no. 3, p. 1758, 1999.

- [103] P. Giannozzi, S. Baroni, N. Bonini, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, G. L. Chiarotti, M. Cococcioni, I. Dabo, A. Dal Corso, S. de Gironcoli, S. Fabris, G. Fratesi, R. Gebauer, U. Gerstmann, C. Gougoussis, A. Kokalj, M. Lazzeri, L. Martin-Samos, N. Marzari, F. Mauri, R. Mazzarello, S. Paolini, A. Pasquarello, L. Paulatto, C. Sbraccia, S. Scandolo, G. Sclauzero, A. P. Seitsonen, A. Smogunov, P. Umari, and R. M. Wentzcovitch, “Quantum espresso: a modular and open-source software project for quantum simulations of materials,” *Journal of Physics: Condensed Matter*, vol. 21, no. 39, p. 395502 (19pp), 2009.
- [104] P. Giannozzi, O. Andreussi, T. Brumme, O. Bunau, M. B. Nardelli, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, M. Cococcioni, N. Colonna, I. Carnimeo, A. D. Corso, S. de Gironcoli, P. Delugas, R. A. D. Jr, A. Ferretti, A. Floris, G. Fratesi, G. Fugallo, R. Gebauer, U. Gerstmann, F. Giustino, T. Gorni, J. Jia, M. Kawamura, H.-Y. Ko, A. Kokalj, E. Küçükbenli, M. Lazzeri, M. Marsili, N. Marzari, F. Mauri, N. L. Nguyen, H.-V. Nguyen, A. O. de-la Roza, L. Paulatto, S. Poncé, D. Rocca, R. Sabatini, B. Santra, M. Schlipf, A. P. Seitsonen, A. Smogunov, I. Timrov, T. Thonhauser, P. Umari, N. Vast, X. Wu, and S. Baroni, “Advanced capabilities for materials modelling with quantum espresso,” *Journal of Physics: Condensed Matter*, vol. 29, no. 46, p. 465901, 2017.
- [105] P. Giannozzi, O. Baseggio, P. Bonfà, D. Brunato, R. Car, I. Carnimeo, C. Cavazzoni, S. de Gironcoli, P. Delugas, F. Ferrari Ruffino, A. Ferretti, N. Marzari, I. Timrov, A. Urru, and S. Baroni, “Quantum espresso toward the exascale,” *The Journal of Chemical Physics*, vol. 152, no. 15, p. 154105, 2020.
- [106] K. Schwarz, P. Blaha, and G. K. Madsen, “Electronic structure calculations of solids using the wien2k package for material sciences,” *Computer physics communications*, vol. 147, no. 1-2, pp. 71–76, 2002.
- [107] N. Mehtougui, D. Rached, R. Khenata, H. Rached, M. Rabah, and S. Bin-Omran, “Structural, electronic and mechanical properties of RuO₂ from first-principles calculations,” *Materials Science in Semiconductor Processing*, vol. 15, pp. 331–339, Aug. 2012.
- [108] M. Wang, K. Tanaka, S. Sakai, Z. Wang, K. Deng, Y. Lyu, C. Li, D. Tian, S. Shen, N. Ogawa, N. Kanazawa, P. Yu, R. Arita, and F. Kagawa, “Emergent

- zero-field anomalous hall effect in a reconstructed rutile antiferromagnetic metal,” *Nat. Commun.*, vol. 14, p. 8240, Dec. 2023.
- [109] D. T. Plouff, L. Scheuer, S. Shrestha, W. Wu, N. J. Parvez, S. Bhatt, X. Wang, L. Gundlach, M. B. Jungfleisch, and J. Q. Xiao, “Revisiting altermagnetism in RuO₂: a study of laser-pulse induced charge dynamics by time-domain terahertz spectroscopy,” *npj Spintronics*, vol. 3, p. 17, May 2025. Publisher: Nature Publishing Group.
- [110] X. Wang, J. R. Yates, I. Souza, and D. Vanderbilt, “Ab initio calculation of the anomalous hall conductivity by wannier interpolation,” *Phys. Rev. B*, vol. 74, p. 195118, Nov 2006.
- [111] J. Qiao, J. Zhou, Z. Yuan, and W. Zhao, “Calculation of intrinsic spin hall conductivity by wannier interpolation,” *Phys. Rev. B*, vol. 98, p. 214402, Dec 2018.
- [112] J. Sinova, S. O. Valenzuela, J. Wunderlich, C. H. Back, and T. Jungwirth, “Spin hall effects,” *Rev. Mod. Phys.*, vol. 87, pp. 1213–1260, Oct 2015.
- [113] L.-D. Yuan, Z. Wang, J.-W. Luo, and A. Zunger, “Prediction of low-*z* collinear and noncollinear antiferromagnetic compounds having momentum-dependent spin splitting even without spin-orbit coupling,” *Phys. Rev. Materials*, vol. 5, p. 014409, Jan 2021.
- [114] S. V. Gallego, J. M. Perez-Mato, L. Elcoro, E. S. Tasci, R. M. Hanson, K. Momma, M. I. Aroyo, and G. Madariaga, “MAGNDATA: towards a database of magnetic structures. I. The commensurate case,” *Journal of Applied Crystallography*, vol. 49, pp. 1750–1776, Oct 2016.
- [115] S. Gražulis, D. Chateigner, R. T. Downs, A. F. T. Yokochi, M. Quirós, L. Lutterotti, E. Manakova, J. Butkus, P. Moeck, and A. Le Bail, “Crystallography Open Database – an open-access collection of crystal structures,” *Journal of Applied Crystallography*, vol. 42, pp. 726–729, Aug 2009.
- [116] S. Gražulis, A. Daškevič, A. Merkys, D. Chateigner, L. Lutterotti, M. Quirós, N. R. Serebryanaya, P. Moeck, R. T. Downs, and A. Le Bail, “Crystallography open database (cod): an open-access collection of crystal structures and platform for world-wide collaboration,” *Nucleic Acids Research*, vol. 40, no. D1, pp. D420–D427, 2012.
- [117] M. Thakurdesai, A. Mahadkar, D. Kanjilal, and V. Bhattacharyya, “Nanocrystallisation of TiO₂ induced by dense electronic excitation,” *Vac-*

- uum*, vol. 82, no. 6, pp. 639–644, 2008.
- [118] L. Liu, “A Dense Modification of BaO and Its Crystal Structure,” *Journal of Applied Physics*, vol. 42, pp. 3702–3704, Sept. 1971.
- [119] M. Kristl, S. Gyergyek, S. D. Škapin, and J. Kristl, “Solvent-free mechanochemical synthesis and characterization of nickel tellurides with various stoichiometries: NiTe, NiTe₂ and Ni₂Te₃,” *Nanomaterials*, vol. 11, no. 8, 2021.
- [120] F. Ersan, E. Vatansever, S. Sarikurt, Y. Yüksel, Y. Kadioglu, H. D. Ozaydin, O. Üzengi Aktürk, Ümit Akıncı, and E. Aktürk, “Exploring the electronic and magnetic properties of new metal halides from bulk to two-dimensional monolayer: RuX₃ (X = Br, I),” *Journal of Magnetism and Magnetic Materials*, vol. 476, pp. 111–119, 2019.
- [121] W. H. Butler, X.-G. Zhang, T. C. Schulthess, and J. M. MacLaren, “Spin-dependent tunneling conductance of Fe|MgO|Fe sandwiches,” *Physical Review B*, vol. 63, p. 054416, Jan. 2001. Publisher: American Physical Society.
- [122] J. Mathon and A. Umerski, “Theory of tunneling magnetoresistance of an epitaxial Fe/MgO/Fe(001) junction,” *Physical Review B*, vol. 63, p. 220403, May 2001. Publisher: American Physical Society.
- [123] P. Soven, “Coherent-potential model of substitutional disordered alloys,” *Phys. Rev.*, vol. 156, pp. 809–813, Apr 1967.
- [124] N. D. Mermin and H. Wagner, “Absence of ferromagnetism or antiferromagnetism in one- or two-dimensional isotropic Heisenberg models,” *Phys. Rev. Lett.*, vol. 17, pp. 1133–1136, Nov 1966.
- [125] B. Huang, G. Clark, E. Navarro-Moratalla, D. R. Klein, R. Cheng, K. L. Seyler, D. Zhong, E. Schmidgall, M. A. McGuire, D. H. Cobden, W. Yao, D. Xiao, P. Jarillo-Herrero, and X. Xu, “Layer-dependent ferromagnetism in a van der Waals crystal down to the monolayer limit,” *Nature*, vol. 546, pp. 270–273, Jun 2017.
- [126] D. Pines and D. Bohm, “A collective description of electron interactions: II. Collective vs individual particle aspects of the interactions,” *Phys. Rev.*, vol. 85, pp. 338–353, Jan 1952.