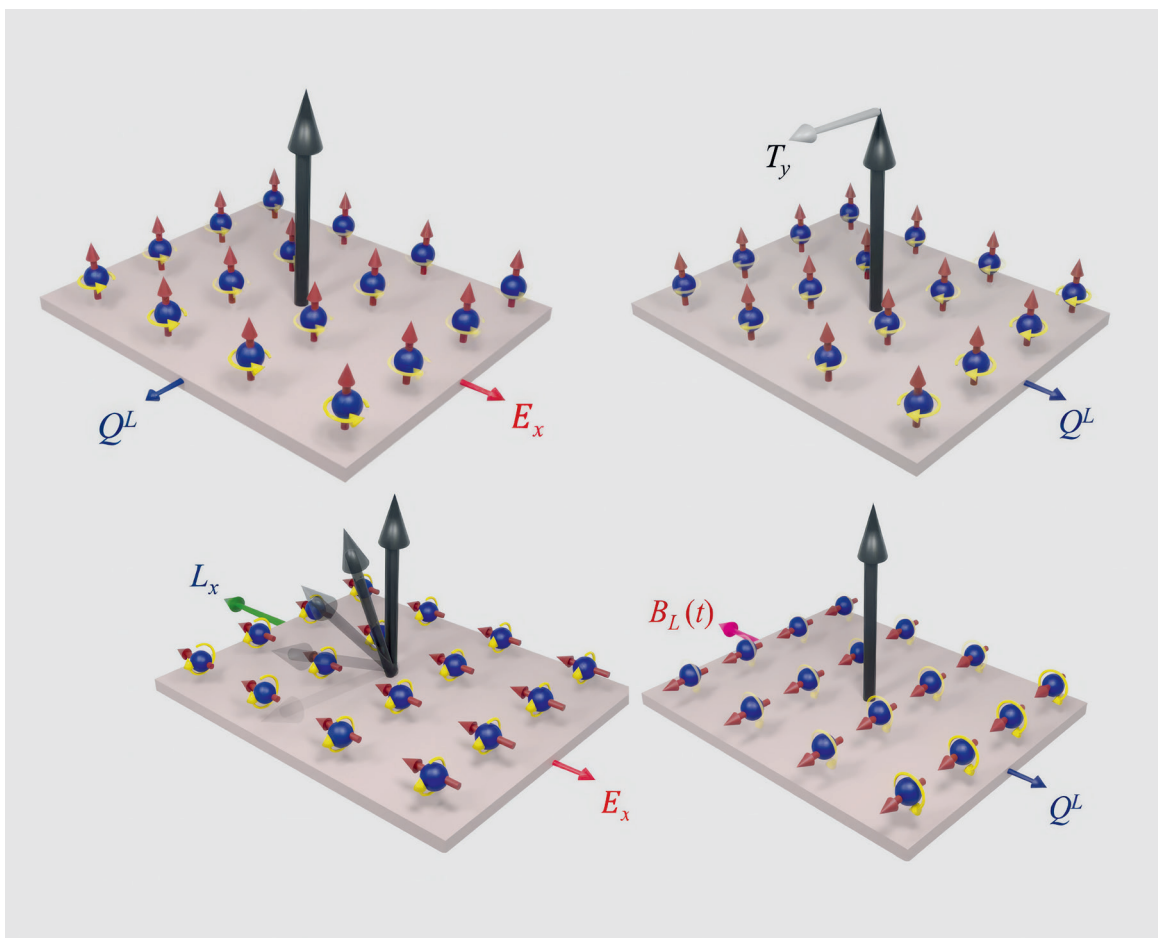




# Orbitronics with Two-Dimensional Rare-Earth Dichalcogenides from First Principles

Mahmoud Zeer

Information

Band / Volume 131

ISBN 978-3-95806-961-9





Forschungszentrum Jülich GmbH  
Peter Grünberg Institut (PGI)  
Quanten-Theorie der Materialien (PGI-1)

# **Orbitronics with Two-Dimensional Rare-Earth Dichalcogenides from First Principles**

Mahmoud Zeer

Schriften des Forschungszentrums Jülich  
Reihe Information / Information

Band / Volume 131

ISSN 1866-1777

ISBN 978-3-95806-961-9



Bibliografische Information der Deutschen Nationalbibliothek.  
Die Deutsche Nationalbibliothek verzeichnet diese Publikation in der  
Deutschen Nationalbibliografie; detaillierte Bibliografische Daten  
sind im Internet über <http://dnb.d-nb.de> abrufbar.

Herausgeber  
und Vertrieb: Forschungszentrum Jülich GmbH  
Zentralbibliothek, Verlag  
52425 Jülich  
Tel.: +49 2461 61-5368  
Fax: +49 2461 61-6103  
[zb-publikation@fz-juelich.de](mailto:zb-publikation@fz-juelich.de)  
[www.fz-juelich.de/zb](http://www.fz-juelich.de/zb)

Umschlaggestaltung: Grafische Medien, Forschungszentrum Jülich GmbH

Druck: Grafische Medien, Forschungszentrum Jülich GmbH

Copyright: Forschungszentrum Jülich 2026

Schriften des Forschungszentrums Jülich  
Reihe Information / Information, Band / Volume 131

D 82 (Diss. RWTH Aachen University, 2026)

ISSN 1866-1777  
ISBN 978-3-95806-960-2 (Print)  
ISBN 978-3-95806-961-9 (E-Book)

Vollständig frei verfügbar über das Publikationsportal des Forschungszentrums Jülich (JuSER)  
unter [www.fz-juelich.de/zb/openaccess](http://www.fz-juelich.de/zb/openaccess).



This is an Open Access publication distributed under the terms of the [Creative Commons Attribution License 4.0](https://creativecommons.org/licenses/by/4.0/),  
which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

# Abstract

The emerging field of orbitronics, which exploits the orbital degree of freedom of electrons rather than spin for information processing, offers a promising platform for next-generation eco-friendly electronic devices. It is also regarded a viable alternative to spintronics, with orbital degree of freedom playing a central role in transport phenomena and magnetization manipulation. A major focus of this work is the exploration of orbital-related effects in two-dimensional (2D) magnetic materials, which remain underexplored in the context of orbitronics. In this study the electronic, magnetic, and transport properties of 2D rare-earth dichalcogenides (REDs) are investigated using first-principles density functional theory (DFT). By utilizing the strong spin-orbit coupling (SOC) and localized  $4f$  electrons of rare-earth elements, REDs exhibit a rich interplay of magnetism, orbital Hall effects (OHE), and spin-orbit torques (SOT), making them ideal candidates for topological spintronic and orbitronic applications.

This thesis first investigates the spin and orbital Hall effects in nonmagnetic monolayer and bilayer transition-metal dichalcogenides (TMDs) and their heterostructures. It is demonstrated that the OHE is a fundamental phenomenon, independent of SOC, and often dominates the spin Hall effect (SHE) in 2D materials. This finding suggests that the OHE could serve as a more efficient mechanism for generating spin currents in spintronic devices. The study is then extended to magnetic 2D materials, focusing on a novel family of magnetic REDs based on europium (Eu) and gadolinium (Gd). It is shown that electronic structure engineering through  $p$ - $d$ - $f$  hybridization in 2D materials can lead to a prominent topological orbital response, which is highly sensitive to the details of crystal lattice design. Furthermore, the results reveal that strain-induced modifications in the electronic structure of Gd-based REDs can enable quantized charge, spin, and orbital transport.

Finally, this thesis explores the role of orbital magnetism in mediating SOT and orbital pumping in 2D magnets based on  $f$ -electrons. Using low-symmetry Janus-type REDs, characterized by strong magnetism, significant SOC, and pronounced orbital polarization, we demonstrate that the current-induced torque acting on  $4f$  states is purely orbital in nature and reaches magnitudes comparable to those in leading 2D magnetic materials. Furthermore, we predict that magnetization dy-



namics can drive in-plane orbital angular momentum currents via orbital pumping, with the response showing a non-trivial dependence on direction and polarization due to structural asymmetry. We also identify the origin of the orbital pumping as an effect of orbital-to-orbital current conversion, establishing these REDs as promising platforms for spintronic applications based on in-plane spin and orbital angular momentum injection.



# Zusammenfassung

Das neu entstehende Gebiet der Orbitronik, das für die Informationsverarbeitung die Orbital- statt die Spin-Freiheitsgrade von Elektronen nutzt, bietet eine vielversprechende Plattform für umweltfreundliche elektronische Bauelemente der nächsten Generation. Sie wird auch als praktikable Alternative zur Spintronik angesehen, wobei der orbitale Freiheitsgrad eine zentrale Rolle bei Transportphänomenen und der Magnetisierungsmanipulation spielt. Ein Fokus dieser Arbeit liegt auf der Erforschung von orbitalen Effekten in zweidimensionalen (2D) magnetischen Materialien, die im Zusammenhang mit der Orbitronik bisher wenig erforscht sind. In dieser Studie werden die elektronischen, magnetischen und Transporteigenschaften von 2D Seltenerd-Dichalkogenide (REDs) mit Hilfe der *ab initio* Dichtefunktionaltheorie (DFT) untersucht. Durch die Nutzung der starken Spin-Bahn-Kopplung (SOC) und der lokalisierten  $4f$ -Elektronen der Seltenerdelemente zeigen REDs ein reichhaltiges Zusammenspiel von Magnetismus, Orbital-Hall-Effekten (OHE) und Spin-Bahn-Drehmomenten (SOT), was sie zu vielversprechenden Kandidaten für topologische spintronische und orbitronische Anwendungen macht.

In dieser Arbeit werden zunächst die Spin- und Orbital-Hall-Effekte in nichtmagnetischen ein- und zweilagigen Übergangsmetall-Dichalkogeniden (TMDs) sowie in deren Heterostrukturen untersucht. Es wird gezeigt, dass der OHE ein grundlegendes Phänomen ist, das unabhängig vom SOC auftritt und in 2D Materialien häufig den Spin-Hall-Effekt (SHE) übertrifft. Diese Erkenntnis legt nahe, dass der OHE als effizienterer Mechanismus zur Erzeugung von Spinströmen in spintronischen Bauelementen dienen könnte. Die Studie wird anschließend auf magnetische 2D-Materialien ausgeweitet, wobei der Fokus auf einer neuartigen Familie von magnetischen REDs auf der Basis von Europium (Eu) und Gadolinium (Gd) liegt. Es wird gezeigt, dass die elektronische Strukturentwicklung durch  $p$ - $d$ - $f$ -Hybridisierung in 2D Materialien zu einer ausgeprägten topologischen Orbitalreaktion führen kann, die sehr empfindlich auf die Details des Kristallgitters reagiert. Darüber hinaus zeigen die Ergebnisse, dass dehnungsinduzierte Veränderungen in der elektronischen Struktur von Gd-basierten REDs einen quantisierten Ladungs-, Spin- und Orbital-Transport ermöglichen können.



Abschließend wird in dieser Arbeit die Rolle des Orbitalmagnetismus bei der Vermittlung von SOT und orbitalem Pumpen in 2D-Magneten in auf  $f$ -Elektronen basierenden zweidimensionalen Magneten untersucht. Unter Verwendung von Janus-artigen REDs mit geringer Symmetrie, die sich durch starken Magnetismus, signifikante SOC und ausgeprägte orbitale Polarisierung auszeichnen, zeigen wir, dass das strominduzierte Drehmoment, das auf  $4f$ -Zustände wirkt, rein orbitaler Natur ist und Größenordnungen erreicht, die mit denen in führenden 2D-Magnetmaterialien vergleichbar sind. Darüber hinaus sagen wir voraus, dass die Magnetisierungsdynamik durch orbitales Pumpen orbitale Drehimpulsströme in der Ebene antreiben kann, wobei die Antwort aufgrund struktureller Asymmetrie eine nicht-triviale Abhängigkeit von Richtung und Polarisierung aufweist. Wir identifizieren auch den Ursprung des Orbitalpumpens als einen Effekt der Umwandlung von Orbital- in Orbitalströme und etablieren diese REDs als vielversprechende Plattformen für spintronische Anwendungen, die auf der Injektion von Spins und Orbitaldrehimpulsen in der Ebene basieren.



# Contents

|          |                                                                             |           |
|----------|-----------------------------------------------------------------------------|-----------|
| <b>1</b> | <b>Introduction</b>                                                         | <b>3</b>  |
| <b>2</b> | <b>Theory</b>                                                               | <b>9</b>  |
| 2.1      | Magnetism . . . . .                                                         | 9         |
| 2.1.1    | Fundamentals of Magnetism . . . . .                                         | 9         |
| 2.1.2    | Origin of Magnetism in Solids . . . . .                                     | 10        |
| 2.2      | Spin-Orbit Coupling . . . . .                                               | 11        |
| 2.3      | Berry Phase . . . . .                                                       | 12        |
| 2.3.1    | Geometric Properties of the Berry Curvature in the Brillouin Zone . . . . . | 14        |
| 2.3.2    | Berry Curvature for Bloch Electrons in a Two-Dimensional System . . . . .   | 15        |
| 2.4      | Anomalous Hall Effect . . . . .                                             | 17        |
| 2.5      | Spin Hall Effect . . . . .                                                  | 18        |
| 2.6      | Orbital Hall Effect . . . . .                                               | 20        |
| 2.6.1    | Orbital Hall Effect in Transition Metal Dichalcogenides . . . . .           | 23        |
| 2.7      | Spin-Orbit Torque . . . . .                                                 | 24        |
| 2.7.1    | Magnetization Dynamics . . . . .                                            | 25        |
| 2.8      | Spin and Orbital Torque . . . . .                                           | 25        |
| 2.8.1    | Anatomy of Spin Orbit Torque . . . . .                                      | 26        |
| 2.8.2    | Orbital and Spin Pumping . . . . .                                          | 28        |
| 2.9      | Orbital to Orbital-Current Conversion . . . . .                             | 30        |
| 2.10     | Transition Metal Dichalcogenides . . . . .                                  | 31        |
| 2.10.1   | Rare-Earth Dichalcogenides . . . . .                                        | 33        |
| <b>3</b> | <b>Density Functional Theory</b>                                            | <b>35</b> |
| 3.1      | The Many-Body Schrödinger Equation . . . . .                                | 35        |
| 3.2      | Born-Oppenheimer Approximation . . . . .                                    | 36        |
| 3.3      | Density Functional Theory . . . . .                                         | 37        |
| 3.3.1    | Hohenberg-Kohn Theorem . . . . .                                            | 37        |
| 3.3.2    | Kohn-Sham System . . . . .                                                  | 38        |
| 3.4      | Exchange-Correlation Functionals . . . . .                                  | 40        |



|          |                                                                                                                |           |
|----------|----------------------------------------------------------------------------------------------------------------|-----------|
| 3.4.1    | Local Density Approximation (LDA)                                                                              | 40        |
| 3.4.2    | Generalized Gradient Approximation (GGA)                                                                       | 40        |
| 3.5      | Spin Density Functional Theory                                                                                 | 41        |
| 3.6      | Hubbard U Correction                                                                                           | 43        |
| 3.7      | Full-Potential Linearized Augmented Plane-Wave Method                                                          | 44        |
| 3.8      | Wannier Functions                                                                                              | 47        |
| 3.8.1    | Maximally Localized Wannier Functions                                                                          | 48        |
| 3.9      | ORBITRANS                                                                                                      | 49        |
| 3.9.1    | Workflow and Methodology                                                                                       | 49        |
| <b>4</b> | <b>Engineering the Orbital Hall Effect in nonmagnetic TMDs</b>                                                 | <b>51</b> |
| 4.1      | Introduction                                                                                                   | 51        |
| 4.2      | Computational Details                                                                                          | 52        |
| 4.3      | Spin and Orbital Hall Effects in Monolayer and Bilayer H-MoS <sub>2</sub>                                      | 52        |
| 4.3.1    | Spin and Orbital Hall Effects in Monolayer H-Phase MoS <sub>2</sub>                                            | 53        |
| 4.3.2    | Analysis of Orbital Hall Conductivity in Monolayer H-MoS <sub>2</sub>                                          | 55        |
| 4.3.3    | Spin and Orbital Hall Effects in Bilayer H-Phase MoS <sub>2</sub>                                              | 55        |
| 4.4      | Spin and Orbital Hall Effects in Monolayer and Bilayer NbSe <sub>2</sub>                                       | 56        |
| 4.5      | Transport Characteristics of MoS <sub>2</sub> -NbSe <sub>2</sub> Hetero-Bilayers                               | 58        |
| 4.6      | Transport Properties as a Function of the Biaxial Strain of MoS <sub>2</sub> /WSe <sub>2</sub> Hetero-Bilayers | 61        |
| 4.7      | Spin and Orbital Hall Effects in Bulk H-Phase MoS <sub>2</sub>                                                 | 63        |
| 4.8      | Spin and Orbital Hall Effects in Bulk H-phase NbSe <sub>2</sub>                                                | 65        |
| 4.9      | Conclusion                                                                                                     | 67        |
| <b>5</b> | <b>Spin and Orbital Transport in Rare-Earth Dichalcogenides: The Case of EuS<sub>2</sub></b>                   | <b>69</b> |
| 5.1      | Introduction                                                                                                   | 69        |
| 5.2      | Computational Details                                                                                          | 71        |
| 5.3      | Electronic Structure of EuS <sub>2</sub>                                                                       | 72        |
| 5.4      | Anomalous Hall Effect in EuS <sub>2</sub>                                                                      | 74        |
| 5.5      | Spin Hall Effect in EuS <sub>2</sub>                                                                           | 77        |
| 5.6      | Orbital Hall effect in EuS <sub>2</sub>                                                                        | 78        |
| 5.7      | Orbital Berry Curvature in EuS <sub>2</sub>                                                                    | 79        |
| 5.8      | Conclusion                                                                                                     | 80        |
| <b>6</b> | <b>Promoting p-based Hall effects by <i>pdf</i> hybridization and strain in rare-earth dichalcogenides</b>     | <b>83</b> |
| 6.1      | Introduction                                                                                                   | 84        |
| 6.2      | Computational Details                                                                                          | 84        |

|          |                                                                                                             |            |
|----------|-------------------------------------------------------------------------------------------------------------|------------|
| 6.3      | Electronic Structure of the $\text{EuX}_2$ Monolayers . . . . .                                             | 85         |
| 6.4      | Electronic Structure of the $\text{GdX}_2$ Monolayers . . . . .                                             | 88         |
| 6.5      | Transport Properties of $\text{EuX}_2$ . . . . .                                                            | 91         |
| 6.6      | Transport Properties of $\text{GdX}_2$ . . . . .                                                            | 93         |
| 6.7      | Tuning Electronic Structure by Strain . . . . .                                                             | 95         |
| 6.8      | Quantized Anomalous, Spin and Orbital Hall Effect in Strained $\text{GdTe}_2$                               | 98         |
| 6.9      | Conclusion . . . . .                                                                                        | 101        |
| <b>7</b> | <b>Orbital topology, orbital torques, and orbital pumping in two-dimensional rare-earth dichalcogenides</b> | <b>103</b> |
| 7.1      | Introduction . . . . .                                                                                      | 103        |
| 7.2      | Computational Details . . . . .                                                                             | 104        |
| 7.3      | Electronic and Magnetic Properties of $\text{EuSX}$ . . . . .                                               | 106        |
| 7.4      | Transport Properties of $\text{EuSX}$ . . . . .                                                             | 110        |
| 7.4.1    | General Features of Transport Properties in $\text{EuSX}$ . . . . .                                         | 113        |
| 7.5      | Orbital Torque in $\text{EuSX}$ . . . . .                                                                   | 114        |
| 7.5.1    | Non-equilibrium Spin and Orbital Accumulation Induced by an External Electric Field . . . . .               | 116        |
| 7.6      | Orbital Pumping in $\text{EuSX}$ . . . . .                                                                  | 118        |
| 7.6.1    | Orbital to Orbital-Current Conversion . . . . .                                                             | 121        |
| 7.7      | Discussion and Conclusion . . . . .                                                                         | 122        |
| <b>8</b> | <b>Summary and Outlook</b>                                                                                  | <b>125</b> |
| 8.1      | Summary . . . . .                                                                                           | 125        |
| 8.2      | Outlook . . . . .                                                                                           | 126        |
| <b>A</b> | <b>Appendix Chapter 5</b>                                                                                   | <b>129</b> |
| <b>B</b> | <b>Appendix Chapter 6</b>                                                                                   | <b>131</b> |
| <b>C</b> | <b>Appendix Chapter 7</b>                                                                                   | <b>133</b> |
| <b>D</b> | <b>Publications</b>                                                                                         | <b>135</b> |
| D.1      | Journal articles related to this thesis . . . . .                                                           | 135        |
| D.2      | Journal articles in preparation . . . . .                                                                   | 136        |
| <b>E</b> | <b>Some Personal Reflections</b>                                                                            | <b>137</b> |
|          | <b>Bibliography</b>                                                                                         | <b>139</b> |







# List of Abbreviations

|           |                                                               |
|-----------|---------------------------------------------------------------|
| 2D        | Two-Dimensional                                               |
| AHC       | Anomalous Hall Conductivity                                   |
| BZ        | Brillouin Zone                                                |
| DFT       | Density Functional Theory                                     |
| DOS       | Density of States                                             |
| E         | Electric Field                                                |
| FLAPW     | Full-Potential Linearized Augmented Planewave                 |
| FLEUR     | <i>DFT code implementing the FLAPW method</i>                 |
| GGA       | Generalized Gradient Approximation                            |
| GGA+U     | <i>GGA with an added Hubbard U</i>                            |
| LAPW      | Linearized Augmented Planewave                                |
| LDA       | Local Density Approximation                                   |
| MT        | Muffin-Tin Sphere                                             |
| NM        | Non-Magnetic                                                  |
| OAM       | Orbital Angular Momentum                                      |
| OHC       | Orbital Hall Conductivity                                     |
| OHE       | Orbital Hall Effect                                           |
| ORBITRANS | <i>In-house code for transport calculations</i>               |
| OT        | Orbital Torque                                                |
| REDs      | Rare-Earth Dichalcogenides                                    |
| SHE       | Spin Hall Effect                                              |
| SHC       | Spin Hall Conductivity                                        |
| SOC       | Spin-Orbit Coupling                                           |
| SOT       | Spin-Orbit Torque                                             |
| STT       | Spin-Transfer Torque                                          |
| TMDs      | Transition Metal Dichalcogenides                              |
| U         | Hubbard U                                                     |
| Wannier90 | <i>Code to generate maximally localized Wannier functions</i> |





# Acknowledgement

First and foremost, I would like to express my deepest gratitude to my supervisor, **Prof. Dr. Yuriy Mokrousov**, for his invaluable guidance, patience, and continuous support throughout my PhD journey. Professor Mokrousov, your insightful advice and encouragement have been a cornerstone of my development. I deeply appreciate the time and effort you have dedicated to mentoring me, for your genius mind, and for making my PhD period such a special and positive time in my life. Your unwavering support has not only helped me academically but also personally, creating an environment that was always filled with inspiration, motivation, and everything positive. I will always be grateful for your profound influence on my growth as a researcher and as an individual.

I would also like to sincerely thank Prof. **Dr. Stefan Blügel** for giving me the opportunity to write my thesis at **the Peter Grünberg Institute, Quantum Theory of Materials (PGI-1)** and under his supervision. Professor Blügel, I am deeply thankful for your trust, support, and for creating such an inspiring and productive research environment. Being part of your institute has been a great privilege, and I am truly grateful for the chance to develop myself both professionally and personally during this journey.

I would like to thank **Dr. Dongwook Go** for his guidance and for teaching me how to perform research with precision and dedication. Dr. Go, your expertise and willingness to share your knowledge have been invaluable to my development as a researcher, and I truly appreciate your mentorship and patience throughout this journey.

I would like to express my sincere gratitude to **Dr. Jamal Ghabboun** for his guidance and support at the beginning of this journey were instrumental in shaping the path of my PhD, and I deeply appreciate your efforts.

I would like to thank numerous people for supporting me throughout my work on this thesis. I am especially grateful to Dr. Tom Saunderson, Dr. Johanna P. Carbone, Dr. Sumit Ghosh, and Dr. Hiroshi Katsumoto for their help and support

---

in daily discussions about physics and life. Your insights, encouragement, and friendship have made this journey not only productive but also truly enjoyable.

I would like to express my deepest gratitude to my mother for her endless love, and constant encouragement throughout my PhD journey. I also wish to honor the memory of my late father, whose guidance, values, and dreams continue to inspire me every day. I am incredibly thankful to my brothers, sisters, and friends for their steadfast belief in me and their continuous support. Your care and encouragement have been a source of strength and motivation, making this journey not only possible but also truly meaningful.

I would like to thank my wonderful wife **Walaa** for her support, patience, and love throughout my PhD journey. Your encouragement and understanding have been a constant source of strength for me.

To my dear children, **Qays**, **Layan**, and **Yamen**, it has been a great joy to watch you grow during this period. Balancing my PhD work and family duties has not always been easy, but it has been one of the most rewarding experiences of my life. You have filled my days with love, laughter, and inspiration, and striving to be a good father while pursuing my academic goals has brought me immense happiness.

The **color code** in the bottom left or right corner of each page represents my family members, from left to right: Pink represents my daughter, Layan; double dark blue represents my sons, Qays and Yamen; gold represents my wife, and olive represents me.



# Introduction

# 1

Historically and consistently up to the present moment, human beings have developed countless innovative tools and technologies to overcome ongoing challenges, progressing from simple tools to increasingly complex systems. In today's world, rapid technological advancements in fields such as artificial intelligence and big data have resulted in massive volumes of data that conventional storage and processing methods, such as hard disk drives and silicon-based systems, can no longer handle effectively. Hard disk drives face capacity barriers and performance limitations when dealing with large-scale data [1], while silicon-based computing technologies are approaching their physical limits in design complexity, processing power, and energy consumption [2]. This demand highlights the need for new materials and techniques to manage information more efficiently. One promising approach is the development of two-dimensional (2D) materials, which, along with the emerging field of spin-orbitronics [3], offer potential for creating faster, more efficient data processing and storage technologies suited to the demands of today's information-driven era [4].

The discovery of graphene in 2004 [5] marked a significant milestone in materials science, paving the way for extensive research into a diverse range of 2D materials, including transition metal dichalcogenides (TMDs) [6–8], hexagonal boron nitride [9], and black phosphorus [10]. These materials exhibit unique physical and chemical properties, enhancing their significance in materials science and expanding their applications in modern nanotechnology [11]. Notably, 2D materials display exceptional mechanical, electronic, and optical properties that are absent in their bulk counterparts [12]. Furthermore, materials such as TMDs can be precisely engineered through strain, composition tuning, and heterostructure assembly, unlocking their potential for advanced technological applications [13]. Additionally, many 2D materials, including TMDs, are relatively easy to synthesize, making them highly accessible for experimental and industrial purposes [14].

Significant progress has been made in exploring the magnetic properties of 2D materials, primarily due to their potential applications in spintronics and quantum computing. Materials such as  $\text{CrI}_3$  [15],  $\text{Fe}_3\text{GeTe}_2$  [16], and bilayer  $\text{Cr}_2\text{Ge}_2\text{Te}_6$  [17, 18]



---

have significantly advanced the study of novel 2D ferromagnetic materials and their unique magnetic properties. Among these materials, 2D TMDs are particularly noteworthy. Several representative TMD compounds, including metallic  $\text{VX}_2$  [19–21],  $\text{CrX}_2$  [21–23], and semiconducting  $\text{MnX}_2$  ( $\text{X} = \text{S}, \text{Se}, \text{Te}$ ) [21, 24, 25], exhibit magnetic ordering in their ground states, drawing considerable attention for their strong potential as platforms for various spintronic effects. Additionally, the unique combination of low crystal symmetry and spin-orbit coupling (SOC) in TMDs allows these materials to host exotic excitations and demonstrate diverse transport properties, such as anomalous, spin, and orbital Hall conductivities.

However, the magnetism in TMDs tends to be relatively fragile. Addressing this limitation involves incorporating  $4f$  electrons from rare-earth atoms into 2D materials, leading to the development of 2D rare-earth dichalcogenides (REDs). These localized  $4f$  electrons play a pivotal role in generating high magnetic moments, which contribute to robust magnetocrystalline anisotropies when combined with the strong SOC and unique crystal field in 2D geometry [26–29]. Several compounds of  $4f$  elements, such as Eu with chalcogenides, are experimentally known in bulk, at interfaces, and in thin films, *e.g.*, based on  $\text{EuS}$  [30–34],  $\text{EuSe}_2$  [35], and  $\text{EuTe}_2$  [36], offering curiosity to study these compounds in 2D material monolayers. These 2D REDs offer substantial advantages for the development of advanced magnetic storage devices and spintronics applications.

Orbitronics is a field within spintronics that focuses on exploiting the orbital degree of freedom of electrons rather than their spin degree of freedom. Orbitronics, holding significant potential to shape information technology, has emerged as a field capable of developing eco-friendly electronic devices [37]. By harnessing the unique properties of electron’s orbital angular momentum (OAM), which can be generated in a highly efficient manner by the direct transfer of angular momentum from the lattice, orbitronics opens up new possibilities for energy-efficient technologies that can reduce environmental impact and optimize device performance [38]. The study of OAM offers the opportunity to explore novel data storage and processing mechanisms, which could revolutionize modern computing [39, 40]. The OAM is the pivotal degree of electronic freedom, which can result in various phenomena such as the orbital Hall effect (OHE) [41, 42], orbital magnetoelectric effects, and orbital torques [43–45]. In this context, orbital torque and orbital pumping emerge as critical mechanisms in orbitronics. Orbital torque, akin to spin torque in spintronics [44, 46, 47], allows for efficient manipulation of magnetization via the transfer of orbital angular momentum. Similarly, orbital pumping facilitates the generation of orbital currents through dynamic coupling between orbitals and magnetic systems.

Together, these mechanisms bridge theoretical insights with practical applications, advancing the potential of orbitronics in modern technology.

In addition to their magnetic properties, our interest in REDs is strongly motivated by their potential to advance the study of the orbital Hall effect (OHE), particularly in complex magnetic systems. The essence of OHE lies in the interaction between the orbital motion of electrons and an applied electric field, which induces a transverse flow of orbital angular momentum, as opposed to spin angular momentum in the spin Hall effect (SHE) [48–53]. To date, OHE has been studied predominantly in nonmagnetic crystals [54], and relatively little is known about orbital currents in magnetic systems, especially in low-dimensional contexts. REDs provide a unique platform for exploring OHE due to the presence of strongly localized  $4f$  electrons, which generate significant magnetic moments and robust magnetocrystalline anisotropies. Additionally, the  $4f$  electrons play a crucial role in enhancing the magnetism of the  $p$ -state chalcogenide atoms, thereby contributing to the transport properties of these materials. This combination of strong magnetism and SOC in REDs makes them ideal candidates for studying the interplay between magnetism and orbital transport phenomena.

The OHE can be utilized for magnetization control in spin-orbit torque devices relying on an injection of an orbital current, which often originates from light elements and their surfaces [43, 55–59]. In the complex magnetic environment of REDs, orbital Hall currents can lead to measurable effects, such as inducing orbital torques on magnetization [43, 60], or generating strong non-local spin currents via SOC [61]. This creates an entirely new channel for information transfer—the orbital channel—which is expected to push the boundaries of spintronics towards the emerging field of orbitronics [62]. By leveraging the distinctive properties of REDs, particularly their strong SOC and magnetism, we anticipate that further exploration of OHE in these materials will lead to significant breakthroughs in the manipulation of quantum states and the development of next-generation spin-orbitronics devices.

The Janus structure of REDs, characterized by its inherent inversion symmetry breaking, significantly enhances spin-related properties. Examples like EuSP, EuSSe, and EuSCl exhibit notable spin-orbit torques (SOT) due to the substantial orbital angular momentum (OAM) response to electric fields, predominantly from Eu's  $4f$ -electrons. These torques facilitate orbital pumping, where magnetization dynamics generates an orbital current, a process beneficial for spin-orbitronic applications. The efficiency of orbital torque in REDs makes them promising candidates



---

for low-dimensional spintronic devices, as they can produce strong in-plane currents of angular momentum, potentially revolutionizing memory technologies.

The Berry curvature arises from the geometric (Berry) phase acquired by Bloch electronic wavefunctions during their adiabatic evolution in crystal-momentum  $\mathbf{k}$  space [63]. It plays a central role in transport phenomena such as the AHE, SHE, and OHE. REDs provide a particularly fertile platform for exploring these effects due to their geometries, which can give rise to nontrivial topological features in the electronic structure and contribute to quantized Hall responses. As a result, REDs enable systematic investigation of Berry-curvature-driven Hall effects.

In this thesis, first-principles density functional theory (DFT) calculations using the full-potential linearized augmented plane-wave (FLAPW) method are employed to investigate the structural, electronic, magnetic, and transport properties of 2D REDs. Applying DFT with the GGA+ $U$  approach enables accurate modeling of the strong on-site Coulomb repulsion within the  $4f$  orbitals, which is crucial for predicting the complex behavior of rare-earth elements in these materials. Additionally, Wannier functions are utilized to construct accurate tight-binding models from the DFT results, enabling detailed calculations of transport properties and spin-orbit torques with high computational efficiency. These *ab initio* calculations provide an in-depth understanding of the band structure, density of states, various Hall conductivities, and spin-orbit torques, which are fundamental to evaluating the transport properties of REDs and their potential applications in spintronics and orbitronics.

This thesis explores key questions regarding the impact of rare-earth elements, specifically europium (Eu) and gadolinium (Gd), on the magnetic and electronic properties of 2D REDs within various chalcogenide compounds and structures. Central to this investigation are the roles of spin-orbit coupling (SOC), Berry curvature, and  $p$ - $d$ - $f$  hybridization in shaping the transport properties of REDs. Additionally, this study examines how strain engineering can be employed to modulate Hall effects in REDs, potentially inducing quantized transport phenomena. Finally, this research assesses the potential of REDs as platforms for realizing topological quantum states, with promising applications in the fields of spintronics and orbitronics. **The following provides an overview of the thesis structure:**

- **Chapter 2:** This chapter presents the theoretical foundations of spinorbit coupling (SOC), Berry curvature, Hall effects, and spin and orbital torques, with particular focus on their roles in spin and orbital transport phenomena in two-dimensional materials.

- **Chapter 3:** This chapter describes the density functional theory (DFT) and GGA+ $U$  approaches, detailing how these methods are employed to model the electronic structure and magnetic properties of REDs. It also discusses the theoretical foundation of the Full-Potential Linearized Augmented Plane-Wave (FLAPW) method, as implemented in the FLEUR code, which is used for all calculations in this thesis. Furthermore, the construction of maximally localized Wannier functions is presented, enabling a more accurate description of electronic properties.
- **Chapter 4:** In this chapter, we begin by reproducing the spin Hall conductivity (SHC) and orbital Hall conductivity (OHC) in the H-phase monolayer and bilayer of MoS<sub>2</sub> [51]. Following this, we investigate the SHC and OHC in the metallic H-phase monolayer and bilayer NbSe<sub>2</sub>, highlighting the plateau-like behavior of the OHC in the bandgap above the Fermi level. Furthermore, we examine the SHC and OHC transport properties in MoS<sub>2</sub>/NbSe<sub>2</sub> heterostructures. Finally, we address the SHC and OHC in MoS<sub>2</sub>/WS<sub>2</sub> under biaxial strain, demonstrating the robustness of the orbital Chern number in these materials.
- **Chapter 5:** This chapter delves into the spin and orbital transport properties of 2D REDs, with a focus on H-phase monolayer of EuS<sub>2</sub> as a representative material. Leveraging first-principles calculations, the study investigates the influence of 4*f* electrons in generating large magnetic moments and robust spin-orbit coupling (SOC), which are essential for realizing substantial spin and orbital Hall conductivities. A key finding presented is the sensitivity of EuS<sub>2</sub>s electronic structure to the Coulomb repulsion parameter  $U$ , which modulates hybridization between Eu's 4*f* and Ss 3*p* states, significantly impacting transport properties. This chapter also explores the AHC, SHC, and OHC revealing that the OHE can reach magnitudes six times greater than the SHE, making EuS<sub>2</sub> a potential platform for orbitronics.
- **Chapter 6:** This chapter investigates the enhancement Hall effects in Gd- and Eu-based REDs, focusing specifically on *p-d-f* hybridization in monolayers of EuS<sub>2</sub>, EuSe<sub>2</sub>, EuTe<sub>2</sub>, GdS<sub>2</sub>, GdSe<sub>2</sub>, and GdTe<sub>2</sub>. Using first-principles calculations, the chapter demonstrates how the unique electronic structures of these REDs – characterized by high magnetic moments and wide band gaps – promote substantial anomalous, spin, and orbital Hall conductivities. In Gd-based compounds, the spin-polarized *p* states near the Fermi level significantly contribute to transport properties, while EuS<sub>2</sub>, EuSe<sub>2</sub> and EuTe<sub>2</sub>

---

exhibit a slightly different electronic configuration that enhances interactions with their  $4f$  states. The chapter also explores the effects of strain in tuning these Hall conductivities, with  $\text{GdTe}_2$  displaying strain-induced orbital inversion that leads to quantized charge, spin, and orbital transport. These findings underscore the versatility of Gd- and Eu-based REDs as promising materials for realizing topological phases and advancing spintronic and orbitronic applications.

- **Chapter 7:** This chapter highlights the unique role of orbital angular momentum (OAM) in mediating spin-orbit torques and orbital pumping in 2D REDs. Focusing on Janus-type ferromagnetic monolayers of Eu-based compounds like EuSP, EuSSe, and EuSCL, this study demonstrates how the strong orbital response of  $4f$  electrons under current generates significant orbital torques. These torques arise primarily from the robust coupling of OAM with spin-orbit interaction, leading to effective current-driven magnetization dynamics. The chapter also explores the reciprocal process of orbital pumping, wherein magnetization dynamics induces in-plane currents of orbital angular momentum, offering a pathway for orbital current generation. These findings highlight the potential of Eu-based REDs as a versatile platform for advanced spin-orbitronic applications, with the non-trivial directional polarization of orbital currents offering novel functionalities for information transfer in 2D materials.
- **Chapter 8:** This chapter summarizes the key findings and provides suggestions for future research directions.

In this chapter, I present the theoretical background of spin and orbital transport in 2D materials. I begin by introducing the fundamentals of magnetism, followed by spin-orbit coupling, and then provide a detailed discussion of Berry curvature and its role in topological transport phenomena. I then discuss the anomalous, spin, and orbital Hall effects, highlighting their physical origins and differences. Afterwards, I introduce the concepts and mechanisms of spin-orbit torque and orbital torque, which are essential for understanding current-driven magnetization dynamics. Finally, I discuss orbital pumping and orbital-to-orbital current conversion, presenting their roles in the generation and manipulation of orbital currents.

## 2.1 Magnetism

### 2.1.1 Fundamentals of Magnetism

Magnetism is a fundamental natural phenomenon arising from the motion of electric charges, such as electrons, and their intrinsic properties, including spin and orbital angular momentum [64]. In magnetic materials, the magnetic moments of electrons are the primary contributors to magnetism, with their alignment governed by exchange interactions. These magnetic moments comprise two key components: the intrinsic spin magnetic moment and the orbital magnetic moment. In most cases, particularly in 3d transition metals, magnetism is predominantly governed by the spin magnetic moment, while the contribution of the orbital magnetic moment is often considered relatively small in bulk materials. For instance, in bulk bcc Fe, the spin magnetic moment is approximately  $2.34 \mu_B$ , whereas the orbital magnetic moment is significantly smaller, measuring around  $0.08 \mu_B$  [65].

The magnetic moment  $\mathbf{m}$  that arises from the spin angular momentum  $\mathbf{S}$  of the electrons can be expressed as:

$$\mathbf{m}_{\text{spin}} = -\frac{1}{\hbar} \mu_B g_e \mathbf{S}, \quad (2.1)$$



where  $\mu_B$  is the Bohr magneton, defined as  $\mu_B = \frac{e\hbar}{2m_e}$ ,  $g_e$  is the Landé  $g$ -factor (approximately 2.0023 for free electrons),  $e > 0$  is the elementary unit of charge,  $\hbar$  is the reduced Planck's constant, and  $m_e$  is the mass of the electron.

Similarly, the orbital magnetic moment  $\mathbf{m}_{\text{orb}}$  originates from the motion of electrons along their orbital paths around the nucleus and is directly related to the orbital angular momentum  $\mathbf{L}$  as expressed by:

$$\mathbf{m}_{\text{orb}} = -\frac{1}{\hbar}\mu_B \sum_{\alpha} \mathbf{L}_{\alpha}, \quad (2.2)$$

where  $\mathbf{L}_{\alpha} = \mathbf{r}_{\alpha} \times (m_e \mathbf{v}_{\alpha})$  represents the angular momentum of the  $\alpha$ -th electron. The orbital magnetic moment of an electron is oriented opposite to its orbital angular momentum due to the negative charge of the electron. Together, the spin and orbital components define the total magnetic moment of an electron, expressed as:

$$\mathbf{m} = -\frac{1}{\hbar}\mu_B(\mathbf{L} + 2\mathbf{S}). \quad (2.3)$$

As described earlier, the spin magnetic moment typically dominates in most materials; however, the orbital contribution can become significant in specific cases, particularly in systems with strong spin-orbit coupling and low crystal symmetry or orbital degeneracy in combination with strong Coulomb interaction.

### 2.1.2 Origin of Magnetism in Solids

In an isolated atom, a single electron can be characterised by the four quantum numbers  $n$  (the principal quantum number),  $l$  (the orbital angular momentum quantum number),  $m_l$  (the magnetic quantum number specifying the orientation of the orbital angular momentum), and  $m_s$  (the spin projection quantum number), and thus the magnetic moment of the single electron arises from its orbital angular momentum and the intrinsic and intrinsic spin (see Eq. (2.3)). The magnetism of multi-electron systems is a result of the Pauli exclusion principle that the total wavefunction of a system of electrons must be antisymmetric under exchange of any two electrons. As a result, each atomic orbital  $(n, l, m_l)$ , can hold at most two electrons, and they must have opposite spins. In atoms with partially filled degenerate subshells, intra-atomic electron-electron interactions combined with the antisymmetric nature of the total electron wave function lift the degeneracy of single-particle states. Coulomb repulsion and intra-atomic exchange interactions favor configurations in which electrons occupy different orbitals with parallel spins, maximizing the total spin  $S$  (Hunds first rule), because this lowers the energy through exchange.

For a given  $S$ , the electrons arrange their orbital magnetic quantum numbers to maximize the total orbital angular momentum  $L$  (Hunds second rule), and spinorbit coupling then selects the total angular momentum  $J$  of the ground state (Hunds third rule). Together, these rules determine the atoms ground-state term and its magnetic moment.

Atoms with filled subshells, such as noble gases, exhibit no net magnetic moment due to spin cancellation. This atomic physics is particularly evident in the atomic-like, localized  $4f$ -electrons of the rare-earth elements of the REDs investigated in this thesis.

In solid state systems, valence electrons delocalize between atoms, intra-atomic exchange extends to inter-atomic exchange that competes with the kinetic energy of electrons. Thus, the strength of the Coulomb interaction, the hopping of the electrons, the overlap of their wave functions, the degeneracy of their states determine whether solids are nonmagnetic or have a magnetic ground state and what magnetic order they adopt, *e.g.* ferro- or antiferromagnetic or even a more complex magnetic order. In solids orbital moments are typically quenched by the crystal fields with exceptions mentioned above.

Magnetism prominently arises in systems with localized electrons, which means for the purpose of this thesis such as the  $3d$ -orbitals in transition metals and the  $4f$ -orbitals in rare-earth elements. These systems, characterized by near-degenerate orbital energy levels (defined by  $m_l$ ), are pivotal in enabling robust magnetic behavior.

## 2.2 Spin-Orbit Coupling

Exploring spin-orbit coupling (SOC) within the framework of the Dirac equation represents a significant milestone in our understanding of the quantum behavior of electrons. The SOC arises from the relativistic interaction between the electron's spin angular momentum and its orbital angular momentum around the nucleus. To understand the origin of SOC, consider an electron orbiting in the electric field generated by the nucleus. From electron's perspective, it perceives the positively charged nucleus as rotating around itself. The relative motion induces a magnetic field in the electron's rest frame. This effective magnetic field interacts with the electron's spin magnetic moment and the interaction energy differs if the spin is parallel or antiparallel to the orbital angular momentum. In solids, SOC lifts the degeneracy of bands of the same principle quantum number  $n$  and the same orbital

quantum number  $l$  but different  $j$ , and links in general band topology to spin-splitting and to magnetization.

Typically the interaction energy ranges from a few to several hundred meV. Consequently, SOC is treated as a perturbation to the non-relativistic Hamiltonian. For a spherically symmetric potential, this interaction can be expressed as

$$H_{\text{SOC}} = \frac{1}{2m_e^2 c^2} \frac{1}{r} \frac{\partial V(r)}{\partial r} \mathbf{L} \cdot \mathbf{S} = \xi(r) \mathbf{L} \cdot \mathbf{S}, \quad (2.4)$$

where  $\xi(r)$  is the radial SOC parameter,  $V(r)$  represents the electrostatic potential energy at a point located at a distance  $r$  from the center of the atom and  $\mathbf{L}$  and  $\mathbf{S}$  denote the angular momentum and spin operator of the electron, respectively. Here,  $c$  is the speed of light, and  $m_e$  is the mass of a free electron. Due to the Coulomb-like form of the potential  $V(r)$  near the atomic nucleus, the SOC constant  $\xi = \langle \xi(r) \rangle$  is a significantly increasing function of the atomic number  $Z$ . Electronic structure calculations [66] reveal that within a series of atoms in the periodic system,  $\xi$  increases proportional to the atomic number  $Z^2$ .

Consequently,  $\xi$  increases for heavier elements, and thus heavier chemical elements as those in the  $4d$ ,  $5d$ , and  $4f$  series are typically selected when strong spin-orbit coupling is required. In reality, most modern first-principles methods include the spin-orbit interaction on top of the scalar-relativistic approximation of the Dirac equation [67], which effectively means that the mass of the free electron is replaced by the velocity-dependent relativistically enhanced mass of the electrons.

Despite its relatively weak magnitude, SOC is a crucial factor in various quantum phenomena, including the anomalous Hall effect, the spin-Hall effect, and spin-orbit torques.

## 2.3 Berry Phase

Berry curvature is a fundamental concept in materials science, essential for understanding phenomena such as the anomalous Hall effect, spin Hall effect, topological insulators, and magnetization dynamics [63, 68]. It arises from the *geometric phase*, a concept that has historical roots in multiple fields. The modern quantum mechanical formulation of the *Berry phase* was introduced by Michael Berry in 1984 [69].

In quantum systems undergoing *adiabatic parameter changes*, the system's Hamiltonian evolves gradually, leading to the accumulation of a phase factor consisting of

both a *dynamical phase* and a *geometric (Berry) phase* [70]. A deep understanding of Berry curvature begins with the Berry phase, which characterizes the geometric phase accumulated by a quantum state under cyclic evolution. This implies that the final and initial states may differ by a constant phase factor, which remains gauge-invariant modulo  $2\pi$  [71]. In the quantum realm, the Berry curvature phenomenon emerges as a cornerstone of geometric phases, elegantly captured by the following fundamental equations. The time-dependent Schrödinger equation with parametric dependence is given as:

$$H(\lambda(t)) |n(\lambda(t))\rangle = E_n(\lambda(t)) |n(\lambda(t))\rangle, \quad (2.5)$$

where  $H(\lambda(t))$  represents the Hamiltonian, which depends on a set of time-varying parameters  $\lambda(t) = (\lambda_1(t), \lambda_2(t), \dots, \lambda_n(t))$ . Here,  $|n(\lambda(t))\rangle$  denotes the instantaneous eigenstates of the Hamiltonian, and  $E_n(\lambda(t))$  are the corresponding eigenenergies. To account for the phase evolution of the quantum state, the wave function is expressed as:

$$|\psi(t)\rangle = c(t)e^{-i\Phi(t)} |n(\lambda(t))\rangle, \quad (2.6)$$

where  $c(t)$  represents time-varying expansion coefficients, and  $\Phi(t)$  is the dynamical phase factor, given by:

$$\Phi(t) = \frac{1}{\hbar} \int_0^t E_n(\lambda(t')) dt'. \quad (2.7)$$

Substituting the eigenstate equation (2.5) into the time-dependent Schrödinger equation:

$$H(\lambda(t)) |\psi(t)\rangle = i\hbar \frac{\partial}{\partial t} |\psi(t)\rangle, \quad (2.8)$$

we derive the equation governing the time evolution of the coefficient  $c(t)$ :

$$\dot{c}(t) = ic(t)A_n(t), \quad (2.9)$$

where  $A_n(t) = \langle n(\lambda(t)) | i \frac{\partial}{\partial t} | n(\lambda(t)) \rangle$  is the Berry connection, a key quantity encoding the geometric properties of the system. The Berry phase  $\gamma(t)$ , a geometric phase acquired by the quantum state, is then defined as:

$$\gamma(t) = \int_0^t A_n(t') dt'. \quad (2.10)$$

This phase can be reformulated in terms of the parameters  $\lambda(t)$ . For slow variations of  $\lambda(t)$ , the adiabatic approximation allows us to apply the chain rule:

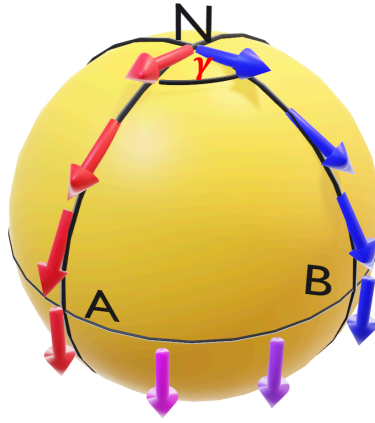
$$\frac{\partial}{\partial t} |n(\lambda(t))\rangle = \dot{\lambda} \cdot \nabla_{\lambda} |n(\lambda(t))\rangle, \quad (2.11)$$



which transforms the Berry phase into a path integral in parameter space:

$$\gamma(t) = \int_{\lambda_i}^{\lambda_f} A_n(\lambda) \cdot d\lambda, \quad (2.12)$$

where  $A_n(\lambda) = \langle n(\lambda) | i\nabla_\lambda | n(\lambda) \rangle$  is the Berry connection in parameter space. This profound result reveals that the Berry phase does not depend on the eigen energy of the state, but on the wave function  $|n\rangle$  and depends solely on the path traversed in parameter space, independent of the rate of traversal, provided the evolution is adiabatic. This geometric phase is behind a wide range of phenomena in quantum mechanics, from the Aharonov-Bohm effect to topological insulators, highlighting the deep interplay between geometry and quantum dynamics.



**Fig. 2.1.:** When a tangent vector is parallel transported along a closed path on a sphere, starting from the north pole N through points A and B back to N, it becomes misaligned with the original vector due to the sphere's non-trivial curvature. The red and blue vectors illustrate this misalignment; red represents the original vector, while blue represents the transported vector. The angle  $\gamma$ , referred to as a geometric phase, arises during this process of parallel transport. Figure inspired by [72].

### 2.3.1 Geometric Properties of the Berry Curvature in the Brillouin Zone

At the heart of this geometric framework lies a remarkable insight: the relationship described in (2.12) depends not on time but on the intrinsic geometry of the Berry

connection. Specifically, it reflects the influence of the Berry vector potential along the path  $C$ . When  $\lambda_i = \lambda_f$ , the geometric phase is given as follows:

$$\gamma = \oint_C i \langle n(\lambda) | \nabla_\lambda | n(\lambda) \rangle \cdot d\lambda = \oint_C \mathbf{A}(\lambda) \cdot d\lambda \quad (2.13)$$

Stokes' theorem allows for the transformation of the line integral in equation (2.13) over the closed path  $C$  into a surface integral over the surface  $S$  enclosed by the loop  $C$ :

$$\gamma = \oint_C \mathbf{A}(\lambda) \cdot d\lambda = \int_S \Omega(\lambda) \cdot dS, \quad (2.14)$$

where  $\Omega(\lambda) = \nabla_\lambda \times \mathbf{A}(\lambda)$ , mirroring the electromagnetic analogue, akin to the curl of the Berry vector potential in electromagnetics. This quantity, termed the Berry curvature, characterizes the geometric properties of the system. When the Berry vector potential is a gradient field of a smooth function, the Berry curvature field vanishes across the surface  $S$ , indicating a trivial Berry phase due to the pure gauge condition. However, deviations from this pure gauge scenario lead to non-trivial geometric or topological phases [72]. The Berry curvature, represented as a rank-2 antisymmetric tensor, has its components defined as follows:

$$\Omega_{ij}(\lambda) = \frac{\partial}{\partial \lambda_i} A_j(\lambda) - \frac{\partial}{\partial \lambda_j} A_i(\lambda) = -2 \operatorname{Im} \left\langle \frac{\partial}{\partial \lambda_i} n_\lambda \left| \frac{\partial}{\partial \lambda_j} n_\lambda \right. \right\rangle. \quad (2.15)$$

Within the parallel transport gauge, a highly useful expression for the curvature can be derived, allowing the perturbation theory expression for the derivative of  $|n_\lambda\rangle$  in (2.15) to be formulated as follows:

$$\frac{\partial}{\partial \lambda_i} |n_\lambda\rangle = \sum_{m \neq n} \frac{\langle m_\lambda | \frac{\partial}{\partial \lambda_i} H(\lambda) | n_\lambda \rangle}{E_n(\lambda) - E_m(\lambda)} |m_\lambda\rangle, \quad (2.16)$$

consequently, we obtain the gauge-invariant expression for the curvature:

$$\Omega_{ij}(\lambda) = -2 \operatorname{Im} \sum_{m \neq n} \frac{\langle n_\lambda | \frac{\partial}{\partial \lambda_i} H(\lambda) | m_\lambda \rangle \langle m_\lambda | \frac{\partial}{\partial \lambda_j} H(\lambda) | n_\lambda \rangle}{(E_n(\lambda) - E_m(\lambda))^2}. \quad (2.17)$$

### 2.3.2 Berry Curvature for Bloch Electrons in a Two-Dimensional System

In a two-dimensional (2D) periodic solid, the eigenstates of the Hamiltonian for electrons can be described by quantum numbers  $(\mathbf{k}, n)$ , where  $\mathbf{k} = (k_x, k_y)$  lies

within the 2D Brillouin zone and  $n$  is a discrete index labeling the energy bands. The eigenfunctions  $\psi_{n\mathbf{k}}(\mathbf{r})$  take the form:

$$\psi_{n\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} u_{n\mathbf{k}}(\mathbf{r}), \quad (2.18)$$

where  $u_{n\mathbf{k}}(\mathbf{r})$  is a function with the same periodicity as the 2D crystal lattice. This allows the Schrödinger equation to be rewritten in terms of a  $\mathbf{k}$ -dependent Hamiltonian  $H(\mathbf{k})$ :

$$H(\mathbf{k})u_{n\mathbf{k}}(\mathbf{r}) = \epsilon_{n\mathbf{k}}u_{n\mathbf{k}}(\mathbf{r}), \quad (2.19)$$

with  $H(\mathbf{k}) \equiv H_{\mathbf{k}} = e^{-i\mathbf{k}\cdot\mathbf{r}} H e^{i\mathbf{k}\cdot\mathbf{r}}$ . Here,  $H_{\mathbf{k}}$  acts on the Hilbert space of periodic functions, making the problem suitable for analyzing geometric phases and topological effects [63, 73]. In this context, the parameter  $\lambda$  in the general theory of Berry phases is identified with the 2D crystal momentum  $\mathbf{k} = (k_x, k_y)$ . The Berry connection for a non-degenerate band  $n$  is given by:

$$\mathbf{A}_n(\mathbf{k}) = i\langle u_{n\mathbf{k}} | \nabla_{\mathbf{k}} u_{n\mathbf{k}} \rangle, \quad (2.20)$$

and the Berry curvature for the 2D system is:

$$\Omega_n(\mathbf{k}) = -2 \operatorname{Im} \left\langle \frac{\partial}{\partial \mathbf{k}_x} u_{n\mathbf{k}} \left| \frac{\partial}{\partial \mathbf{k}_y} u_{n\mathbf{k}} \right. \right\rangle. \quad (2.21)$$

Using perturbation theory, the Berry curvature in  $\mathbf{k}$ -space can be expressed as:

$$\Omega_n(\mathbf{k}) = -2 \operatorname{Im} \sum_{m \neq n} \frac{\langle u_{n\mathbf{k}} | \frac{\partial}{\partial \mathbf{k}_x} H_{\mathbf{k}} | u_{m\mathbf{k}} \rangle \langle u_{m\mathbf{k}} | \frac{\partial}{\partial \mathbf{k}_y} H_{\mathbf{k}} | u_{n\mathbf{k}} \rangle}{(E_{n\mathbf{k}} - E_{m\mathbf{k}})^2}. \quad (2.22)$$

This expression highlights the interplay between different bands and their contributions to the Berry curvature. In a 2D system, the Berry curvature  $\Omega_n(\mathbf{k})$  acts as an effective magnetic field in reciprocal space. It influences the dynamics of electrons under external perturbations, such as in-plane electric fields. For example, an in-plane electric field  $\mathbf{E}$  can drive electrons along specific orbits in  $\mathbf{k}$ -space, leading to a transverse velocity proportional to the Berry curvature.

To calculate the Berry curvature in momentum space, it is crucial to take into consideration the symmetry properties. When time-reversal symmetry is present in a crystal,  $\Omega_n(\mathbf{k}) = -\Omega_n(-\mathbf{k})$ , leading to a zero integral of Berry curvature over all occupied states. Conversely, in the presence of spatial inversion symmetry,  $\Omega_n(\mathbf{k}) = \Omega_n(-\mathbf{k})$ . Therefore, if a crystal exhibits both time-reversal and spatial inversion symmetries,  $\Omega_n(\mathbf{k}) = 0$ , indicating that the Berry curvature vanishes

throughout the Brillouin zone. So, to obtain a non-zero Berry curvature, a crystal must exhibit broken inversion symmetry or broken time-reversal symmetry.

## 2.4 Anomalous Hall Effect

It required nearly a century to unravel the enigma of the Anomalous Hall Effect (AHE). The phenomenon was first observed as an extraordinarily large Hall effect in ferromagnetic materials such as nickel (Ni) and cobalt (Co) [74]. In 1879, Edwin H. Hall discovered that when an electric current  $j_y$  flows through a conductor, charge carriers experience a Lorentz force in the presence of an external magnetic field  $H_z$ , giving rise to a transverse voltage perpendicular to the current. One year later, Hall discovered that this effect is tenfold larger in ferromagnetic iron than in nonmagnetic conductors. To address this difference, a practical equation for the Hall resistivity was introduced to describe the resistivity governing this effect:

$$\rho_{xy}^H = \rho^{\text{OHE}} + \rho^{\text{AHE}} = R_0 H_z + R_s M_z, \quad (2.23)$$

where the first term corresponds to the resistivity of the ordinary Hall effect (OHE) – not to be confused with the orbital Hall effect discussed in section 2.6 – which is proportional to the applied magnetic field  $H_z$  and arises due to the Lorentz force on charge carriers. The second term represents the anomalous Hall effect (AHE) and depends on the magnetization  $M_z$ ; it can be observed even in the absence of an external magnetic field. Here,  $R_0$  and  $R_s$  are material-dependent coefficients.

In 1954, Karplus and Luttinger provided the foundational understanding of the intrinsic AHE. They revealed that when an electric field is applied to a perfect crystal with spin-orbit coupling (SOC), the electrons gain an extra component to their group velocity, known as the anomalous velocity. The common explanation of AHE is based on the role of SOC. This interaction influences the trajectory of electrons by curving the paths of those with up and down spins in opposite directions. When these electrons are subject to exchange splitting, an imbalance arises between the populations of up-spin and down-spin electrons, leading to the emergence of a net Hall current (see Figure 2.2). Intriguingly, this effect is rooted in the band structure of the material; the concept of anomalous velocity is often associated with the Berry curvature. The interaction between the electric field and the Berry phase curvature in momentum space results in a spatial separation of electrons with different spins, driving them in opposite directions perpendicular to the field [68].

The AHC follows the same symmetry arguments as the Berry curvature discussed in the previous section. Therefore, we can observe AHC when the Berry curvature is nonzero, which occurs upon breaking either time-reversal symmetry or spatial inversion symmetry. Consequently, AHC is nonzero in ferromagnetic materials.

The intrinsic contribution to the 2D AHC is defined as the DC limit of the interband conductivity, derived from the Kubo formula [75]:

$$\sigma_{xy}^{\text{AH}} = -\frac{e^2}{\hbar} \sum_n \int_{\text{BZ}} \frac{d^2 \mathbf{k}}{(2\pi)^2} f_{n\mathbf{k}} \Omega_{n\mathbf{k}}, \quad (2.24)$$

where  $f_{n\mathbf{k}}$  is the Fermi-Dirac distribution function, and  $\Omega_{n\mathbf{k}}$  is the Berry curvature of the  $n$ -th band at momentum  $\mathbf{k}$ . The Berry curvature is given by:

$$\Omega_{n\mathbf{k}} = 2\hbar^2 \sum_{m \neq n} \text{Im} \left[ \frac{\langle u_{n\mathbf{k}} | v_x | u_{m\mathbf{k}} \rangle \langle u_{m\mathbf{k}} | v_y | u_{n\mathbf{k}} \rangle}{(E_{n\mathbf{k}} - E_{m\mathbf{k}} + i\eta)^2} \right], \quad (2.25)$$

where  $E_{n\mathbf{k}}$  is the energy of the Bloch state,  $u_{n\mathbf{k}}$  is the lattice-periodic part of the Bloch wave function, and  $v_i$  is the  $i$ -th component of the velocity operator related to the Hamiltonian  $H$  as:

$$v_i = \frac{1}{\hbar} \frac{\partial H}{\partial k_i}, \quad i = x, y.$$

The anomalous velocity, arising from the Berry curvature, is expressed as:

$$\mathbf{v}_{\text{anomalous}} = \frac{e}{\hbar} \mathbf{E} \times \boldsymbol{\Omega}_{n\mathbf{k}},$$

this anomalous velocity induces a transverse electron flow, leading to the intrinsic AHC, which is proportional to the integral of the Berry curvature over the occupied states in the Brillouin zone. In this context, the Berry curvature acts as an effective magnetic field in momentum space, deflecting electron motion analogous to the Lorentz force.

## 2.5 Spin Hall Effect

The Spin Hall Effect (SHE) occurs in nonmagnetic systems with finite spin-orbit coupling, where an external electric field induces a transverse flow of pure spin current without any net charge current (see Figure 2.2). The spin current leads to the accumulation of spins on the edges of a sample. The SHE was first proposed by Dyakonov and Perel in 1971 [76, 77], but it gained significant attention after Hirsch's theoretical description, who coined the term "spin Hall effect" [78]. Despite the

absence of a Hall voltage in SHE, numerous experimental studies have successfully measured the effect [79–82].

The SHE and AHE originate from the same microscopic mechanisms, differing only in the requisite presence of spin polarization and time-reversal symmetry breaking in the examined material for AHE. Consequently, both AHE and SHE can be analyzed in a similar manner. Both effects can be deconstructed into two distinct contributions: the extrinsic and intrinsic contributions. The extrinsic contribution is rooted in spin-dependent scattering, such as scattering by impurities. Skew scattering, arising from asymmetric scattering, and side-jump scattering are the two dominant microscopic mechanisms responsible for spin-dependent scattering [83, 84], which will not be discussed further. The intrinsic SHE is seen in pure systems without impurity scattering and is connected to the Berry curvature [68, 85–87]. The intrinsic scattering mechanism depends only on the electronic structure of the material. When an electric field  $\mathbf{E}$  is applied (in  $\hat{\mathbf{x}}$  direction), electrons acquire a spin-dependent anomalous velocity (in  $\hat{\mathbf{y}}$  direction), leading to a transverse spin current. The intrinsic 2D SHE can be calculated using the Kubo formula:

$$\sigma^{\text{SH}} = \frac{e}{\hbar} \sum_n \int \frac{d^2 \mathbf{k}}{(2\pi)^2} f_{n\mathbf{k}} \Omega_{n\mathbf{k}}^{J_z}, \quad (2.26)$$

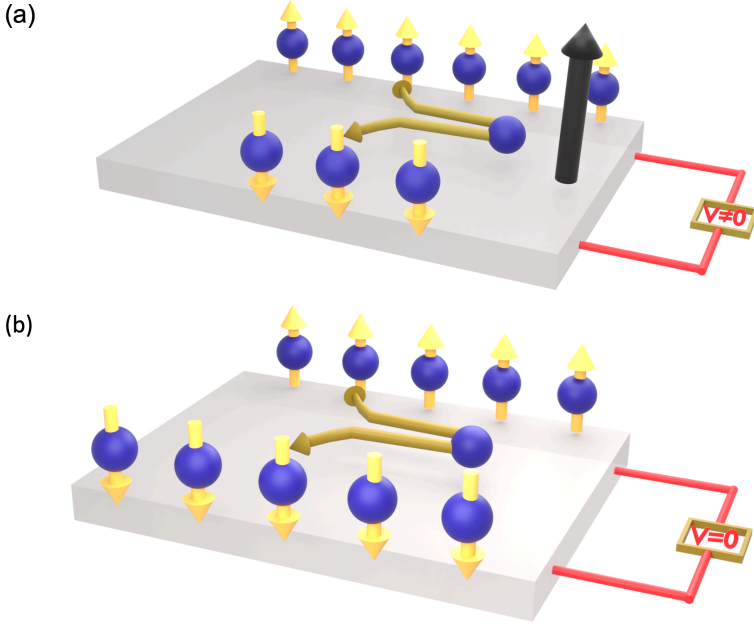
where  $f_{n\mathbf{k}}$  is the Fermi-Dirac distribution function, and  $\Omega_{n\mathbf{k}}^{J_z}$  is the spin Berry curvature (pointing in  $\hat{\mathbf{z}}$  direction), defined as:

$$\Omega_{n\mathbf{k}}^{J_z} = 2\hbar^2 \sum_{m \neq n} \text{Im} \left[ \frac{\langle u_{n\mathbf{k}} | j_y^{J_z} | u_{m\mathbf{k}} \rangle \langle u_{m\mathbf{k}} | v_x | u_{n\mathbf{k}} \rangle}{(E_{n\mathbf{k}} - E_{m\mathbf{k}} + i\eta)^2} \right]. \quad (2.27)$$

Here,  $j_y^{J_z}$  is the spin current operator, given by:

$$j_y^{J_z} = \frac{1}{2} (v_y J_z + J_z v_y),$$

where  $J_z = S_z$  is the  $z$ -component of the local angular momentum operator, and  $v_y$  is the velocity operator in the  $y$ -direction. The spin Berry curvature  $\Omega_{n\mathbf{k}}^{J_z}$  is associated with the  $n$ -th band, and the integral is performed over the Brillouin zone for all occupied bands below the Fermi surface. The SHC thus depends on the detailed spin character of the electronic bands and the geometry of the Berry curvature in momentum space.



**Fig. 2.2.:** (a) The Anomalous Hall Effect (AHE) is observed in magnetic materials characterized by an asymmetry in the distribution of spin-up and spin-down electrons. This asymmetry induces spin-dependent Hall transport, leading to an uneven charge accumulation at opposing boundaries and, consequently, the generation of a finite Hall voltage. Conversely, (b) the Spin Hall Effect (SHE) is analogous to the AHE but occurs in nonmagnetic materials. Despite the SHE causing spin accumulation at opposite boundaries, the quantities of spin-up and spin-down electrons are balanced, resulting in a zero Hall voltage.

## 2.6 Orbital Hall Effect

In addition to the Anomalous Hall Effect and Spin Hall Effect, the Orbital Hall Effect (OHE) describes the generation of a transverse flow of orbital angular momentum (OAM) in response to an applied electric field. Unlike the SHE, which relies on spin-orbit coupling to produce a transverse spin current, the OHE can emerge even in systems with weak or negligible SOC [42]. This makes the OHE a more general and fundamental phenomenon, driven by non-relativistic orbital textures in momentum space that are intrinsic to the electronic band structure of materials. As a result, the OHE is particularly prominent in multi-orbital systems, where the complex interplay of orbital degrees of freedom dominates the transport properties.

The OHE shares the same theoretical foundation as the AHE and SHE, relying on the Berry curvature of electronic bands. However, instead of spin, the key quantity in the OHE is the orbital angular momentum operator  $\mathbf{L}$ . The OHC, which quantifies the magnitude of the OHE, can be derived using the Kubo formula:

$$\sigma^{\text{OH}} = \frac{e}{\hbar} \sum_n \int \frac{d^2 \mathbf{k}}{(2\pi)^2} f_{n\mathbf{k}} \Omega_{n\mathbf{k}}^{J_z}, \quad (2.28)$$

where  $f_{n\mathbf{k}}$  is the Fermi-Dirac distribution function, and  $\Omega_{n\mathbf{k}}^{J_z}$  is the orbital Berry curvature, defined as:

$$\Omega_{n\mathbf{k}}^{J_z} = 2\hbar^2 \sum_{m \neq n} \text{Im} \left[ \frac{\langle u_{n\mathbf{k}} | j_y^{J_z} | u_{m\mathbf{k}} \rangle \langle u_{m\mathbf{k}} | v_x | u_{n\mathbf{k}} \rangle}{(E_{n\mathbf{k}} - E_{m\mathbf{k}} + i\eta)^2} \right]. \quad (2.29)$$

Here,  $j_y^{J_z}$  is the orbital current operator, given by:

$$j_y^{J_z} = \frac{1}{2} (v_y J_z + J_z v_y),$$

Here,  $J_z = L_z$  denotes the  $z$ -component of the orbital angular momentum operator, while  $v_y$  is the velocity operator along the  $y$ -direction. The orbital Berry curvature  $\Omega_{n\mathbf{k}}^{J_z}$  captures the interband coherence associated with orbital degrees of freedom in the  $n$ -th band. The Brillouin-zone integration is performed over all occupied states below the Fermi level. Consequently, the OHC is governed by the orbital texture of the Bloch states and the momentum-space distribution of the Berry curvature.

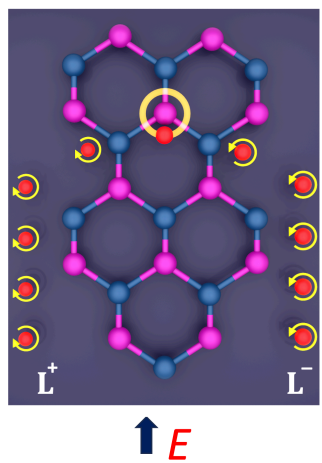
Moreover, in systems where sizable SOC exists, the OHE can be converted into the SHE, indicating that the OHE is more fundamental phenomenon. Theoretical studies have shown that in a variety of materials, the OHE can significantly surpass the SHE in magnitude, making it a more efficient mechanism for manipulating and detecting quantum states [42]. This discovery offers new opportunities in fields like spintronics and orbitronics, where controlling orbital angular momentum offers an alternative channel for information transfer.

The exploration of the OHE has been extended to a wide range of materials, including  $4d$  and  $5d$  transition metals, graphene, and two-dimensional transition-metal dichalcogenides (TMDs) [41, 42, 51, 88]. This research is particularly impactful because the OHE circumvents some of the material limitations posed by the SHE, which typically requires heavy elements with strong SOC. The ability of the OHE to arise in a wide class of materials, including systems with weak SOC, significantly expands its potential for device applications.



**Tab. 2.1.:** Comparison of anomalous, spin, and orbital Hall effects.

| Effect | Calculation Basis                                                                                  | Berry Curvature                                                            | SOC Dependence                                       | Example Materials                                                          |
|--------|----------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------|------------------------------------------------------|----------------------------------------------------------------------------|
| AHE    | Calculated using the intrinsic Kubo formalism and Berry curvature integration over occupied bands. | Requires finite Berry curvature from broken symmetries.                    | Strong SOC enhances the Hall response.               | Ferromagnets such as Nickel [74], Cobalt [89], and Iron [75].              |
| SHE    | Calculated from spin Berry curvature within linear-response theory.                                | Berry curvature generates transverse spin currents under electric fields.  | Finite SOC is required for efficient spin transport. | Platinum [90] and graphene with enhanced SOC [91, 92].                     |
| OHE    | Calculated using orbital Berry curvature and the Kubo formalism.                                   | Can exist with finite orbital Berry curvature even in nonmagnetic systems. | May occur without strong SOC.                        | Titanium [93] and TMDs such as MoS <sub>2</sub> and WSe <sub>2</sub> [94]. |



**Fig. 2.3.:** Orbital Hall Effect (OHE): When an electric field  $\mathbf{E}$  is applied, orbital angular momentum with a positive sign flows in one direction, while orbital angular momentum with a negative sign flows in the opposite direction, generating a transverse orbital current. Remarkably, the OHC can occur even in the absence of SOC.

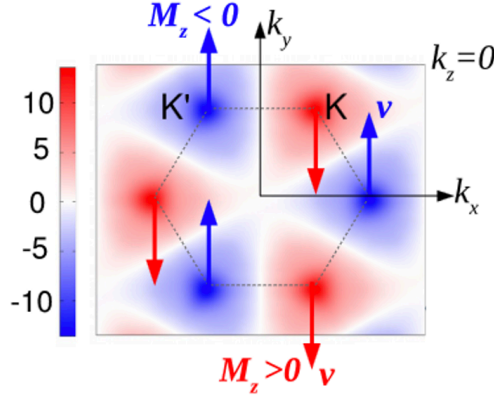
The OHE remained largely unknown until recently due to the quenching of OAM in solids in the presence of crystal fields. However, recent studies have shown that orbital quenching does not prevent the nonequilibrium dynamics and transport of the orbital degree of freedom [42, 45]. Additionally, it facilitates the essential hybridization between different atomic orbitals, playing a vital role in orbital dynamics [42].

This implies that external stimuli, such as an electric field, can steer the dynamics and transport of orbital information, irrespective of the orbital quenching that dominates ground state properties [95]. In 2023, the OHE-induced orbital moment in face-centered cubic titanium was detected experimentally using the magneto-optical Kerr effect (MOKE). The OHE-induced MOKE signal was significantly stronger than the SHE-induced spin moment. This discovery is notable, especially considering that titanium is a light element with negligible SOC [93].

The intrinsic mechanism of the OHE exhibits a striking contrast between centrosymmetric and non-centrosymmetric materials. In centrosymmetric systems, the presence of inversion symmetry enforces a ground state with zero orbital angular momentum (OAM). This arises because the OAM expectation values satisfy  $\langle \mathbf{L} \rangle_{n,-\mathbf{k}} = \langle \mathbf{L} \rangle_{n,\mathbf{k}}$ . Furthermore, time-reversal symmetry imposes an additional constraint:  $\langle \mathbf{L} \rangle_{n,-\mathbf{k}} = -\langle \mathbf{L} \rangle_{n,\mathbf{k}}$ . When both symmetries are present, these constraints force the OAM to vanish at every  $\mathbf{k}$ -point in the Brillouin zone. To induce nonzero OAM in such materials, external perturbations such as an applied electric field  $\mathbf{E}$  are required to break inversion symmetry and hybridize states with distinct orbital characters [42, 95]. In contrast, non-centrosymmetric materials inherently lack inversion symmetry, allowing for the existence of nonzero OAM in the ground state. However, even in these materials, an external electric field  $\mathbf{E}$  is still necessary to drive the OHE. The absence of inversion symmetry facilitates the generation of OAM, but the electric field is essential to create the transverse flow of orbital angular momentum. Thus, while non-centrosymmetric materials provide a natural platform for observing and harnessing the OHE, the application of an electric field remains a key requirement for its manifestation.

### 2.6.1 Orbital Hall Effect in Transition Metal Dichalcogenides

TMDs characterized by their non-centrosymmetric nature with time-reversal symmetry, can manifest a substantial OHC with a mechanism notably distinct from that observed in centrosymmetric cases. The absence of inversion symmetry in non-centrosymmetric systems enables the spatial accumulation of orbital moments as shown in Fig. 2.4. In contrast to centrosymmetric materials, an intrinsic  $k$ -space orbital moment exists even in the absence of an external electric field. These orbital moments in  $\mathbf{k}$ -space exhibit a transverse flow in response to the applied electric field  $E$ , giving rise to the Hall effect due to the anomalous velocity  $\mathbf{v} \propto \mathbf{E} \times \boldsymbol{\Omega}$ , stemming from the nonzero Berry curvature present in the system [54]. Subsequently, this orbital moment flow results in a pronounced OHC.



**Fig. 2.4.:** In a monolayer of TMD, the broken spatial inversion symmetry results in a valley-dependent orbital angular momentum (OAM) in equilibrium (highlighted by red/blue color). Consequently, the anomalous velocity (illustrated by red/blue arrows), propelled by the valley-dependent Berry curvature, induces a Hall current that is inherently orbitally polarized. Adapted from ref [96].

## 2.7 Spin-Orbit Torque

Switching the magnetization of magnetic moments is a primary focus in developing next-generation data storage devices. Various techniques, including magnetic fields, spin-transfer torque (STT) [97], and spin-orbit torques (SOT) [37, 98], have been explored for efficient and energy-effective magnetization switching. STT involves using a current of spin-polarized electrons to change the magnetization orientation by applying torque, without the need for external magnetic fields. However, STT relies on the exchange of spin angular momentum between two ferromagnetic layers and requires spatial variation in magnetization, leading to significant energy dissipation and device complexity.

In contrast, SOT arises from spin-orbit coupling, where an electric field transfers orbital angular momentum to spin angular momentum within either a single material or a heterostructure. Conventionally, an electric field creates nonequilibrium orbital occupation, which leads to SOT through SOC. This process encompasses mechanisms such as the spin Hall effect [99], the Rashba-Edelstein effect [4, 100], and the orbital Hall effect [101]. The result is nonequilibrium spin accumulation that generates magnetic torque on the magnetization. The advantage of SOT over STT is that it can operate in collinear magnetic systems when inversion symmetry is broken. Unlike STT, SOT does not require a second ferromagnetic layer, offering greater flexibility in device design and significantly lower energy dissipation.

### 2.7.1 Magnetization Dynamics

The Landau-Lifshitz-Gilbert (LLG) equation [102] describes how the magnetization  $\hat{\mathbf{M}}$  responds to the effective magnetic fields within the material:

$$\frac{d\hat{\mathbf{M}}}{dt} = -\gamma\hat{\mathbf{M}} \times \mathbf{H}_{\text{eff}} + \alpha\hat{\mathbf{M}} \times \frac{d\hat{\mathbf{M}}}{dt}, \quad (2.30)$$

where the first term represents the precession of the magnetization vector  $\hat{\mathbf{M}}$  around the direction of the effective magnetic field  $\mathbf{H}_{\text{eff}}$ , and the second term accounts for the damping of the magnetization dynamics. Here,  $\gamma = \frac{ge}{2m_e}$  is the gyromagnetic ratio, and  $\alpha$  is the Gilbert damping parameter. Figure 2.5 provides a schematic representation of the magnetization dynamics governed by the LLG equation.

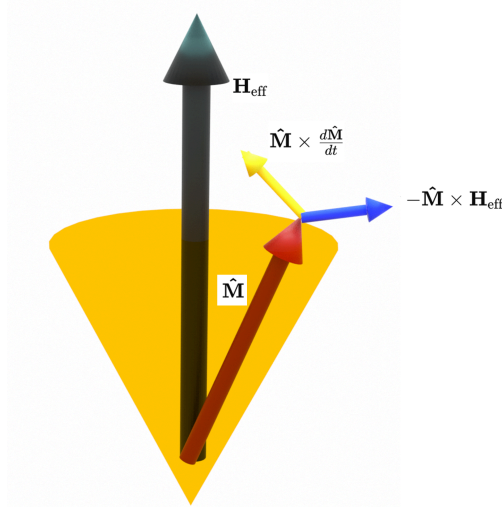
When spin-polarized electrons are injected into a ferromagnetic layer, the magnetization of the layer is influenced by a torque arising from the interaction with the polarized spins. In 1996, Slonczewski revised the LLG equation to incorporate this torque generated by the current. This updated equation, referred to as the Landau-Lifshitz-Gilbert-Slonczewski (LLGS) equation, is formulated as follows:

$$\frac{d\hat{\mathbf{M}}}{dt} = -\gamma\hat{\mathbf{M}} \times \mathbf{H}_{\text{eff}} + \alpha\hat{\mathbf{M}} \times \frac{d\hat{\mathbf{M}}}{dt} + T_{\text{FL}}\boldsymbol{\sigma} \times \hat{\mathbf{M}} + T_{\text{DL}}\hat{\mathbf{M}} \times (\boldsymbol{\sigma} \times \hat{\mathbf{M}}), \quad (2.31)$$

where the third term on the right-hand side corresponds to the current-induced field-like (FL) torque, while the fourth term corresponds to the damping-like (DL) torque.  $\boldsymbol{\sigma}$  is the spin polarization orientation.

## 2.8 Spin and Orbital Torque

Injection of an orbital current into a ferromagnetic material can induce an orbital torque (OT) on the local magnetic moments, similar to how the injection of a spin current into a ferromagnet induces magnetization dynamics, which is known as spin torque as described by SOT [103]. One key difference between spin torque and orbital torque is that orbital torque requires SOC because the local moments in a ferromagnet are coupled to spin by exchange interaction, and OAM interacts with magnetization indirectly via SOC [43]. The mechanism of orbital torque, as detailed by Go in [43], involves three main steps: the generation of OAM or the orbital current in the nonmagnetic (NM) layer, the injection of OAM into the ferromagnetic (FM) layer, and the conversion of OAM to spin. This spin then interacts with the local magnetic moment through SOC, ultimately resulting in torque on the magnetization, as shown in Fig. 2.6. The advantage of relying on an



**Fig. 2.5.:** Magnetization dynamics described by the Landau-Lifshitz-Gilbert (LLG) equations. The red arrow ( $\mathbf{M}$ ) represents the magnetization of a ferromagnet, while the black arrow illustrates the effective magnetic field ( $H_{eff}$ ). The blue and yellow arrows correspond to the damping torque and field-like torque, respectively, which are perpendicular to each other.

orbital torque is that the OHC is often one order of magnitude larger than SHC in various materials and does not rely on SOC to occur. However, to exert an OT on a ferromagnet, one still needs SOC to convert the orbital current into a spin current. Unlike the conventional SOT device which needs high SOC in the NM layer, in an OT device one requires only moderate SOC in the ferromagnet and vanishing SOC in the NM layer. It has been shown experimentally that the torque efficiency was greatly increased with the inclusion of orbital currents in a SOT device [104, 105], positioning OT as a promising path towards achieving enhanced torque efficiency in spintronic devices.

### 2.8.1 Anatomy of Spin Orbit Torque

To investigate the spin and orbital torque that is exerted on the magnetization upon application of an electric field, we utilize the spin continuity equation for the time evolution of spin density [106]:

$$\frac{\partial \mathbf{S}}{\partial t} = \Phi^{\mathbf{S}} + \mathbf{T}_{\text{SO}}^{\mathbf{S}} + \mathbf{T}_{\text{XC}}^{\mathbf{S}}, \quad (2.32)$$

where  $\Phi^{\mathbf{S}}$  stands for the spin flux contribution,  $T_{\text{SO}}$  is what we refer to as the spin-orbital, or, simply orbital torque, and  $T_{\text{XC}}$  is the torque on the magnetization due to transfer of angular momentum from the spin of excited electronic states.

For the total Hamiltonian  $H$ , the spin flux coming into an atom defined by the projection  $P$  is given by

$$\Phi^{\mathbf{S}} = \frac{1}{2i\hbar} \{[P, H], \mathbf{S}\}, \quad (2.33)$$

where  $[A, B] = AB - BA$  and  $\{A, B\} = AB + BA$  are the commutator and anti-commutator for the pair of operators  $A$  and  $B$ . Here,  $P$  acts as a projection onto a specific atomic site, allowing us to isolate the spin flux contribution for that site.

The spin-orbital torque and exchange torque are given as

$$T_{\text{SO}}^{\mathbf{S}} = \frac{1}{2i\hbar} \{[\mathbf{S}, H_{\text{SO}}], P\}, \quad T_{\text{XC}}^{\mathbf{S}} = \frac{1}{2i\hbar} \{[\mathbf{S}, H_{\text{XC}}], P\}, \quad (2.34)$$

respectively, where  $H_{\text{SO}}$  and  $H_{\text{XC}}$  stand for the spin-orbit and spin-dependent exchange-correlation part of the single-particle Kohn-Sham Hamiltonian, respectively. That is, the total Hamiltonian as given by (for further details, see next Chapter 3 on Density Functional Theory):

$$H = H_0 + H_{\text{SO}} + H_{\text{XC}}, \quad (2.35)$$

where  $H_0$  incorporates the spin-independent part of the Hamiltonian, including the kinetic energy and the crystal-field potential generated by the lattice and the mean-field of electron cloud. Note that the exchange Hamiltonian is given as  $H_{\text{XC}} = (2\mu_{\text{B}}/\hbar)\mathbf{S} \cdot \mathbf{\Omega}_{\text{XC}}$ , where  $\mathbf{\Omega}_{\text{XC}} = \hat{\mathbf{M}} \cdot \mathbf{\Omega}_{\text{XC}}(\mathbf{r})$  is the so-called exchange field which determines the magnitude of the magnetic moments and spatial properties of the spin density, and  $\hat{\mathbf{M}}$  is the unit vector along the magnetization direction. In the evaluation of the corresponding torques, only muffin-tin regions around atoms are considered, as discussed in Chapter 3.

In the steady state, the torque on the magnetization is given by

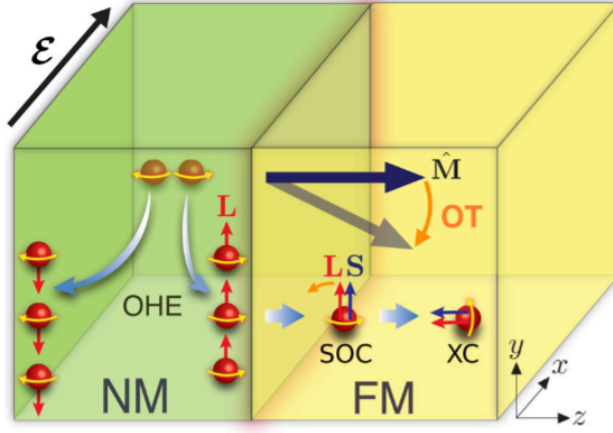
$$\langle -T_{\text{XC}}^{\mathbf{S}} \rangle \approx \langle \Phi^{\mathbf{S}} \rangle + \langle T_{\text{SO}}^{\mathbf{S}} \rangle, \quad (2.36)$$

where  $\langle \cdots \rangle$  stands for the statistical average in the steady state. However, in a 2D magnet, which is periodic in the  $xy$  plane, the flux is absent by construction. Thus, the exchange torque is purely given by the spin-orbital torque, the angular momentum transfer from the OAM to the spin,  $\langle -T_{\text{XC}}^{\mathbf{S}} \rangle \approx \langle T_{\text{SO}}^{\mathbf{S}} \rangle$ . Under an external

electric field  $E_x$  applied along  $x$ , the response of the torque in the steady state as given by the Kubo formula:

$$\langle T_{XC}^S \rangle = \frac{e\hbar E_x}{N_k} \sum_{nm} \sum_{\mathbf{k}} (f_{n\mathbf{k}} - f_{m\mathbf{k}}) \frac{\text{Im} [\langle \psi_{n\mathbf{k}} | T_{XC}^S | \psi_{m\mathbf{k}} \rangle \langle \psi_{m\mathbf{k}} | v_x | \psi_{n\mathbf{k}} \rangle]}{(E_{n\mathbf{k}} - E_{m\mathbf{k}} + i\eta)^2} \equiv t_{yx} E_x, \quad (2.37)$$

where  $e > 0$  is the unit charge,  $N_k$  is the number of  $k$ -points used for the summation such that the response is given in the unit per unit cell,  $v_x$  is the  $x$ -component of the velocity operator,  $\psi_{n\mathbf{k}}$  is an eigenstate of the total Hamiltonian with band index  $n$ ,  $E_{n\mathbf{k}}$  is its energy eigenvalue, and  $t_{yx}$  is the  $yx$ -component of the torkance tensor. Here,  $f_{n\mathbf{k}}$  is the corresponding Fermi-Dirac distribution function. The calculation for the spin-orbital torque is done by replacing  $T_{XC}^S$  by  $T_{SO}^S$  in Eq. (2.37).



**Fig. 2.6.:** The orbital torque (OT) mechanism in a nonmagnetic/ferromagnetic (NM/FM) bilayer is illustrated as follows: It begins with the injection of orbital angular momentum (OAM) in the NM layer, where torque arises purely from the orbital contribution. Since the NM is composed of light elements, it exhibits negligible SOC. The injected OAM is then converted into spin through the SOC present in the FM layer. This conversion ultimately generates torque on the local magnetic moment  $M$  under the influence of an applied electric field. The figure is adapted from [43].

## 2.8.2 Orbital and Spin Pumping

The reciprocal phenomenon of spin torque, termed spin pumping, occurs when magnetization produces non-equilibrium spin. This mechanism is deeply rooted in the fundamental dynamics of magnetization, characterized by the transfer of

angular momentum. Notably, this transfer extends beyond spin to include orbital angular momentum (OAM). Similarly, orbital pumping refers to the process where magnetization dynamics drives an orbital current, causing the flow of OAM [44, 107–109]. This process is the orbital analogue of spin pumping and extends the concept of angular momentum transfer to include orbital contributions. Orbital pumping has been recognized as one of the most flexible and effective techniques for generating spin currents [44]. Recent studies have demonstrated that orbital pumping can significantly advance spintronics, enabling more efficient spin-current generation and providing new routes to control magnetization dynamics through the interplay of spin and orbital angular momentum [44]. In particular, orbital pumping can drive the conversion of orbital currents into spin currents via spinorbit coupling [110].

To explore the generation of spin and orbital currents driven by magnetization dynamics, we need to develop a model that accounts for the effects of these dynamic processes. A key assumption is that the exchange field becomes time-dependent due to the time-dependent nature of the magnetization direction. This approach enables us to systematically investigate how time-varying magnetization influences the generation of spin and orbital currents:

$$\mathbf{V}_{\text{XC}}(t) = \hat{\mathbf{M}}(t) \cdot \mathbf{V}_{\text{XC}}(\mathbf{r}). \quad (2.38)$$

It can be shown [111] that this results in the following rate of change of the Hamiltonian of the system:

$$\frac{dH}{dt} = \mathcal{T}_{\text{XC}} \cdot \left( \hat{\mathbf{M}}(t) \times \frac{d\hat{\mathbf{M}}(t)}{dt} \right). \quad (2.39)$$

In terms of the torkance tensor  $t_{\alpha\beta}$ , by using perturbation theory, one can arrive at the expression for the so-called inverse spin-orbit torque effect, *i.e.* an effect of charge pumping by magnetization dynamics:

$$j_{\alpha}(t) = \frac{1}{V} \sum_{\beta} t_{\beta\alpha} \left( \hat{\mathbf{M}}(t) \right) \left( \hat{\mathbf{M}}(t) \times \frac{d\hat{\mathbf{M}}(t)}{dt} \right)_{\beta}, \quad (2.40)$$

where  $V$  is the volume and  $j_{\alpha}$  is the  $\alpha$ 'th component of the generated by magnetization dynamics charge current density [111]. Thus, following Onsager's reciprocity principle, the torkance tensor describes both the response of torque to the current, and the response of current to the magnetization dynamics.



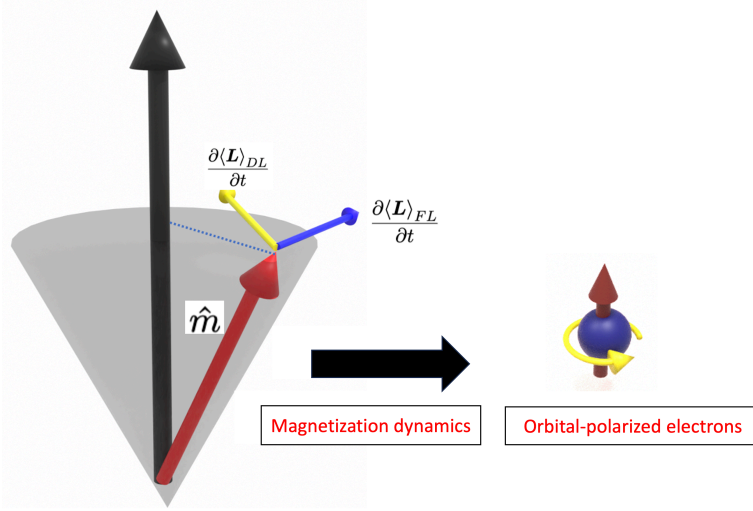
To assess the effects of orbital and spin pumping, we introduce the inverse spin and orbital torque tensors  $t_{\beta\alpha}^{J_\gamma}$  according to

$$Q_\alpha^{J_\gamma}(t) = \frac{1}{V} \sum_\beta t_{\beta\alpha}^{J_\gamma} \left( \hat{\mathbf{M}}(t) \right) \left( \hat{\mathbf{M}}(t) \times \frac{d\hat{\mathbf{M}}(t)}{dt} \right)_\beta, \quad (2.41)$$

where  $J_\gamma$  stands for the  $\gamma$ -component of either spin ( $S_\gamma$ ) or orbital ( $L_\gamma$ ) angular momentum, and  $Q_\alpha^{J_\gamma}$  is the current density of  $J_\gamma$  along direction  $\alpha$ . Within the Kubo linear response the intrinsic contribution to the inverse spin (orbital) torque tensor reads:

$$\frac{t_{\alpha\beta}^{J_\gamma}}{e\hbar} = 2 \sum_{n\mathbf{k} < E_F} \text{Im} \left[ \frac{\langle u_{n\mathbf{k}} | j_\alpha^{J_\gamma} | u_{m\mathbf{k}} \rangle \langle u_{m\mathbf{k}} | T_{XC,\beta}^S | u_{n\mathbf{k}} \rangle}{N_{\mathbf{k}}(E_{n\mathbf{k}} - E_{m\mathbf{k}} + i\eta)^2} \right], \quad (2.42)$$

where  $j_\alpha^{J_\gamma} = \frac{1}{2}(v_\alpha J^\gamma + J^\gamma v_\alpha)$  is the operator of spin (orbital) velocity.



**Fig. 2.7.:** The schematic depicts the concept of orbital pumping, where magnetization dynamics drive the generation of an orbital-polarized current. As the magnetization precesses or undergoes dynamic changes, it induces a flow of orbital angular momentum (OAM) within the system.

## 2.9 Orbital to Orbital-Current Conversion

To explore the concept of orbital-to-orbital current conversion, we consider the response of the system in terms of a pumped orbital angular momentum (OAM)

current when the orbital magnetization increases over time. This effect can be modeled by introducing an additional orbital term in the Hamiltonian,  $B_L(t)\hat{\mathbf{L}}$ , where the orbital exchange field,  $B_L(t)$ , increases linearly with time,  $B_L(t) = \eta t$ . Such a term, introduced for spin systems, has previously been employed to investigate phenomena such as the inverse (spin) Edelstein effect [112] and its magnetic counterpart [113], both associated with the pumping of charge currents.

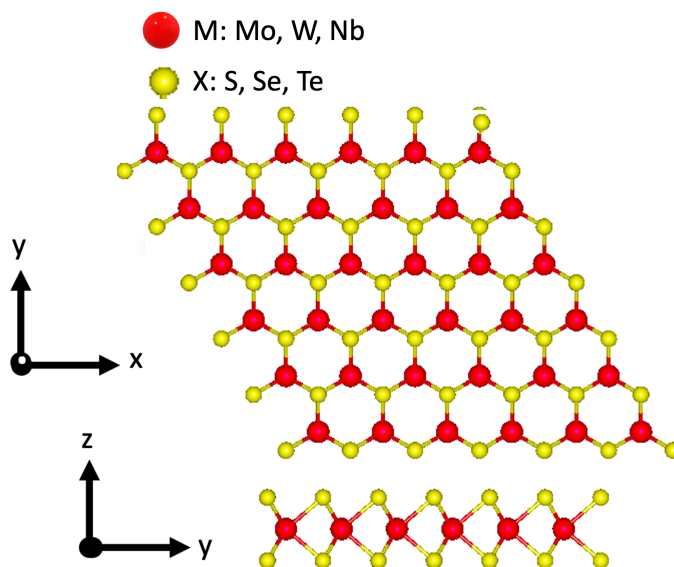
In this context, we treat the time-dependent orbital exchange field as a perturbation and focus on the resulting orbital current response. This response, proportional to the rate of change in the orbital magnetic field  $\eta$ , is characterized by the orbital-to-orbital-current conversion tensor, which governs the relationship between the orbital dynamics and the generated orbital currents.

$$\frac{\tau_{\alpha}^{\beta\gamma}}{e\hbar} = 2 \sum_{nk < E_F}^{mk > E_F} \text{Im} \left[ \frac{\langle u_{nk} | j_{\beta}^{L\gamma} | u_{mk} \rangle \langle u_{mk} | \hat{L}_{\alpha} | u_{nk} \rangle}{N_{\mathbf{k}}(E_{nk} - E_{mk} + i\eta)^2} \right]. \quad (2.43)$$

The calculations for the  $\tau_x^{xz}$  and  $\tau_x^{xy}$  components corresponding to a generation of  $y$ - or  $z$ -polarized OAM currents along  $x$  in response to the orbital field applied along  $x$ .

## 2.10 Transition Metal Dichalcogenides

Two-dimensional (2D) transition metal dichalcogenides (TMDs), denoted as  $\text{MX}_2$  where M represents a transition metal element (such as Mo, W, or Nb) and X is a chalcogen atom (such as S, Se, or Te), as shown in Fig. 2.8, constitute a diverse class of van der Waals (vdW) crystals. These materials can be mechanically exfoliated from their bulk form into individual monolayers. Monolayers of TMDs exhibit unique properties that differ significantly from their bulk counterparts. For example, while many bulk TMDs possess an indirect bandgap, their monolayer forms often transition to a direct bandgap, as demonstrated in studies such as [114–116].  $\text{MX}_2$  monolayers can exist in two primary structural phases: the hexagonal (H-phase  $\text{MX}_2$ ) and the trigonal (T-phase  $\text{MX}_2$ ). In the H-phase  $\text{MX}_2$  structure, each M atom is positioned at the center of a trigonal prism formed by six surrounding X atoms, resulting in hexagonal symmetry. This structure is the focus of this thesis. In contrast, the T-phase  $\text{MX}_2$  structure features M atoms octahedrally coordinated with six X atoms, leading to trigonal symmetry. These structural distinctions give rise to significantly different electronic properties: the H-phase is typically semiconducting, whereas the T-phase is metallic, as highlighted in [117].



**Fig. 2.8.:** Top and side view of the H-phase monolayer MX<sub>2</sub>. The red balls and yellow balls represent M and X atoms, respectively.

Magnetism is an essential characteristic within the realm of low-dimensional spintronic devices. However, exhibiting magnetism in 2D systems is challenging due to the well-known Mermin-Wagner theorem, which states that thermal fluctuations can prevent spontaneous magnetization in low-dimensional systems [118]. Introducing magnetic anisotropy can stabilize magnetism in 2D materials by overcoming these fluctuations and maintaining magnetic order [119]. Consequently, considerable progress has been made in investigating the magnetism of 2D materials. Since the groundbreaking discovery of ferromagnetism in monolayer CrI<sub>3</sub> [120], a plethora of 2D magnetic materials has been discovered, including monolayer Fe<sub>3</sub>GeTe<sub>2</sub> [121] and monolayer FePS<sub>3</sub> [122]. Additionally, 2D TMDs such as VSe<sub>2</sub> have attracted substantial interest due to their fascinating magnetic properties. However, there is an ongoing debate regarding whether VSe<sub>2</sub> exhibits ferromagnetic behavior [123] or if its magnetism is frustrated [124]. Further research into the magnetism of 2D TMDs is expected to reveal new phenomena and enhance our comprehension of magnetic interactions at the nanoscale.

|    |    |       |    |    |    |    |    |    |    |    |    |     |    |     |    |     |     |  |
|----|----|-------|----|----|----|----|----|----|----|----|----|-----|----|-----|----|-----|-----|--|
| H  |    |       |    |    |    |    |    |    |    |    |    |     |    |     |    |     | He  |  |
| Li | Be |       |    |    |    |    |    |    |    |    |    | B   | C  | N   | O  | F   | Ne  |  |
| Na | Mg |       |    |    |    |    |    |    |    |    |    | Al  | Si | P   | S  | Cl  | Ar  |  |
| K  | Ca | Sc    | Ti | V  | Cr | Mn | Fe | Co | Ni | Cu | Zn | Ga  | Ge | As  | Se | Br  | Kr  |  |
| Rb | Sr | Y     | Zr | Nb | Mo | Tc | Ru | Rh | Pd | Ag | Cd | In  | Sn | Sb  | Te | I   | Xe  |  |
| Cs | Ba | La-Lu | Hf | Ta | W  | Re | Os | Ir | Pt | Au | Hg | Tl  | Pb | Bi  | Po | At  | Rn  |  |
| Fr | Ra | Ac-Lr | Rf | Db | Bh | Hs | Ms | Mt | Ds | Rg | Cn | Uut | Fl | Uup | Lv | Uus | Uuo |  |
|    |    |       | 57 | 58 | 59 | 60 | 61 | 62 | 63 | 64 | 65 | 66  | 67 | 68  | 69 | 70  |     |  |
|    |    |       | La | Ce | Pr | Nd | Pm | Sm | Eu | Gd | Tb | Dy  | Ho | Er  | Tm | Yb  |     |  |

**Fig. 2.9.:** Highlighted in the periodic table are Europium (Eu) and Gadolinium (Gd), representing rare-earth elements, with their electron configurations being  $[\text{Xe}] 4f^7 6s^2$  for Eu and  $[\text{Xe}] 4f^7 5d^1 6s^2$  for Gd. These elements are significant for their partially filled  $4f$  orbitals, which contribute to unique magnetic and electronic properties. Sulfur (S), Selenium (Se), and Tellurium (Te) are also highlighted as representatives of dichalcogenides, showcasing their interactions with rare-earth elements.

### 2.10.1 Rare-Earth Dichalcogenides

In this thesis, we proceeded to scan promising materials capable of hosting complex magnetic properties. For this purpose, we focus on  $4f$  rare-earth elements. In particular, we adopt the well-established crystal structure of transition metal dichalcogenides (TMDs) and replace the transition-metal atoms with rare-earth elements, leading to the formation of rare-earth dichalcogenides (REDs). The utilization of localized  $4f$  electrons in these 2D materials presents distinct advantages for advanced magnetic storage devices and spintronics applications. The  $4f$  electrons give rise to large magnetic moments and strong magnetocrystalline anisotropy. Combined with strong spinorbit coupling and the unique crystal field in a two-dimensional geometry [26–29], these features significantly enhance the magnetic performance of the materials. This robust magnetic behavior is difficult to achieve in conventional TMDs, where magnetism is typically more fragile, making REDs particularly attractive for next-generation technologies.



# Density Functional Theory

# 3

The dawn of the 20th century was marked by the emergence of quantum mechanics, which sparked a transformative era in the realm of physics [125, 126]. This revolutionary theory reshaped our understanding of the microscopic world, offering a novel perspective on the behavior of matter. One of its most profound implications was the ability to predict the properties of atomic systems with unparalleled accuracy.

In this chapter, I offer a brief overview of the electronic structure problem, and its resolution through quantum mechanics, specifically using density functional theory (DFT). We begin by explaining the interacting many-body Schrödinger equation and the theoretical foundation of DFT. We then explore various extensions to DFT that allow to model the many-electron problem that we encounter in different materials systems more accurately. This introductory section follows along the lines of similar introductions to DFT [127–133].

## 3.1 The Many-Body Schrödinger Equation

The time-independent Schrödinger equation accurately describes the ground state of a system comprising electrons and atomic nuclei:

$$\mathcal{H}\psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_\alpha, \mathbf{R}_1, \mathbf{R}_2, \dots, \mathbf{R}_\beta) = E\psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_\alpha, \mathbf{R}_1, \mathbf{R}_2, \dots, \mathbf{R}_\beta). \quad (3.1)$$

Here,  $\Psi$  represents the many-body wave function that depends on the coordinates of  $\mathbf{r}_i$  of all  $\alpha$  electrons as well as on the positions of  $\mathbf{R}_\mu$  of the  $\beta$  nuclei, and the Hamiltonian operator  $\mathcal{H}$  is defined as:

$$\mathcal{H} = \underbrace{-\frac{\hbar^2}{2m_e} \sum_i^\alpha \nabla_i^2}_{\mathcal{T}_e} - \underbrace{\frac{\hbar^2}{2} \sum_\mu^\beta \frac{1}{M_\mu} \nabla_\mu^2}_{\mathcal{T}_n} - \underbrace{\sum_{i,\mu} \frac{e^2 Z_\mu}{|\mathbf{r}_i - \mathbf{R}_\mu|}}_{\mathcal{V}_{e-n}} + \underbrace{\frac{1}{2} \sum_{i \neq j} \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|}}_{\mathcal{V}_{e-e}} + \underbrace{\frac{1}{2} \sum_{\mu \neq \nu} \frac{e^2 Z_\mu Z_\nu}{|\mathbf{R}_\mu - \mathbf{R}_\nu|}}_{\mathcal{V}_{n-n}}. \quad (3.2)$$

The first two terms in the Hamiltonian, represented by  $\mathcal{T}_e$  and  $\mathcal{T}_n$ , describe the kinetic energy of the electrons and nuclei, respectively, the following three terms

represent the potential energies. In detail, the third term,  $\mathcal{V}_{e-n}$ , represents the attractive Coulomb interaction between an electron located at  $\mathbf{r}_i$  and a nucleus positioned at  $\mathbf{R}_\mu$ . The remaining two terms,  $\mathcal{V}_{e-e}$  and  $\mathcal{V}_{n-n}$ , describe the Coulomb repulsion between electrons and between nuclei, respectively. Here,  $i$  and  $j$  represent the electrons, while  $\mu$  and  $\nu$  are indices for the nuclei. The quantities  $\alpha$  and  $\beta$  represent the total number of electrons and nuclei, respectively, while  $Z_\mu$  and  $M_\mu$  denote the atomic number and mass of the nuclei  $\mu$ .

## 3.2 Born-Oppenheimer Approximation

With nuclei being at least a thousand times heavier than electrons, their motion is significantly slower, making them appear almost stationary compared to the rapidly moving electrons. This difference arises from the equal forces acting on both electrons and nuclei; however, the much smaller mass of electrons leads to greater accelerations. Consequently, it is assumed that the nuclei remain stationary. This assumption represents a key simplification of the many-body problem and is referred to as the adiabatic *Born-Oppenheimer approximation* [134]. In systems featuring stationary nuclei, the kinetic energy ( $\mathcal{T}_n$ ), associated with nuclei can be neglected, as a result, the electrostatic potential energy from the nucleus can be treated as an external potential energy, similar to other energies due to externally applied fields, enabling the Hamiltonian to be expressed as:

$$\mathcal{H}_{BO} = \mathcal{T}_e + \mathcal{V}_{ee} + \mathcal{V}_{\text{ext}}, \quad (3.3)$$

where

$$\mathcal{V}_{\text{ext}} = -\sum_{i,\mu} \frac{e^2 Z_\mu}{|\mathbf{r}_i - \mathbf{R}_\mu|} + \mathcal{V}_{n-n}. \quad (3.4)$$

$\mathcal{V}_{\text{ext}}$  is the external potential which includes the nuclear electrostatic potential. While treating atomic nuclei as stationary objects significantly simplifies the problem, solving Eq. 3.3 remains computationally challenging. The solution is a  $3N_\alpha$ -dimensional function, represented as  $|\psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_\alpha)\rangle$ . Due to the mutual interactions among the electrons, the system depends on the coordinates of all electrons and cannot be decoupled into a system of non-interacting particles. Consequently, obtaining an exact solution is practically unattainable. Even for moderately sized systems with relatively small values of  $N_\alpha$ , storing or computing this solution becomes infeasible.

### 3.3 Density Functional Theory

The fundamental idea behind DFT is to utilize the electronic charge density  $n(\mathbf{r})$  rather than the wave function to determine all the observables of a given system.

#### 3.3.1 Hohenberg-Kohn Theorem

The cornerstone of density functional theory (DFT) is the replacement of the ground-state many-body wavefunction as the central descriptor with the ground-state electron density of the electrons,  $n(\mathbf{r})$ . This was made possible thanks to theorems by Hohenberg and Kohn [135]. Unlike the many-body wavefunction,  $\psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_\alpha)$ , which depends on the coordinates of all electrons in the system, the charge density depends only on one spatial variable, making it significantly simpler to calculate, as shown schematically in Fig. 3.1.

For a system of  $N$  interacting electrons described by the Hamiltonian in Eq. 3.3, the ground-state electron density is expressed as:

$$n_{gs}(\mathbf{r}) = \int |\psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_\alpha)|^2 d\mathbf{r}_2 d\mathbf{r}_3 \dots d\mathbf{r}_\alpha. \quad (3.5)$$

The first Hohenberg-Kohn theorem states [135]:

1. **Theorem 1:** *The external potential  $V_{ext}$  in Eq. 3.3 is determined by the electronic many-body density  $n(\mathbf{r})$  up to a trivial constant. As a consequence, the total energy of the system is a unique functional of the density:*

$$E = E[n(\mathbf{r})] = \mathcal{T}[n(\mathbf{r})] + \mathcal{V}[n(\mathbf{r})] + \mathcal{V}_{e-e}[n(\mathbf{r})]. \quad (3.6)$$

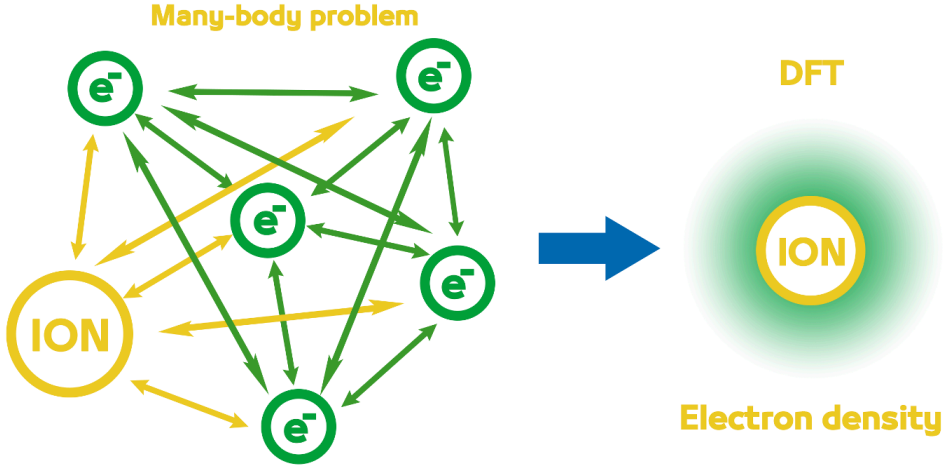
Thus, the total energy and all observables are given as functional of the ground-state density.

2. **Theorem 2:** *The total energy functional  $E[n(\mathbf{r})]$  is minimized for the exact ground-state density  $n(\mathbf{r})$  under the constraint that the total number of electrons remains fixed. This implies that:*

$$E[n_{gs}(\mathbf{r})] \leq E[n(\mathbf{r})] \quad (3.7)$$

Hence, minimizing the total energy functional determines the ground-state density. Furthermore, this demonstrates that the variational principle can be applied to calculate the ground-state density.





**Fig. 3.1.:** The core principle of Density Functional Theory (DFT) is to reduce the complexity of many-body electron interactions by describing the system in terms of its electron density distribution. Instead of tracking individual electrons, DFT treats each electron as moving within an effective potential generated by the combined effects of all other electrons and the nuclei.

### 3.3.2 Kohn-Sham System

The Hohenberg-Kohn theorems establish that the ground-state density can be used to compute any observable. This density is determined by minimizing the total energy functional, with all physical properties of the ground state expressed as functionals of  $n_{gs}(\mathbf{r})$ . In 1964, Kohn and Sham introduced single-particle Schrödinger-like equations to describe a system of  $N$  non-interacting electrons, instead of tackling the fully interacting many-body problem [136]. In this framework, all properties are derived from single-particle wavefunctions. They proposed that a fictitious non-interacting system could be constructed such that its ground-state density matches that of the interacting many-body system. To address this non-interacting problem, one must solve the Kohn-Sham equation:

$$\left[-\nabla^2 + V_{\text{eff}}(\mathbf{r})\right] \psi_i(\mathbf{r}) = \mathcal{E}_i \psi_i(\mathbf{r}), \quad (3.8)$$

where  $\psi_i(\mathbf{r})$  represents the Kohn-Sham orbital, and  $\mathcal{E}_i$  denotes the Kohn-Sham orbital's eigenvalues. The effective Kohn-Sham potential  $V_{\text{eff}}(\mathbf{r})$  is an unknown functional of the ground-state electron density and can be decomposed into three components:

$$V_{\text{eff}}(\mathbf{r}) = V_{\text{ext}}(\mathbf{r}) + V_{\text{H}}(\mathbf{r}) + V_{\text{xc}}(\mathbf{r}), \quad (3.9)$$

where the electron density can be calculated using the Kohn-Sham orbitals as:

$$n_{\text{KS}}(\mathbf{r}) = \sum_i^\alpha |\psi_i(\mathbf{r})|^2. \quad (3.10)$$

The total energy functional of Kohn-Sham system, which needs to be minimized is defined as follows:

$$E_{\text{KS}}[n] = T_s[n] + \int n(\mathbf{r})V_{\text{ext}}(\mathbf{r}) d\mathbf{r} + E_{\text{H}}[n] + E_{\text{xc}}[n]. \quad (3.11)$$

Here,  $T_s[n]$  represents the kinetic energy of the non-interacting Kohn-Sham orbitals, calculated as the sum of the individual contributions from all electrons:

$$T_s[n] = -\frac{\hbar^2}{2m_e} \sum_i^\alpha \int \psi_i^*(\mathbf{r}) \nabla^2 \psi_i(\mathbf{r}) d\mathbf{r}. \quad (3.12)$$

The  $E_{\text{H}}[n]$  is the Hartree term, which describes the electrostatic interaction between charge densities:

$$E_{\text{H}}[n] = \frac{e^2}{2} \iint \frac{n(\mathbf{r})n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r} d\mathbf{r}' = \frac{e^2}{2} \int V_{\text{H}}(\mathbf{r})n(\mathbf{r}) d\mathbf{r}. \quad (3.13)$$

Finally, the exchange–correlation energy  $E_{\text{xc}}[n]$  accounts for all many-body effects beyond the classical Hartree term and is defined as

$$E_{\text{xc}}[n] = (T[n] - T_s[n]) + (U[n] - E_{\text{H}}[n]), \quad (3.14)$$

where  $T[n]$  and  $U[n]$  are the exact kinetic and electron–electron interaction energies of the interacting system. The corresponding exchange–correlation potential is given by

$$V_{\text{xc}}(\mathbf{r}) = \frac{\delta E_{\text{xc}}[n]}{\delta n(\mathbf{r})}. \quad (3.15)$$

It's important to highlight that even though the density of the Kohn-Sham system corresponds to that of the many-body system, the Kohn-Sham orbitals  $\psi_i$  lack direct physical interpretation. Despite this disconnection from the many-body wave function, Kohn-Sham orbitals have proven highly valuable. Notably, their band structures often offer surprisingly accurate representations of experimental data, underscoring their practical utility.

## 3.4 Exchange-Correlation Functionals

While the exchange-correlation energy  $E_{xc}[n]$  remains unknown, various approximation methods have been developed to address this challenge. Among these, the most widely used methods are the Local Density Approximation (LDA) and the Generalized Gradient Approximation (GGA).

### 3.4.1 Local Density Approximation (LDA)

The core concept of LDA, introduced by Kohn and Sham in 1965, is to partition the inhomogeneous electron density of the system into many small regions. Each of these small regions is then approximated as a homogeneous electron gas. The exchange-correlation functional for the nonuniform electron gas is derived based on the density function,  $n(\mathbf{r})$ , of the uniform electron gas, enabling a self-consistent calculation. The LDA assumes that the XC energy depends locally on the electron density at each point, similar to the behavior of a homogeneous electron gas. The XC energy in the LDA methods is as follows:

$$E_{xc}^{LDA}[n] = \int n(\mathbf{r}) \epsilon_{xc}(n(\mathbf{r})) d\mathbf{r} \quad (3.16)$$

Here,  $\epsilon_{xc}(n(\mathbf{r}))$  represents the exchange-correlation energy of homogeneous electron gas with density  $n(\mathbf{r})$ , weighted by the probability  $n(\mathbf{r})$  of finding an electron at that position. This LDA is successful when electron density is smooth and does not undergo rapid variation such as metals [136].

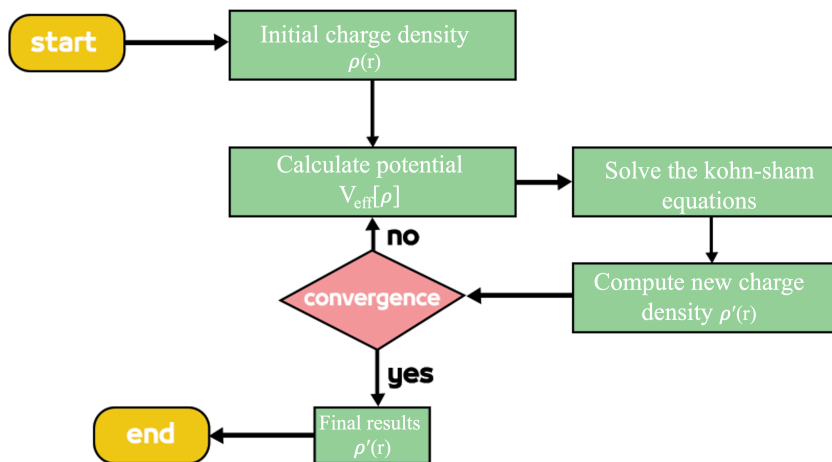
### 3.4.2 Generalized Gradient Approximation (GGA)

To improve accuracy beyond that of the LDA, the *generalized gradient approximation* (GGA) [137, 138] offers a more advanced approach. GGA builds upon LDA by incorporating both the local electron density and its gradient, resulting in a more general form:

$$E_{xc}[n] = \int n(\mathbf{r}), \epsilon_{xc}(n(\mathbf{r})), d\mathbf{r} + E_{XC}^{GGA}(n(\mathbf{r}), \nabla n(\mathbf{r})). \quad (3.17)$$

GGA functionals are favored over LDA for their enhanced depiction of electron density  $n(\mathbf{r})$ , which provides a more accurate representation of physical principles. Among the available GGA formulations, the Perdew, Burke, and Ernzerhof (PBE) functional [139] is widely adopted. GGAs offer improvements in total energies, atomization energies, energy barriers, and structural energy differences compared to

LDA [139]. GGA typically exhibits a stronger preference for density inhomogeneity compared to LDA [140]. In this thesis, the PBE functional [139], a widely adopted GGA method, is employed for the DFT study. The PBE functional remains one of the most commonly used approaches in contemporary DFT calculations due to its reliability and accuracy.



**Fig. 3.2.:** The self-consistency loop of density functional theory, employing the Kohn-Sham approach.

## 3.5 Spin Density Functional Theory

This study focuses on exploring the electronic and magnetic properties of rare-earth dichalcogenides (REDs), a class of materials that exhibit intriguing behaviors due to their strong spin-orbit coupling and correlated electron effects. To accomplish this purpose, the existing Density Functional Theory (DFT) framework must be expanded to incorporate spin-polarized systems. This extension involves decomposing the electron density,  $n(\mathbf{r})$ , into its spin components, enabling a more comprehensive analysis of magnetic and electronic behavior. The foundation for this approach lies in utilizing a  $2 \times 2$  density matrix, which replaces the conventional electronic and magnetization densities in spin-polarized systems [141]. This formalism is particularly useful for studying materials where spin interactions play a dominant role, such as in REDs.

The spin density matrix is defined as

$$\boldsymbol{\rho}(\mathbf{r}) = \frac{1}{2} [n(\mathbf{r}) \mathbb{I} + \mathbf{m}(\mathbf{r}) \cdot \boldsymbol{\sigma}] = \frac{1}{2} \begin{pmatrix} n(\mathbf{r}) + m_z(\mathbf{r}) & m_x(\mathbf{r}) - im_y(\mathbf{r}) \\ m_x(\mathbf{r}) + im_y(\mathbf{r}) & n(\mathbf{r}) - m_z(\mathbf{r}) \end{pmatrix}. \quad (3.18)$$

Here,  $\boldsymbol{\rho}(\mathbf{r})$  is the local  $2 \times 2$  spin-density matrix,  $n(\mathbf{r})$  denotes the total electron (charge) density, and  $\mathbf{m}(\mathbf{r}) = (m_x(\mathbf{r}), m_y(\mathbf{r}), m_z(\mathbf{r}))$  is the magnetization density vector, which characterizes the local spin polarization of the system. The symbol  $\boldsymbol{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$  denotes the vector of Pauli matrices, and  $\mathbb{I}$  is the  $2 \times 2$  identity matrix. The dot product  $\mathbf{m}(\mathbf{r}) \cdot \boldsymbol{\sigma}$  is understood as a scalar contraction in spin space, yielding a  $2 \times 2$  matrix.

For collinear magnetic systems, the spin-polarized Kohn-Sham equations are expressed as:

$$\begin{cases} \left[ -\frac{\hbar^2}{2m_e} \nabla + V_{\text{eff}}^{\uparrow}(\mathbf{r}) \right] \psi_i^{\uparrow}(\mathbf{r}) = \varepsilon_i^{\uparrow} \psi_i^{\uparrow}(\mathbf{r}) \\ \left[ -\frac{\hbar^2}{2m_e} \nabla + V_{\text{eff}}^{\downarrow}(\mathbf{r}) \right] \psi_i^{\downarrow}(\mathbf{r}) = \varepsilon_i^{\downarrow} \psi_i^{\downarrow}(\mathbf{r}). \end{cases} \quad (3.18)$$

These equations can be solved independently for spin-up and spin-down electrons. The effective potentials,  $V_{\text{eff}}^{\uparrow}(\mathbf{r})$  and  $V_{\text{eff}}^{\downarrow}(\mathbf{r})$ , incorporate both the external potential and the exchange-correlation potential, which depends on the spin density. As a result, physical observables such as the total energy depend on the electron density,  $n(\mathbf{r})$ , and the magnetization density,  $\mathbf{m}(\mathbf{r})$ , rather than the specific orientation of the magnetization. This simplification is valid for collinear systems, where the magnetization is aligned along a single axis.

The electronic structure problem in collinear magnets can also be described in terms of the spin-up and spin-down electron densities, which are given by:

$$n^{\uparrow}(\mathbf{r}) = \sum_i^{N_{\uparrow}} |\psi_i^{\uparrow}(\mathbf{r})|^2 \quad (3.19)$$

$$n^{\downarrow}(\mathbf{r}) = \sum_i^{N_{\downarrow}} |\psi_i^{\downarrow}(\mathbf{r})|^2. \quad (3.20)$$

The total electron density,  $n(\mathbf{r})$ , is then obtained by summing the spin-up and spin-down contributions:

$$n(\mathbf{r}) = n^{\uparrow}(\mathbf{r}) + n^{\downarrow}(\mathbf{r}). \quad (3.21)$$

Similarly, the magnetization density,  $m(\mathbf{r})$ , is derived from the difference between the spin-up and spin-down densities:

$$m(\mathbf{r}) = n^\uparrow(\mathbf{r}) - n^\downarrow(\mathbf{r}). \quad (3.22)$$

The magnetization density provides insight into the local magnetic moment distribution within the material. For instance, regions with a large  $m(\mathbf{r})$  indicate strong spin polarization, which is often associated with magnetic ordering.

Finally, the magnetic moment per unit volume, arising from spin polarization, is calculated by integrating the magnetization density over the entire volume,  $V$ :

$$M = \int_V m(\mathbf{r}) d^3\mathbf{r}. \quad (3.23)$$

This approach provides a robust framework for understanding the magnetic and electronic properties of REDs and other spin-polarized materials. By combining DFT with advanced computational techniques, it is possible to predict and analyze the behavior of these materials under various conditions.

## 3.6 Hubbard U Correction

DFT based on the GGA approach is a powerful method for describing the electronic properties of systems dominated by  $s$ - and  $p$ -orbitals. However, DFT  $\rightarrow$  GGA has notable shortcomings when dealing with localized electrons, such as those in systems containing  $d$ - and  $f$ -orbitals, as found in transition metals and rare-earth systems [142]. Additionally, DFT  $\rightarrow$  GGA is widely recognized for underestimating experimental band gaps [143, 144] and magnetic properties [145], particularly in systems with  $d$ - and  $f$ -orbitals. In strongly localized systems such as  $4f$  electrons it is essential to address the Coulomb interaction to obtain accurate ground-state properties. The DFT+ $U$  scheme, integrating DFT in some local or semilocal approximation to the exchange-correlation potential with the Hubbard model  $U$ , serves as a valuable tool for achieving this goal. This method, known for simplicity of its implementations and computational cost, addresses Coulomb repulsion by appending a  $U$  correction to the DFT Hamiltonian.

The concept behind DFT+ $U$  is straightforward: it combines the description of strongly correlated electronic states, typically associated with localized  $d$ - or  $f$ -

orbitals, using the Hubbard model [146], while treating remaining all delocalized electrons with conventional approximate DFT functionals:

$$E_{\text{DFT}+U}[n(\mathbf{r})] = E_{\text{DFT}}[n(\mathbf{r})] + E_{\text{Hubbard}}[n_{mm'}^{\mu\ell\sigma}] - E_{\text{dc}}[n^{\mu\ell\sigma}]. \quad (3.24)$$

The approach combines the conventional DFT functional,  $E_{\text{DFT}}[n(\mathbf{r})]$ , with the Hubbard functional,  $E_{\text{Hubbard}}[n_{mm'}^{\mu\ell\sigma}]$ , capturing electron-electron interactions among localized electrons within the framework of the Hubbard model. To mitigate issues of double-counting, it is essential to subtract the functional  $E_{\text{dc}}[n^{\mu\ell\sigma}]$  from the formulation, as the DFT component already addresses a segment of the correlation energy. The Hubbard term's calculation depends on the occupation numbers of localized orbitals, characterized by the specific indices attributed to them  $\mu$ ,  $\ell$ ,  $\sigma$ , and  $m$ . This emphasizes the exclusive application of the  $U$  correction to localized electrons. In this context,  $\mu$  denotes the atomic site,  $\sigma$  represents the spin index, and  $m$  corresponds to the subspace of orbitals indicated by the orbital number  $\ell$  of the localized state.

There are two formulations of the DFT+ $U$  method. In Dudarev's approach [147, 148], only the difference between the Coulomb parameter  $U$  and the exchange parameter  $J$ , denoted as  $(U - J)$ , contributes to the energy functional. In contrast, Liechtenstein's formulation [149] considers both  $U$  and  $J$  as independent contributions to the energy functional. In this work, the latter approach is employed.

## 3.7 Full-Potential Linearized Augmented Plane-Wave Method

Different strategies can be used to solve the Kohn-Sham equation (Eq. 3.8) or its spin-polarized counterpart (Eq. 3.18). A common method is to expand the single-particle wave functions in a set of basis functions that are well suited to the physical problem, such as augmented plane waves for periodic solids. A popular choice is to use an orthogonal plane-wave basis. This method is practical because plane waves are simple to handle, they diagonalize the kinetic energy operator, and they are well matched to Bloch's theorem, which makes them a natural solution for periodic systems.

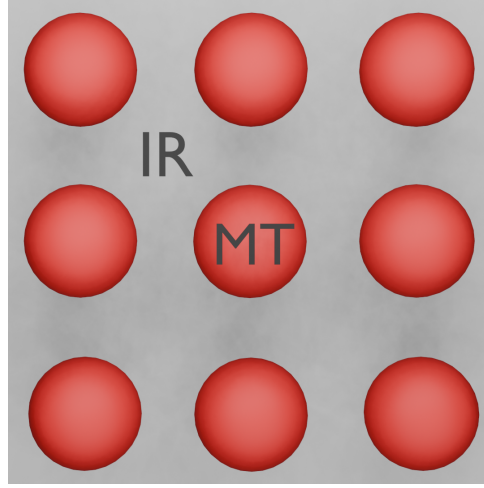
The translational invariance of solids implies that the Kohn-Sham wave functions can be expressed as Bloch states,  $\psi_{\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}}u_{\mathbf{k}}(\mathbf{r})$ , for any given point  $\mathbf{k}$  within

the Brillouin zone. The lattice-periodic functions  $u_{\mathbf{k}}(\mathbf{r})$  are expanded into plane waves using the reciprocal lattice vector  $\mathbf{G}$  as

$$\psi_{\mathbf{k}}(\mathbf{r}) = \sum_{\mathbf{G}} c_{\mathbf{k}}^{\mathbf{G}} \phi_{\mathbf{k}}^{\mathbf{G}}(\mathbf{r}), \quad \text{where} \quad \phi_{\mathbf{k}}^{\mathbf{G}}(\mathbf{r}) = \frac{1}{\sqrt{\Omega}} e^{i(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}}. \quad (3.25)$$

Here,  $\Omega$  is the unit cell volume, and  $c_{\mathbf{k}}^{\mathbf{G}}$  are the expansion coefficients.

The augmented plane-wave method (APW) [127, 150–152] partitions real space into two distinct regions: muffin-tin (MT) spheres centered around the nuclei, and the interstitial region (IR) between the spheres. Within the muffin-tins, the potential is approximated to resemble atomic-like spherical potentials, while in the interstitial region, the potential is often assumed to be constant or smoothly varying. The



**Fig. 3.3.:** The space in the (FL)APW method is divided into two regions: the muffin-tin (MT) spheres, centered on each atom (shown in red), and the interstitial region (IR) between the spheres (shown in gray).

core electrons are described as eigenstates in the spherical muffin-tin potential. The APW basis function of a valence electron is defined as:

$$\phi_{\mathbf{G}\mathbf{k}}(\mathbf{r}) = \begin{cases} \frac{1}{\sqrt{\Omega}} e^{i(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}}, & \text{IR region,} \\ \sum_{lm} a_{lm}^{\alpha\mathbf{G}}(\mathbf{k}) u_l^{\alpha}(r^{\alpha}, E) Y_{lm}(\hat{\mathbf{r}}^{\alpha}), & \text{MT region,} \end{cases} \quad (3.26)$$

where  $l$  and  $m$  are the orbital and magnetic quantum numbers, respectively;  $\alpha$  labels the atoms at positions  $\tau^{\alpha}$ ; and  $\mathbf{r}^{\alpha} = \mathbf{r} - \tau^{\alpha}$  denotes the local coordinate system relative to the center of atom  $\alpha$ . The coefficients  $a_{lm}^{\alpha\mathbf{G}}$  are chosen to ensure continuity of the wave functions at the muffin-tin boundaries.  $Y_{lm}(\hat{\mathbf{r}})$  are the spherical



harmonics functions depending on the angular part of the spatial vector  $\mathbf{r}^\alpha$ , and  $u_l^\alpha(r, E)$  are the solutions of the radial Schrödinger equation:

$$\left[ -\frac{\hbar^2}{2m} \frac{d^2}{dr^2} + \frac{\hbar^2 l(l+1)}{2mr^2} + V(r) \right] u_l(r) = E u_l(r). \quad (3.27)$$

The radial functions  $u_l^\alpha(r, E)$  depend on the band energy  $E$ , which leads to a non-linear eigenvalue problem in the solution of the Kohn-Sham equations. To address this, a linearization technique was proposed [153] based on a Taylor expansion of  $u_l$  around a fixed energy  $E_l^\alpha$ :

$$u_l^\alpha(r^\alpha, E) = u_l^\alpha(r^\alpha, E_l^\alpha) + (E - E_l^\alpha) \dot{u}_l^\alpha(r^\alpha, E_l^\alpha) + \mathcal{O}((E - E_l^\alpha)^2). \quad (3.28)$$

Here,  $\dot{u}_l^\alpha$  is the energy derivative of  $u_l^\alpha$ . Using this Taylor expansion helps to separate valence states from core states. Usually, only terms up to the first order are kept in the expansion. In the linearized augmented plane-wave (LAPW) method, the basis function becomes:

$$\phi_{\mathbf{G}\mathbf{k}}(\mathbf{r}) = \begin{cases} \frac{1}{\sqrt{\Omega}} e^{i(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}}, & \text{IR region,} \\ \sum_{lm} [a_{lm}^{\alpha\mathbf{G}}(\mathbf{k}) u_l^\alpha(r^\alpha, E_l^\alpha) + b_{lm}^{\alpha\mathbf{G}}(\mathbf{k}) \dot{u}_l^\alpha(r^\alpha, E_l^\alpha)] Y_{lm}(\hat{\mathbf{r}}^\alpha), & \text{MT region.} \end{cases} \quad (3.29)$$

The LAPW coefficients  $a_{lm}^{\alpha\mathbf{G}}$  and  $b_{lm}^{\alpha\mathbf{G}}$  are determined by applying boundary conditions at the interface between the interstitial region and the muffin-tin spheres. They are chosen to ensure that the basis functions are both continuous and have continuous first derivatives at the muffin-tin boundaries.

Another major advantage of the LAPW basis set is the simple extension of the APW method to the full potential and charge density of the periodic solid without any shape approximation. The non-spherical potential/charge density in the muffin-tin spheres and the warped potential/charge density in the interstitial region are taken into account variationally in the Hamiltonian. This is the all-electron full-potential linearized augmented plane wave (FLAPW) method, which is currently the gold standard for precision in electron structure methods for periodic solids.

All calculations presented in this thesis were carried out using the FLEUR code [154], an open-source density functional theory package that implements the FLAPW method.

### 3.8 Wannier Functions

In periodic crystals, the electronic structure is typically described using single-particle Bloch wavefunctions, which are expressed as in Equation 2.18. These wavefunctions simultaneously diagonalize the Hamiltonian and the lattice translation operator, making them a natural representation for describing periodic systems. However, the delocalized nature of Bloch states in real space can complicate the understanding of localized chemical and physical phenomena, such as bonding or defect interactions, in solids. To address this issue, Wannier functions [155–157] were introduced as a localized alternative to Bloch wavefunctions. They are constructed as Fourier transforms of Bloch states over the entire Brillouin zone (BZ) and are defined mathematically as:

$$W_0(\mathbf{r}) = \frac{V}{(2\pi)^3} \int_{BZ} |\psi_{n\mathbf{k}}(\mathbf{r})| d^3\mathbf{k}, \quad (3.30)$$

where  $V$  represents the real-space primitive cell volume and the integral is carried over the BZ. Due to the phase factor  $e^{i\mathbf{k}\cdot\mathbf{r}}$  in Bloch's theorem, the superposition of Bloch wavefunctions at different  $\mathbf{k}$ -points ensures that Wannier functions are localized in real space. The definition above confines the Wannier function to the original unit cell. To extend this description across a lattice, Wannier functions can incorporate lattice translational symmetry:

$$|\mathbf{R}n\rangle = \frac{V}{(2\pi)^3} \int_{BZ} d\mathbf{k} e^{-i\mathbf{k}\cdot\mathbf{R}} |\psi_{n\mathbf{k}}\rangle, \quad (3.31)$$

and

$$|\psi_{n\mathbf{k}}\rangle = \sum_{\mathbf{R}} e^{i\mathbf{k}\cdot\mathbf{R}} |\mathbf{R}n\rangle. \quad (3.32)$$

where  $\mathbf{R}$  is a real-space lattice vector. Bloch's theorem implies that Wannier functions exhibit translational symmetry. Specifically, the Wannier function centered at the lattice vector  $\mathbf{R} + \mathbf{R}_{(\text{shift})}$  is identical to the Wannier function at  $\mathbf{R}$ , but shifted in real space by  $\mathbf{R}_{(\text{shift})}$ . This property demonstrates that Wannier functions maintain their shape across unit cells, differing only in their positions.

Equations 3.30 and 3.31 are suitable for describing a single band that does not exhibit degeneracies at any  $\mathbf{k}$ -point. Additionally, these equations can be extended to a set of bands that may be degenerate among themselves at specific symmetry points or lines, but remain separated from other bands by finite energy gaps across the entire Brillouin zone. Such sets of bands are referred to as composite bands. In these cases, it becomes necessary to introduce a specific index, denoted as  $n$ , to label the individual bands within the composite group. This index must be consis-

tently incorporated into both the Wannier function and the Bloch wavefunction to distinguish between the bands effectively. It is well established that while Wannier functions serve as a powerful alternative to Bloch wavefunctions, their construction is not inherently unique. This ambiguity, particularly in systems with composite bands, can be resolved by applying a unitary transformation to the Bloch states. This transformation ensures the proper definition and localization of the Wannier functions while maintaining the physical consistency of the electronic structure:

$$|u_{n\mathbf{k}}(\mathbf{r})\rangle = \sum_m U_{mn}^{(\mathbf{k})} |u_{m\mathbf{k}}(\mathbf{r})\rangle \quad (3.33)$$

with  $U_{mn}^{(\mathbf{k})}$  representing the arbitrary unitary rotation that mixes the bands at each point in  $\mathbf{k}$ -space. The matrix  $U_{mn}^{(\mathbf{k})}$  is chosen such that it optimizes the localization properties of the resulting Wannier functions. Subsequently, the Wannier functions from Eq. 3.30 are given by:

$$|w_{n\mathbf{R}}(\mathbf{r})\rangle = \int_{\text{BZ}} \sum_m U_{mn}^{(\mathbf{k})} |\psi_{m\mathbf{k}}(\mathbf{r})\rangle e^{-i\mathbf{k}\cdot\mathbf{R}} d^3\mathbf{k} \quad (3.34)$$

It is important to emphasize that the selection of Wannier functions is not unique due to the freedom in the choice of the unitary matrix  $U_{mn}^{(\mathbf{k})}$ .

### 3.8.1 Maximally Localized Wannier Functions

Significant progress toward the practical applications of Wannier functions was achieved by Marzari and Vanderbilt [158], who introduced the concept of maximally localized Wannier functions (MLWFs). To address not only composite but also entangled energy bands, the method was later extended. By imposing the constraint of maximal localization, this approach ensures a unique Wannier basis set and establishes an optimally smooth gauge for the Bloch states. This constraint also minimizes the spread of functional  $\Omega$ , defined as:

$$\Omega = \sum_n \left[ \langle w_{n0}(\mathbf{r}) | \mathbf{r}^2 | w_{n0}(\mathbf{r}) \rangle - \langle w_{n0}(\mathbf{r}) | \mathbf{r} | w_{n0}(\mathbf{r}) \rangle^2 \right] = \sum_n \left[ \langle \mathbf{r}^2 \rangle_n - \langle \mathbf{r} \rangle_n^2 \right], \quad (3.35)$$

The detailed method and the technical procedure for calculating MLWFs are outlined in reference [159].

## 3.9 ORBITRANS

Having established the theoretical framework underlying the response functions considered in this work, we now turn to their numerical evaluation. Orbital, spin, and charge transport properties are computed from first principles using the in-house computational code ORBITRANS [160], based on Wannier interpolation and linear-response theory.

ORBITRANS is a specialized in-house computational code developed for calculating orbitronic properties using the Wannier interpolation method. The Wannier interpolation technique enables efficient calculations on dense  $k$ -meshes while retaining first-principles accuracy. ORBITRANS is fully interfaced with the FLEUR [154] and WANNIER90 [161] codes, allowing integration with first-principles DFT calculations. ORBITRANS is based on the theoretical framework introduced in Ref. [106] and enables accurate and efficient evaluation of transport phenomena in materials with complex electronic structures.

Working in the basis of maximally localized Wannier functions (MLWFs) (see Sec. 3.8), ORBITRANS supports the calculation of key quantities such as Berry curvature, anomalous Hall conductivity, spin Hall conductivity, and orbital Hall conductivity. These quantities are computed using the Kubo linear-response formalism [Eqs. 2.24, 2.26, and 2.28]. Its capabilities make ORBITRANS a powerful tool for investigating the interplay between spin-orbit coupling, magnetism, and orbital effects, particularly in low-dimensional and topologically nontrivial systems. With its combination of precision and scalability, ORBITRANS plays a central role in the computational study of phenomena at the heart of the emerging field of orbitronics.

ORBITRANS is well suited for large-scale systems and dense Brillouin-zone sampling, as it supports hybrid MPI/OpenMP parallelization, enabling efficient high-performance computing on modern architectures.

### 3.9.1 Workflow and Methodology

The practical implementation of ORBITRANS follows a three-step workflow, described below.

In the first step, a self-consistent density functional theory (DFT) calculation is performed to determine the electronic structure and density of states of the material.

This is carried out using the FLEUR code [154], which employs the full-potential linearized augmented plane-wave (FLAPW) method. The FLAPW approach provides high accuracy and enables direct access to orbital-resolved quantities, including the orbital angular momentum (OAM). In the second step, maximally localized Wannier functions (MLWFs) are generated from the converged Kohn-Sham states via the interface between FLEUR and WANNIER90 [159]. These functions provide an efficient real-space representation of the electronic structure, preserving the orbital character and enabling accurate interpolation across the Brillouin zone. In the final step, the Hamiltonian and relevant operators, including spin and orbital angular momentum operators, expressed in the MLWF basis are diagonalized to obtain the interpolated Bloch eigenstates and corresponding energy eigenvalues. These quantities are subsequently used to evaluate the desired response functions. All transport calculations presented in this thesis were performed within the Wannier representation using ORBITRANS.

# Engineering the Orbital Hall Effect in nonmagnetic TMDs

In this chapter, we explore the engineering of the orbital Hall effect (OHE) across various examples of transition metal dichalcogenides (TMDs), specifically focusing on monolayers, bilayers, and hetero-bilayer structures with electronic properties that range from semiconducting to metallic and semimetallic. The study of spin and orbital Hall effects in these materials offers valuable insights due to their distinctive electronic and structural characteristics. By analyzing monolayer and bilayer configurations of MoS<sub>2</sub> and NbSe<sub>2</sub>, and extending our investigation to MoS<sub>2</sub>/NbSe<sub>2</sub> and MoS<sub>2</sub>/WSe<sub>2</sub> hetero-bilayers under conditions such as biaxial strain, we aim to reveal the robustness and tunability of the OHE in TMDs. These findings not only demonstrate the stability of the orbital Hall insulator state across diverse structural configurations but also provide a foundation for designing strain-tunable orbitronic devices with promising applications in next-generation spintronics and orbitronics.

*Some results presented in Chapter 4.8 have already been published in Nádorník, Zeer et al., Adv. Mater. Interfaces 9, 2201675 (2022) [162].*

## 4.1 Introduction

2D TMDs have attracted considerable attention due to their potential as a versatile platform for various spintronics effects. TMDs display a wide array of electronic phases, including semiconductors [116, 163, 164], semimetals [165], metals [165], and superconductors [166]. This variety enables innovative design paradigms incorporating multiple complex phases of matter. Furthermore, the combination of low crystal symmetry and spin-orbit coupling (SOC) in TMDs [167] can give rise to exotic excitations and unique transport properties. For example, recent studies suggest that H-phase TMDs, which lack inversion symmetry, can facilitate various Hall effects, such as the anomalous Hall effect (AHE) [168], the valley Hall effect (VHE) [169, 170], the spin Hall effect [171, 172], and even the OHE [51, 173].



## 4.2 Computational Details

Our first-principles calculations were conducted using the density functional theory (DFT) code FLEUR [154], which employs the full-potential linearized augmented plane wave (FLAPW) method [152]. For the exchange-correlation functional, we used the Perdew-Burke-Ernzerhof approximation [139]. The monolayer, bilayer, and hetero-bilayer structures of MoS<sub>2</sub>, NbSe<sub>2</sub>, and WSe<sub>2</sub> were optimized by relaxing the atomic positions to achieve their lowest-energy configurations, as summarized in Table 4.1. The spin-orbit coupling (SOC) effect was included in all calculations using the second-variation approach.

We performed self-consistent calculations on a  $16 \times 16 \times 1$  Monkhorst-Pack grid within the first Brillouin zone to ensure accurate electronic structure convergence. For the TMD materials studied, the SOC influence on orbital and spin Hall conductivities was carefully analyzed through the density of states (DOS) and transport properties. Maximally localized Wannier functions (MLWFs) were constructed from the Bloch wave functions obtained from DFT, using the Wannier90 package [161]. For MoS<sub>2</sub>, WSe<sub>2</sub> and NbSe<sub>2</sub> systems, MLWFs were generated from the  $d$  orbitals of Mo, W and Nb and the  $p$  orbitals of S and Se atoms, which are the primary contributors near the Fermi level. For accurate orbital Hall and spin Hall conductivity calculations, a mesh of  $8 \times 8 \times 1$   $k$ -points was used to obtain MLWFs for each system. The matrix elements of spin and orbital angular momentum operators were first evaluated in the Bloch basis and subsequently transformed into the Wannier function basis, enabling detailed transport property analysis under varying conditions, including biaxial strain.

To analyze the transport properties of MoS<sub>2</sub>, NbSe<sub>2</sub>, WSe<sub>2</sub>, and their hetero-bilayer configurations, we computed the intrinsic Berry curvature contributions to the SHC and OHC using the MLWF framework, as described in Section 3.9. MLWF Hamiltonian was constructed for each system, enabling precise calculations of the  $xy$  components of the SHC and OHC. These quantities were evaluated as Brillouin zone integrals over a dense  $300 \times 300$   $k$ -point mesh, ensuring high-resolution insights into the transport properties using Equations 2.26 and 2.28.

## 4.3 Spin and Orbital Hall Effects in Monolayer and Bilayer H-MoS<sub>2</sub>

In this section, we start by taking the H-phase monolayer and bilayer of MoS<sub>2</sub> as a representative example to demonstrate the consistency of our results with references [51].

**Tab. 4.1.:** Key properties of MoS<sub>2</sub>, NbSe<sub>2</sub>, WSe<sub>2</sub>, and their hetero-bilayer configurations, including lattice constants, band gaps, orbital Hall conductivity (OHC), and spin Hall conductivity (SHC) values. For metallic systems, the OHC is given as two values: the first corresponds to the value at the Fermi energy, and the second value in parentheses corresponds to the value in the main gap above the Fermi energy.

| Material                                           | Lattice Constant (Å) | Band Gap (eV)   | OHC ( $\frac{e}{2\pi}$ ) | SHC ( $\frac{e}{2\pi}$ ) |
|----------------------------------------------------|----------------------|-----------------|--------------------------|--------------------------|
| Monolayer MoS <sub>2</sub>                         | 3.182                | 1.61            | 2.1                      | 0.0                      |
| Monolayer NbSe <sub>2</sub>                        | 3.487                | Metallic        | 1.0 (1.7)                | −0.02                    |
| Monolayer WSe <sub>2</sub>                         | 3.326                | 1.27            | 1.5                      | 0.0                      |
| Bilayer MoS <sub>2</sub>                           | 3.160                | 1.18 (indirect) | 3.8                      | 0.0                      |
| Bilayer NbSe <sub>2</sub>                          | 3.440                | Metallic        | 3.2 (3.4)                | −0.7                     |
| Hetero-bilayer MoS <sub>2</sub> /NbSe <sub>2</sub> | 3.286                | Metallic        | 3.1 (3.4)                | −0.013                   |
| Hetero-bilayer MoS <sub>2</sub> /WSe <sub>2</sub>  | 3.259                | 0.3             | 3.6                      | 0.0                      |

4.3.1 Spin and Orbital Hall Effects in Monolayer H-Phase MoS<sub>2</sub>

The 2H-phase monolayer MoS<sub>2</sub>, known for its noncentrosymmetric structure, serves as a prototypical example for exploring the spin and orbital Hall effects in 2D TMDs. This material’s unique electronic structure, combined with strong SOC primarily originating from the *d*-orbitals of Mo atoms, makes it an ideal candidate for studying spin-orbit-induced transport phenomena [174]. The interplay between inversion symmetry breaking and significant SOC leads to band splitting that is essential for understanding spin and orbital transport in 2D systems.

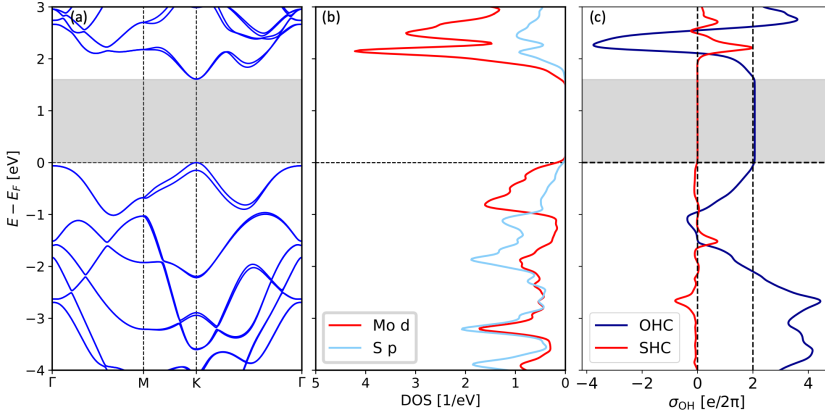
In Fig. 4.1 (a-b), we present the calculated band structure and density of states (DOS) for the H-phase monolayer MoS<sub>2</sub> with SOC. Our DFT calculations reveal a direct energy bandgap of 1.60 eV, with both the conduction and valence bands located at the *K*-point, consistent with previous studies [172, 175]. The inclusion of SOC results in a noticeable splitting of the valence band at the *K*-point, underscoring its effect on the band structure. The DOS plot highlights strong hybridization between the Mo-*d* states and the S-*p* states.

In Fig. 4.1 (c), we present the spin Hall conductivity (SHC) (red line) and orbital Hall conductivity (OHC) (blue line) as functions of band filling. The SHC exhibits two prominent sharp peaks with opposite signs for p-doped monolayer MoS<sub>2</sub>, while





for n-doped MoS<sub>2</sub>, a significant peak is centered around 2.2 eV. Within the primary semiconducting energy gap, the SHC vanishes, reflecting its topologically trivial nature [171]. In contrast, the OHC shows dominant contributions across the set Fermi energy, demonstrating that the OHC is more pronounced than the SHC in these materials. Most notably, within the main bandgap, the OHC exhibits a substantial plateau with a magnitude of approximately  $2.2 \frac{e}{2\pi}$ , consistent with previous findings [51, 94]. This behavior highlights the fundamental characteristic of the OHC, where significant orbital transport occurs even in the absence of SHE. Interestingly, the orbital Hall effect in MoS<sub>2</sub> can occur without SOC, showcasing the intrinsic capability of these materials for robust orbital transport.



**Fig. 4.1.:** Electronic structure and transport properties of monolayer MoS<sub>2</sub> as a function of band filling. Panel (a) displays the band structure with spin-orbit coupling (SOC) included, revealing a direct bandgap at the  $K$  point. The shaded region marks the energy range where significant orbital Hall conductivity (OHC) is observed. In panel (b), the spin-resolved density of states (DOS) is shown, highlighting the contributions from Mo- $d$  orbitals (red) and S- $p$  orbitals (sky-blue). These DOS features illustrate strong hybridization between Mo- $d$  and S- $p$  states near the Fermi level, which influences the electronic and transport properties of MoS<sub>2</sub>. Panel (c) illustrates the calculated orbital Hall conductivity (OHC, blue line) and spin Hall conductivity (SHC, red line) as functions of band filling. The OHC exhibits notable plateaus, particularly in the shaded energy region where it maintains a high value of approximately  $2.2 \frac{e}{2\pi}$ , suggesting a robust orbital Hall effect within the bandgap. In contrast, the SHC vanishes within the main semiconducting gap due to the topologically trivial nature of MoS<sub>2</sub>, but displays small peaks outside this region.

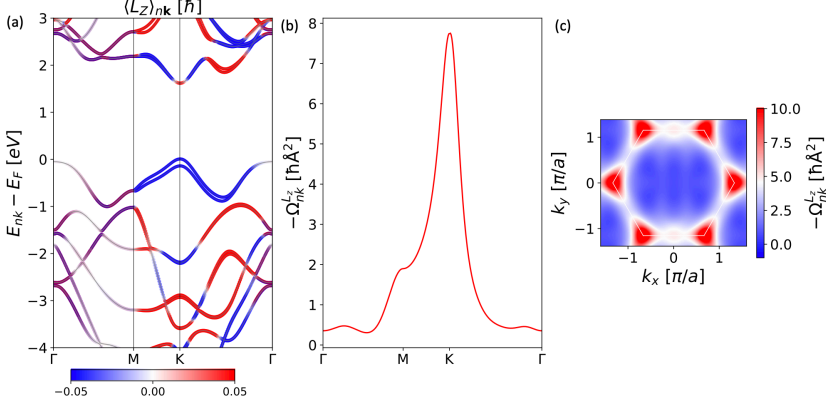
### 4.3.2 Analysis of Orbital Hall Conductivity in Monolayer H-MoS<sub>2</sub>

We turn our attention to analyzing the OHC in monolayer MoS<sub>2</sub>. For this purpose, we plot the out-of-plane orbital polarization  $\langle L_z \rangle_{nk}$  of the occupied states of MoS<sub>2</sub>, as shown in Fig. 4.2 (a). The color map indicates the evolution of orbital character along high-symmetry paths, highlighting changes that result from strong hybridization between Mo-*d* and S-*p* orbitals. This interaction significantly affects the electronic structure and contributes to the overall OHC. Notably, we observe an orbital band inversion around the maximum of the valence band and the minimum of the conduction band at the *K* point. This inversion is a key indicator of the emergence of an orbital Hall insulating phase, where orbital current can be sustained independently of spin transport.

In Fig. 4.2 (b), we present the orbital Berry curvature  $-\Omega_{nk}^{L_z}$  along the high-symmetry paths. The pronounced peak at the *K* point reflects a substantial orbital band inversion and signifies a strong contribution to the OHC at the Fermi energy. Fig. 4.2 (c) shows the distribution of orbital Berry curvature summed over all occupied bands across the Brillouin zone, emphasizing high-curvature magnitude near the *K* point. This reinforces the potential of MoS<sub>2</sub> as an orbital Hall insulator, capable of robust orbital transport that could be utilized in orbitronic applications.

### 4.3.3 Spin and Orbital Hall Effects in Bilayer H-Phase MoS<sub>2</sub>

In the realm of MoS<sub>2</sub> bilayer physics, the OHC exhibits a plateau with a magnitude nearly double that observed in the monolayer case, reaching approximately  $4.2 \frac{e}{2\pi}$ . Meanwhile, the SHC remains null within the main energy gap, consistent with its topologically trivial nature. Notably, the quantized-like plateaus of the OHC extend from the valence band maximum at the *K* symmetry point to the conduction band minimum, even as the maximum of the valence band shifts towards the  $\Gamma$  point. This observation is crucial, as the valley Hall effect would typically predict a zero net OHC due to the cancellation between contributions from the *K* and *K'* valleys [51].

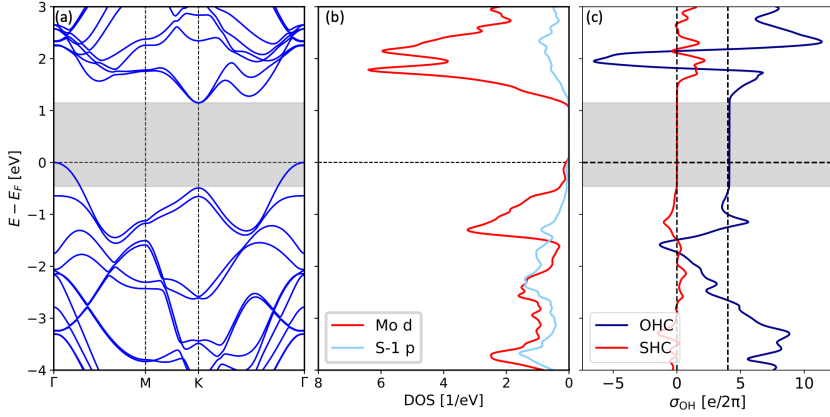


**Fig. 4.2.:** (a) Evolution of the out-of-plane orbital polarization,  $\langle L_z \rangle_{nk}$ , for the occupied states of monolayer MoS<sub>2</sub>, with color bars indicating the magnitude in units of  $\hbar$ . This polarization reflects the orbital angular momentum contributions across the electronic bands, highlighting variations around high-symmetry points in the Brillouin zone. (b) Orbital Berry curvature distribution along high-symmetry paths within the Brillouin zone, showcasing the pronounced peaks that contribute to the orbital Hall conductivity. (c) Summed distribution of the orbital Berry curvature over all occupied bands at each  $k$ -point within the Brillouin zone, evaluated at the Fermi energy. This map emphasizes the concentration of orbital Berry curvature near specific  $K$ -points.

## 4.4 Spin and Orbital Hall Effects in Monolayer and Bilayer NbSe<sub>2</sub>

Our investigation extends to the OHC and SHC in monolayer and bilayer metallic H-NbSe<sub>2</sub>, chosen for its relevance as a metallic representative among TMDs. The monolayer NbSe<sub>2</sub> results reveal plateau-like OHC features above the Fermi energy comparable to those seen in monolayer MoS<sub>2</sub>, exhibiting a magnitude of  $1.7 \frac{e}{2\pi}$ . These features are particularly prominent within the energy range spanning from the top of the valence band to the conduction band minimum at the  $K$  symmetry points above the Fermi energy. At the Fermi level, calculated for comparison with experimental data from our collaboration [162], the SHC displays a negative sign relative to the OHC. Notably, the OHC is approximately 20 times greater than the SHC, emphasizing its dominant role in the transport properties of NbSe<sub>2</sub>. Overall, the OHC also dominates over the SHC across all band fillings.

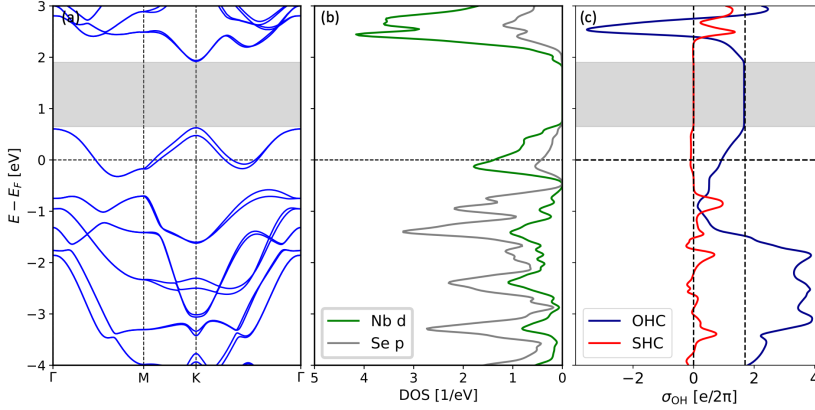
In Fig. 4.4 (a), the band structure of monolayer NbSe<sub>2</sub> with SOC reveals key electronic features around high-symmetry points, underscoring the complex interactions



**Fig. 4.3.:** Electronic structure and transport properties of bilayer  $\text{MoS}_2$  as a function of band filling. Panel (a) shows the band structure with spin-orbit coupling (SOC) included, revealing band splitting due to interlayer interactions at the  $K$ -symmetry points. Panel (b) presents the spin-resolved density of states (DOS) with contributions from Mo- $d$  (red) and S- $p$  (sky-blue) orbitals, showing hybridization effects near the Fermi level that influence electronic and transport behavior. Panel (c) displays the calculated orbital Hall conductivity (OHC, blue line) and spin Hall conductivity (SHC, red line) as functions of band filling. Despite the anticipated cancellation of valley contributions, the OHC exhibits prominent features, whereas the SHC remains minimal. These findings underscore the unique transport properties of bilayer  $\text{MoS}_2$ , where the valley effect does not contribute directly to the OHC.

that influence its transport properties. The spin-resolved density of states (DOS) in panel (b) highlights the contributions from Nb- $d$  states and Se- $p$  states, shown in green and gray, respectively, which illustrate significant hybridization between these orbitals. This hybridization plays a crucial role in shaping the transport characteristics, particularly the OHC and SHC. Fig. 4.4 (c) shows The SHC and OHC as functions of band filling. The results present the potential H-NbSe<sub>2</sub> as a matalic TMDs candidate for orbitronic applications.

The OHC behavior of bilayer H-phase NbSe<sub>2</sub> shows notable similarities to that of bilayer H-phase  $\text{MoS}_2$ , particularly with the presence of plateaus. For bilayer NbSe<sub>2</sub>, the magnitude of these plateaus significantly increases, doubling from  $1.7 \frac{e}{2\pi}$  to  $3.4 \frac{e}{2\pi}$  as shown in Fig 4.5. This change in conductance quantization is confined within the global band gap, situated above the valence band maximum. This outcome challenges the typical predictions of the valley Hall effect, which would suggest a net-zero OHC due to opposite contributions from the valleys. At the Fermi energy, the SHC shows a negative peak, contrasting sharply with the large positive OHC



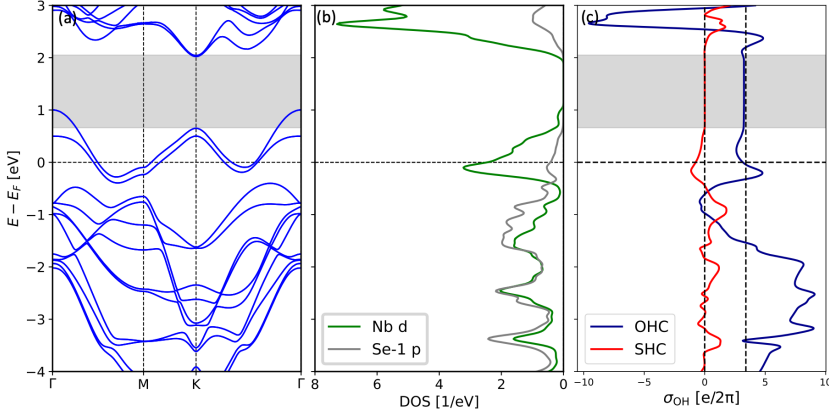
**Fig. 4.4.:** Electronic structure and transport properties of monolayer NbSe<sub>2</sub> in its metallic phase, shown as a function of band filling. Panel (a) displays the band structure with spin-orbit coupling (SOC) included, highlighting metallic characteristics with bands crossing the Fermi level, indicative of a continuous density of states that confirms the metallic nature of monolayer NbSe<sub>2</sub>. Panel (b) illustrates the spin-resolved density of states (DOS), with contributions from Nb-*d* orbitals (green) and Se-*p* orbitals (gray). Notable hybridization between Nb-*d* and Se-*p* states is visible. Panel (c) presents the orbital Hall conductivity (OHC, blue line) and spin Hall conductivity (SHC, red line) as functions of band filling. The OHC shows robust values across the Fermi level, demonstrating substantial orbital contributions in the metallic regime, while the SHC displays smaller peaks, consistent with the influence of SOC on transport properties.

peak. The pronounced OHC highlights substantial orbital contributions originating near the *K* point.

Overall, the results show the complex interaction of orbital dynamics and interlayer effects in bilayer NbSe<sub>2</sub>, providing deeper insight into its transport properties. The persistence of quantized OHC from the valence band maximum to the conduction band minimum indicates potential for orbitronic applications, leveraging orbital currents for advanced functionalities. The observed interplay between Nb-*d* and Se-*p* states supports the understanding of orbital transport mechanisms in metallic TMDs, paving the way for engineered quantum transport in layered materials.

## 4.5 Transport Characteristics of MoS<sub>2</sub>–NbSe<sub>2</sub> Hetero–Bilayers

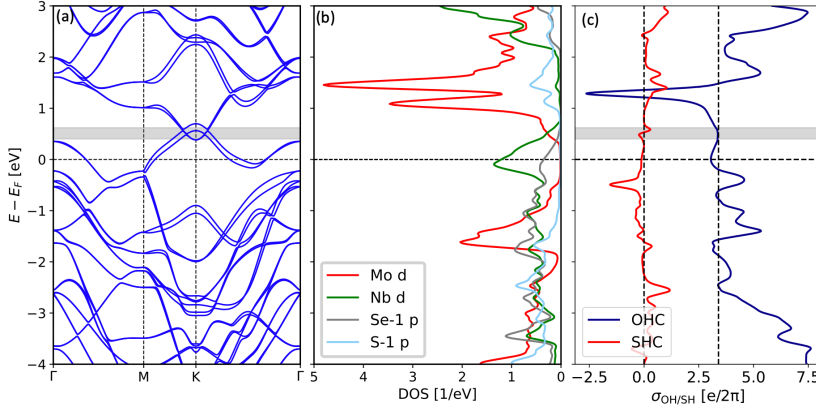
After examining the orbital Hall insulator properties in both semiconducting and metallic monolayers and bilayers, we proceed to explore the transport characteristics of MoS<sub>2</sub>–NbSe<sub>2</sub> hetero-bilayers. This direction allows us to integrate the distinct



**Fig. 4.5.:** Electronic structure and transport properties of bilayer NbSe<sub>2</sub> as a function of band filling. Panel (a) presents the band structure including spin-orbit coupling (SOC), revealing key features near high-symmetry points. Panel (b) shows the spin-resolved density of states (DOS), with contributions from Nb-*d* and Se-1 *p* states depicted in green and gray, respectively, highlighting the dominant role of Nb-*d* states at the Fermi level. In Panel (c), the orbital Hall conductivity (OHC) and spin Hall conductivity (SHC) are plotted as functions of band filling, with the bilayer configuration showing an enhanced small peak in the SHC at the Fermi energy. This enhancement in SHC indicates notable spin-related transport effects in the bilayer structure, alongside a stable and robust OHC response across the energy range.

properties of semiconducting MoS<sub>2</sub> and metallic NbSe<sub>2</sub>, forming a heterostructure that supports complex interlayer interactions and unique transport behavior. By constructing and studying this MoS<sub>2</sub>-NbSe<sub>2</sub> heterostructure, we aim to reveal how the individual monolayer properties contribute to the combined system's spin and orbital Hall conductivities.

The electronic structure of the NbSe<sub>2</sub>-MoS<sub>2</sub> hetero-bilayer exhibits metallic characteristics with a semimetal structure above the Fermi energy. Analysis of the DOS reveals that electronic states at the Fermi level are predominantly contributed by the NbSe<sub>2</sub> layer, while the MoS<sub>2</sub> layer contributes states above the Fermi level, around 0.6 eV. In this energy region, bands come close to crossing or nearly touch (Fig. 4.6 (a-b)), suggesting that doping or introducing defects could shift the Fermi level to this band-touching region. Our electronic structure are in excellent agreement with those reported in [176]. The OHC in this heterostructure appears to be an additive effect, combining contributions from both MoS<sub>2</sub> and NbSe<sub>2</sub> layers, resulting in a robust OHC signal. The behavior of the OHC in the NbSe<sub>2</sub>-MoS<sub>2</sub> bilayer differs from that observed in either monolayer or bilayer forms of MoS<sub>2</sub> and



**Fig. 4.6.:** Electronic structure and transport properties of the MoS<sub>2</sub>/NbSe<sub>2</sub> heterobilayer as a function of band filling. Panel (a) shows the band structure with spin-orbit coupling (SOC), highlighting metallic characteristics and a near-band-touching point just above the Fermi level. The gray-shaded region denotes the energy range around this near-touching point, where significant changes in electronic behavior are observed. Panel (b) presents the spin-resolved density of states (DOS), showing contributions from Mo-*d* (red), Nb-*d* (green), Se-*p* (gray), and S-*p* (sky-blue) orbitals, with the NbSe<sub>2</sub> layer providing the dominant states at the Fermi level. In panel (c), the orbital Hall conductivity (OHC) and spin Hall conductivity (SHC) are plotted as functions of band filling, with the gray-shaded region indicating where the OHC reaches a quantized plateau of  $3.4 \frac{e}{2\pi}$ , signaling a robust orbital Hall effect within this energy range. Notably, the SHC remains minimal around the Fermi level, underscoring the dominance of orbital transport in this heterostructure.

NbSe<sub>2</sub>. Unlike the decay of the OHC signal below the *K* symmetry point, located at the maximum of the valence band in monolayer MoS<sub>2</sub>, the heterostructure configuration stabilizes the OHC, maintaining a high signal without significant decay. This sustained response is closely tied to the positioning of the *K*-point peak in monolayer MoS<sub>2</sub>, highlighting the distinct orbital transport characteristics arising in the combined bilayer system.

Notably, the orbital Hall insulator remains robust around the Weyl crossing, indicated by the shaded region in both the electronic structure in Fig. 4.6 (a) and the orbital Hall conductivity in Fig. 4.6 (c). This region exhibits a quantized OHC of  $3.4 \frac{e}{2\pi}$  within the energy range of 0.3 to 0.6 eV above the Fermi level. In contrast, the SHC displays two small peaks, reversing in sign from  $-0.2 \frac{e}{2\pi}$  at 0.3 eV to  $0.3 \frac{e}{2\pi}$  at 0.6 eV. While the SHC around the Fermi energy is null in individual monolayers, the heterostructure configuration enhances the SHC, leading to a noticeable response as a function of band filling. Exploring alternative stacking configurations,

as demonstrated in studies on MoTe<sub>2</sub>/WSe<sub>2</sub> bilayers, could potentially yield even greater SHC around the Fermi level [177].

## 4.6 Transport Properties as a Function of the Biaxial Strain of MoS<sub>2</sub>/WSe<sub>2</sub> Hetero-Bilayers

Motivated by the robustness of the orbital Hall insulator state observed in various monolayer and bilayer configurations with different structures, we extend our investigation to examine its stability under significant biaxial strain. To this end, we construct a hetero-bilayer structure composed of two distinct H-phase semiconductor monolayers, MoS<sub>2</sub> and WSe<sub>2</sub>. Hetero-bilayers of TMDs have previously garnered considerable attention for their ability to engineer bandgaps in semiconductor TMDs, offering new opportunities for advanced applications in electronics and spintronics [178]. In this study, we specifically aim to evaluate the influence of heterostructure design on the orbital Hall insulator state, focusing on how biaxial strain modulates its properties within the MoS<sub>2</sub>-WSe<sub>2</sub> hetero-bilayer, providing insights into strain-tunable orbitronics and spintronics device design.

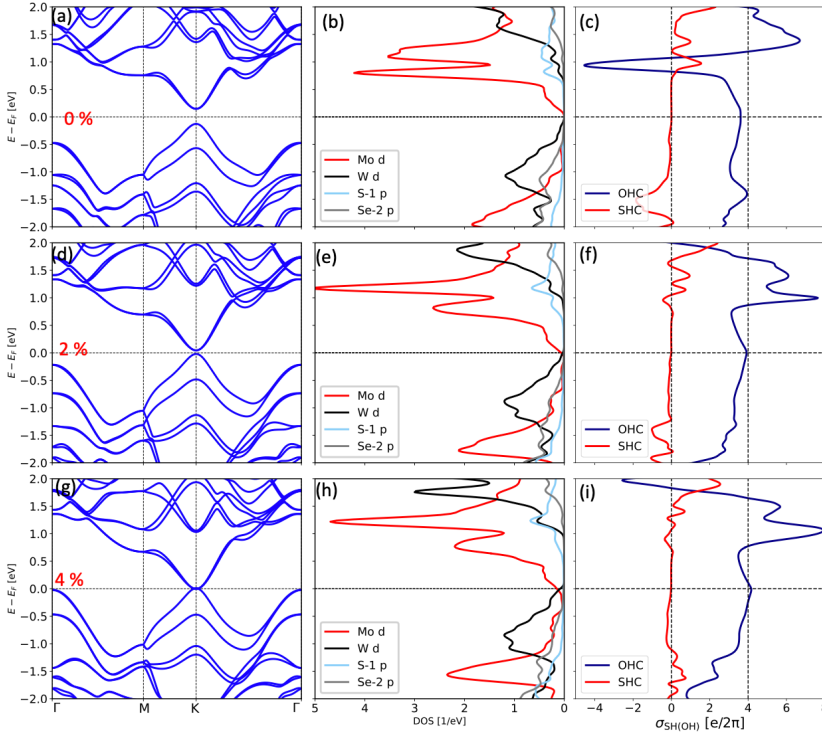
Starting with the unstrained MoS<sub>2</sub>-WSe<sub>2</sub> hetero-bilayer, the electronic structure with SOC is shown in Fig. 4.7. Notably, the MoS<sub>2</sub>-WSe<sub>2</sub> hetero-bilayer displays a direct band gap of approximately 0.3 eV, consistent with previous findings reported in [178]. In this unstrained configuration, both the valence band maximum and conduction band minimum are located at the *K* symmetry point, confirming a direct band gap structure. Under increasing tensile strain of 2% and 4%, the band gap decreases to 0.1 eV and 0.02 eV, respectively, indicating a trend toward metallic behavior at higher strain levels. This semiconducting background provides an ideal platform to explore the contribution of each layer, as both MoS<sub>2</sub> and WSe<sub>2</sub> layers exhibit OHC plateaus within their semiconducting band gaps. This reinforces the potential of the MoS<sub>2</sub>-WSe<sub>2</sub> hetero-bilayer as an effective orbital Hall insulator, even under strain, demonstrating its applicability for strain-tunable orbitronic applications.

The SHC in MoS<sub>2</sub>-WSe<sub>2</sub> primarily depends on the hybridization between the *p*-states of the chalcogenide atoms and the *d*-states of the metal atoms. This strong hybridization is a key factor in driving the SHC, as indicated by the pronounced SHC peaks in regions where this hybridization is significant, as observed in the DOS. Furthermore, the magnitude of the SHC peaks is directly influenced by the extent of *p*-state contributions, with higher *p*-state densities leading to increased



SHC values. This finding highlights the critical role of the  $p$ -states in generating the spin Hall conductivity (SHC), as shown in Figs. 4.5 and 4.6. This role will be further discussed in the context of REDs in Chapters 6 and 7.

Focusing on the OHC in the unstrained case, we observe that the orbital Hall insulator is maintained within the main band gap, gives rise to magnitude of almost  $3.7 \frac{e}{2\pi}$  with zero SHC in the background. Interestingly, the configuration here differs significantly, with the maximum of the valence band originating from the W  $d$ -states and the minimum of the conduction band stemming from the Mo  $d$ -states. The magnitude of the OHC is effectively the combined contributions from both MoS<sub>2</sub> and WSe<sub>2</sub>, reinforcing the hetero-bilayers robust orbital Hall response. Outside the main gap, up to the relative position of the  $K$  symmetry point of Mo, the orbital Hall insulator remains relatively stable. In the strained cases of 2% and 4% tensile strain, the orbital Hall insulator remains stable within the narrow band gap. The OHC increases with decreasing the band gap. Demonstrating that the orbital Hall insulator in TMDs is resilient across different heterostructure configurations and strain conditions.



**Fig. 4.7.:** Electronic structure and transport properties of  $\text{WSe}_2\text{-MoS}_2$  hetero-bilayers under biaxial strain: This figure showcases the impact of biaxial strain on the electronic structure and Hall conductivities of  $\text{WSe}_2\text{-MoS}_2$  hetero-bilayers. Panels (a)-(c) present the unstrained configuration: (a) illustrates the band structure with spin-orbit coupling (SOC), (b) displays the density of states (DOS) with W-d and W-p orbitals in black and gray, and Mo-d and Mo-p in red and sky blue, respectively. Panel (c) shows the spin Hall conductivity (SHC) and orbital Hall conductivity (OHC) as functions of band filling, offering insight into the transport behavior in the absence of strain. Panels (d)-(f) examine the effects of a 2% biaxial strain, while panels (g)-(i) show the impact of a 4% biaxial strain on the band structure, DOS, and Hall conductivities.

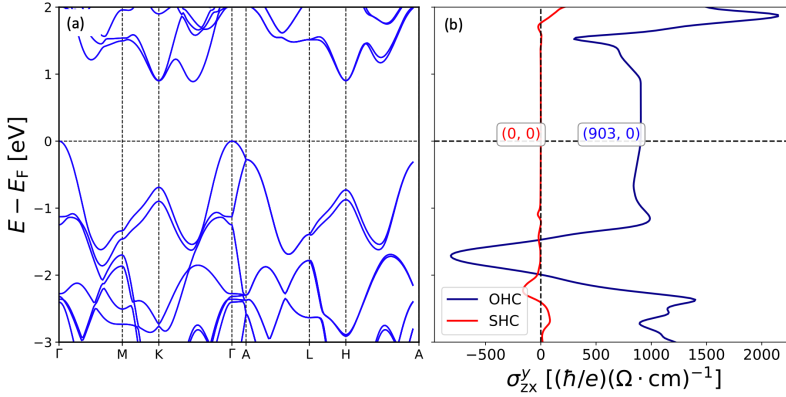
## 4.7 Spin and Orbital Hall Effects in Bulk H-Phase $\text{MoS}_2$

While monolayer and bilayer  $\text{MoS}_2$  have been extensively studied for their spin and orbital transport properties [51, 54, 94, 173], the bulk phase of  $\text{MoS}_2$  remains equally significant due to its widespread use in electronics and spintronics applications [14, 179]. A key motivation behind our interest in understanding spin and orbital transport in bulk H-phase  $\text{MoS}_2$  is that the bulk system provides unique insights into the role of interlayer coupling and the robustness of orbital transport

mechanisms beyond the 2D limit. Additionally, bulk systems are more experimentally accessible [180], making it essential to establish theoretical predictions that can be directly validated through experiments.

To gain insight into the bulk calculations, we focus on the H-phase MoS<sub>2</sub> as shown in Fig. 4.8. The electronic band structure reveals an indirect bandgap of 0.88 eV, which is consistent with the reported result in [181]. To address the experimental scenario of measuring conductivity, we calculate the SHC and OHC  $\sigma_{zx}^y$  for the spin/orbital current in the  $z$ -direction, polarized along  $y$ , under an electric field applied in the  $x$ -direction. Our results show that the OHC dominates, particularly within the main indirect bandgap region. At the Fermi energy, the OHC magnitude reaches  $903 \left(\frac{\hbar}{e}\right) (\Omega \cdot \text{cm})^{-1}$ , while the SHC is negligible, indicating that the orbital Hall insulator state still appears in the gap.

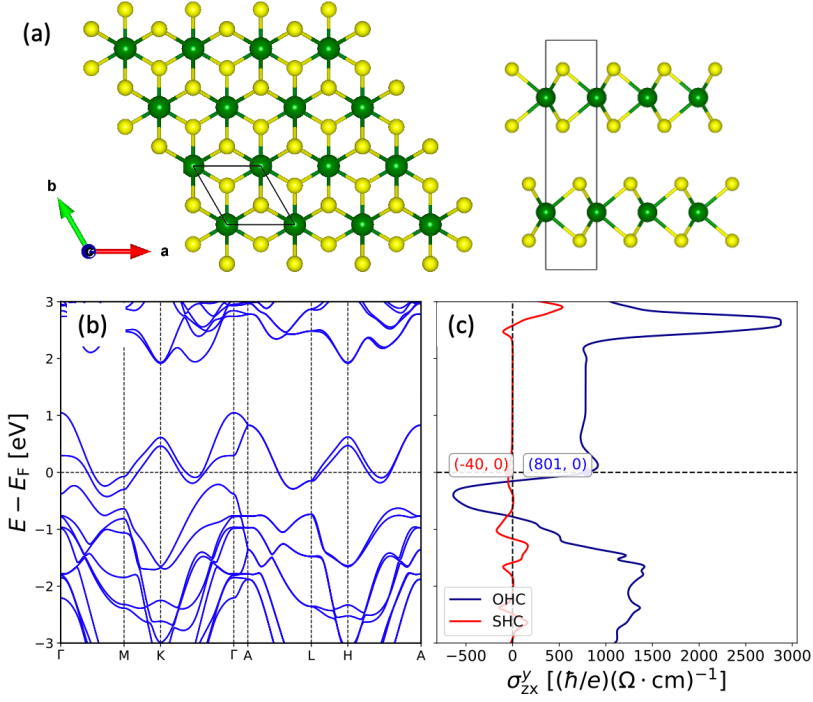
It is important to note that we have converted the units from  $\frac{e}{2\pi}$  to  $\left(\frac{\hbar}{e}\right) (\Omega \cdot \text{cm})^{-1}$  using this formula  $2\pi \cdot G_0 \cdot 10^8 \left(\frac{1}{V_{\text{cell}}}\right)$ , where  $G_0 = \left(\frac{1}{2}\right) \frac{2\hbar}{e^2} \approx 7.748 \times 10^{-5} \Omega^{-1}$ . This unit conversion ensures that the calculated SHC and OHC are dimensionally consistent with respect to the material's physical volume.



**Fig. 4.8.:** Electronic structure and transport properties of bulk H-phase MoS<sub>2</sub>. (a) the electronic band structure along high-symmetry points in the Brillouin zone. The band structure shows H-phase indirect band gap of 0.88 eV. (b) shows the spin Hall conductivity (SHC) and orbital Hall conductivity (OHC) as functions of band filling the red line corresponds to the SHC  $\sigma_{zx}^y$ , indicating the spin current in the  $z$ -direction polarized along  $y$  under an electric field applied in the  $x$ -direction. The region near the Fermi energy ( $E_F$ ) is highlighted, with the values of SHC and OHC marked as (0, 0) for SHC and (903, 0) for OHC.

## 4.8 Spin and Orbital Hall Effects in Bulk H-phase NbSe<sub>2</sub>

Here we perform similar calculations for the metallic H-phase NbSe<sub>2</sub>. At the Fermi energy, the material exhibits a negative SHC of approximately  $40 \left(\frac{\hbar}{e}\right) (\Omega \cdot \text{cm})^{-1}$ , while the OHC is much larger, reaching  $801 \left(\frac{\hbar}{e}\right) (\Omega \cdot \text{cm})^{-1}$ . This indicates that the material retains its orbital Hall insulating behavior, as shown in Fig.4.9. Furthermore, the negative SHC observed at the Fermi energy is consistent with the metallic nature of the H-phase of NbSe<sub>2</sub> and the high density of states around the Fermi level. Experimental measurements, such as those reported in [162], have shown that the primary spin-charge conversion occurs in the bulk of NbSe<sub>2</sub>, with a negative spin Hall angle. This aligns well with the calculated negative values of the SHC at the Fermi energy, further validating the theoretical predictions. These results highlight the interplay between spin and orbital transport phenomena in H-phase NbSe<sub>2</sub>, emphasizing its unique electronic and transport properties.



**Fig. 4.9.:** Electronic structure and transport properties of H-phase bulk NbSe<sub>2</sub>. (a) shows the atomic structure of the H-phase NbSe<sub>2</sub>, the left presents the top view, while the right side displays the side view of the structure with the layered arrangement of Nb (green) and Se (yellow) atoms. (b) presents the electronic band structure along high symmetry points. (c) shows the spin Hall conductivity (SHC) and orbital Hall conductivity (OHC) as functions of band filling. The red line corresponds to the SHC  $\sigma_{zx}^y$ , indicating the spin current in the  $z$ -direction polarized along  $y$  under an electric field applied in the  $x$ -direction. The blue line represents the OHC. The inset zooms into the region of Fermi energy ( $E = 0$ ), showing the values of SHC and OHC, marked as  $(-40, 0)$  for SHC and  $(801, 0)$  for OHC.

## 4.9 Conclusion

In this chapter we provide a comprehensive investigation into the spin and orbital Hall conductivities across various nonmagnetic TMD configurations. The robustness of the orbital Hall insulator is consistently observed in monolayers, bilayers, and hetero-bilayers, with notable resilience under biaxial strain in  $\text{MoS}_2/\text{WSe}_2$  systems with all quantized orbital Hall conductivities appearing on the background of vanishing SHC. Our findings underscore the additive nature of the OHC contributions in hetero-bilayers. The detailed analysis of band structures, density of states, and transport properties across these TMDs emphasizes the strong potential for orbitronic applications. These results not only enhance the fundamental understanding of the OHE in TMDs but also pave the way for tailored device designs that leverage strain and structural modifications to optimize transport characteristics.





# Spin and Orbital Transport in Rare-Earth Dichalcogenides: The Case of $\text{EuS}_2$

In this chapter, we extend our exploration of transition metal dichalcogenides (TMDs) by investigating promising materials that may exhibit intrinsic magnetic properties. For this purpose, we conducted first-principles calculations to investigate the structural, electronic, magnetic, and transport properties of the H-phase monolayer of the  $4f$  rare-earth dichalcogenide  $\text{EuS}_2$ . Using  $\text{EuS}_2$  as a representative case, the goal is to explore the potential of  $4f$  rare-earth dichalcogenides as promising candidates for generating pronounced charge, spin, and orbital currents. Given the significant role of electron correlation effects in  $f$ -electron systems, a range of Coulomb interaction strengths, characterized by the parameter  $U$ , was considered. The results reveal that  $\text{EuS}_2$  exhibits both a large magnetic moment and a sizable band gap across a broad range of correlation strengths. Notably, an increased  $p$ - $d$ - $f$  hybridization between Eu and S atoms is observed with increasing  $U$ , which exerts a substantial impact on the transport properties of the material.

*The results presented in this chapter have already been published in Zeer et al., Phys. Rev. Mater. 6, 074004 (2022) [182].*

## 5.1 Introduction

TMDs exhibit a rich variety of exotic phenomena driven by the interplay between their crystal symmetry and spin-orbit coupling (SOC) [183]. This interplay gives rise to numerous Hall effects, including the anomalous Hall effect (AHE) [184], valley Hall effect [185, 186], spin Hall effect (SHE) [171, 172], and the more recently discovered orbital Hall effect (OHE) [41, 42, 48, 51, 88, 94, 101, 173, 187–189]. Among these, the SHE and OHE are particularly promising for next-generation spintronic applications. While the SHE has been extensively studied, the OHE remains relatively underexplored, despite its potential to revolutionize information



transfer through the orbital degree of freedom, giving rise to the emerging field of orbitronics.

A particularly promising avenue is the exploration of 2D magnetic materials based on  $4f$ -elements, especially rare-earth dichalcogenides. The strong magnetism, significant spin-orbit coupling, and orbital complexity of  $4f$ -electrons make these materials ideal candidates for studying spin and orbital transport phenomena. Rare-earth dichalcogenides, with their unique electronic and magnetic properties, could serve as a platform for realizing robust orbital currents, opening new possibilities for orbitronic applications.

In this work, we focus on  $4f$  rare-earth metal dichalcogenides, using the H-phase monolayer  $\text{EuS}_2$  as a representative system. By employing first-principles calculations with a Hubbard  $U$  parameter to account for strong electron correlations, we investigate the electronic, magnetic, and transport properties of  $\text{EuS}_2$ . Our results reveal that in the strongly correlated limit, the electronic bands of  $\text{EuS}_2$  exhibit non-trivial topology, leading to sizable and nearly quantized values of the anomalous Hall conductivity (AHC) associated with the  $f$ -states. Interestingly, the SHE in this system closely follows the behavior of the AHE due to the dominant majority character of the occupied  $f$ -states. In contrast, the OHE displays a highly non-trivial dependence on band filling, reaching magnitudes more than six times larger than those of the SHE.

Our detailed analysis of the electronic structure and Hall effects in  $\text{EuS}_2$  provides fundamental insights into the mechanisms governing spin and orbital transport in rare-earth dichalcogenides. These findings suggest that rare-earth dichalcogenides, with their strong magnetic tendencies and robust orbital currents, could occupy a unique niche among 2D magnetic materials, offering significant potential for applications in spintronics and orbitronics.

Specifically, we predict that the H-phase monolayer of  $\text{EuS}_2$  exhibits half-metallic behavior with a large magnetic moment when doped. The electronic structure is highly sensitive to the Hubbard  $U$  parameter, which modulates the hybridization between Eu- $f$  and S- $p$  states. This hybridization plays a critical role in determining the transport properties, leading to pronounced anomalous and orbital Hall effects. These results highlight the potential of rare-earth dichalcogenides as efficient sources of charge, spin, and orbital currents, making them promising candidates for next-generation spintronic and orbitronic devices.

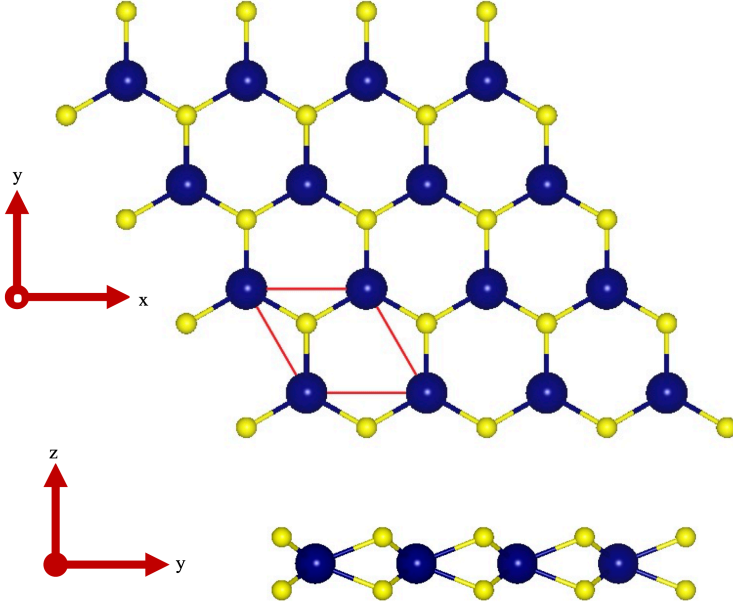
## 5.2 Computational Details

Our first-principles calculations were performed using the film version of full-potential linearized augmented plane wave method [152], as implemented in the Jülich density functional theory (DFT) code FLEUR [190]. We used the Perdew-Burke-Ernzerhof approximation to the exchange-correlation potential [139]. The structure of the monolayer was relaxed so that residual forces are well below 0.001 eV/Å. For self-consistent calculations we used a  $16 \times 16$  Monkhorst-Pack grid in the first Brillouin zone and a plane-wave cutoff of  $4.1 a_0^{-1}$ , where  $a_0$  is the Bohr radius. We set the angular momentum expansion of 10 and 8 for Eu and S atoms, respectively. The muffin-tin radii of Eu and S atoms were set to  $2.8 a_0$  and  $1.94 a_0$ , respectively. Spin-orbit coupling (SOC) was included self-consistently within the second variation scheme for all calculations.

To treat the effect of strongly correlated electrons in the  $4f$ -shell of Eu we applied the GGA+ $U$  method within the self-consistent DFT cycle [191]. The on-site Coulomb interaction strength  $U$  applied to the Eu  $4f$  states was varied from 0 eV to 6.7 eV, and the intra-atomic exchange interaction strength  $J$  was chosen to be 0.7 eV [191].

The optimized structure of H-EuS<sub>2</sub> is shown in Fig. 5.1, along with the definition of the coordinate axes. Initial structural relaxations were performed without including  $U$ , yielding a lattice constant of 4.616 Å and an interplanar distance of 1.1 Å between the Eu and S layers along the  $z$ -axis. Introducing a Coulomb repulsion strength of  $U = 6.7$  eV modified these values to 4.744 Å and 1.08 Å, respectively. All calculations were performed for the latter values of structural constants.

Transport calculations were performed for two values of  $U$ : 2.5 eV and 6.7 eV. The calculations of the anomalous Hall conductivity (AHC), spin Hall conductivity (SHC), and orbital Hall conductivity (OHC) were performed using the expressions discussed below, employing the technique of Wannier interpolation [161, 192]. To achieve this, 36 maximally localized Wannier functions (see Sec. 3.8.1) were constructed from the *ab initio* electronic structure by projecting the Bloch states onto the  $p$ -states of S and the  $f, d$ -states of Eu atoms. This approach ensures the reproduction of the electronic structure within an energy window that includes all states below the Fermi energy and a sufficient number of states above it. Throughout this work, we treat the system as a perpendicularly magnetized ferromagnet. For  $U = 6.7$  eV, the magnetic anisotropy energy was estimated to be on the order of 30 μeV, favoring the out-of-plane direction of magnetization, while the ferromag-

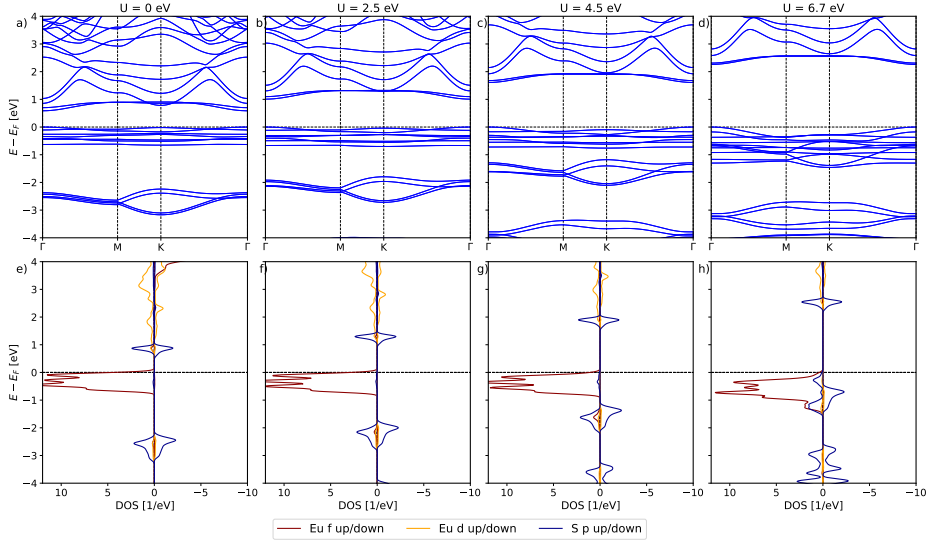


**Fig. 5.1.:** Top and side view of the H-phase monolayer  $\text{EuS}_2$ . The dark blue balls and yellow balls represent Eu and S atoms, respectively. The definition of the axes is shown with arrows.

netic state was found to be 0.3 meV lower in energy than the row-wise antiferromagnetic state. It is important to note that these values may change with variations in the Fermi energy due to doping, which constitutes the primary focus of this study.

### 5.3 Electronic Structure of $\text{EuS}_2$

We begin by discussing the electronic structure of  $\text{EuS}_2$ . Consistent with Hund's first rule, the intra-atomic exchange interaction arranges the half-filled Eu  $4f$  shell into the  $4f^7$  configuration, resulting in a large magnetic moment of  $6.95 \mu_B$ . This value remains robust across varying correlation strengths and is significantly larger than the spin moments observed in other TMDs, as seen in Refs. [20, 24, 25]. In Fig. 5.2, we present the computed band structure (a-d) and density of states (DOS) (e-h) for  $\text{EuS}_2$  at different values of the Hubbard  $U$  parameter:  $U$  of 0 eV (a,e), 2.5 eV (b,f), 4.5 eV (c,g) and 6.7 eV (d,h). The quantitative features of the electronic structure of  $\text{EuS}_2$  are similar irrespective of  $U$ : The highest occupied states are formed by the Eu  $4f$  majority electrons. The position of the minority



**Fig. 5.2.:** Electronic structure of H-phase  $\text{EuS}_2$  monolayer as a function of Coulomb repulsion strength  $U$ . In (a)-(d) the band structures are shown for  $U = 0, 2.5, 4.5$  and  $6.7$  eV, respectively. In (e)-(h) the corresponding evolution of the spin-resolved density of states is shown. In (e)-(h) left and right parts of the figures correspond to the majority and minority spins, respectively, while the density of states of Eu- $f$ , Eu- $d$  and S- $p$  states is shown with dark red, orange and blue thin lines, respectively. While the dominance of the majority Eu- $f$  states at the Fermi level is evident, increasing  $U$  drives the hybridization of Eu- $f$ -states with S- $p$  states. This has drastic influence on the transport properties of the system, as discussed in the main text.

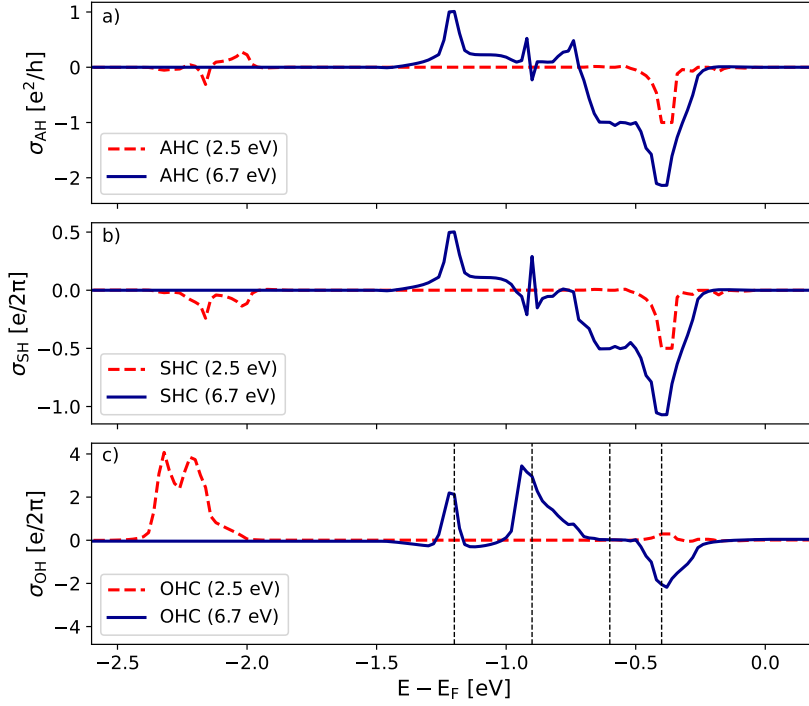
$f$ -states varies from about  $+4$  eV for  $U = 0$  eV to about  $+10$  eV for  $U = 6.7$  eV is shown in Appendix A (Fig. A.1), and we do not discuss these states further. The valence band maximum is positioned at the  $\Gamma$  point. We find  $p$  states of the chalcogen S atoms well separated and lying above and below the majority  $f$  states. Together with Eu  $f$  and  $d$  states the  $p$  states of S form bonding and antibonding states that have an energy separation of about  $3.5$  eV irrespective of the spin-channel. The lowest conduction band is relatively flat and it is formed by the states with primarily S- $p$  character.

The conduction band minimum is also positioned at the  $\Gamma$  point and  $\text{EuS}_2$  monolayer exhibits a direct band gap of  $f$ - $p$  transition. The higher conduction bands following the S- $p$  band are formed by highly dispersing Eu  $6s$  and  $5d$  states. The localized occupied  $4f$  states act as a repulsive scattering potential to the S- $p$  electrons which moves bonding and antibonding S- $p$  states in the majority and minority spin-channel to lower and higher energy, respectively. This introduces a small ex-

change splitting, visible in Fig. 5.2(a)-(d), and a magnetic moment of  $-0.28 \mu_B$  on S-atoms. The values of the orbital moments of Eu and S atoms are practically negligible. Focusing on the  $4f$  states, in the ground state, at  $U = 0$  eV, the majority  $f$ -states are separated from the conduction S  $p$ -states by about 1 eV (direct band gap of 0.59 eV at  $\Gamma$ ), see Fig. 2(a,e). Increasing  $U$  leads to an increase of the  $f$ - $p$  gap without a change of dispersion, and the  $4f$  states move down in energy closer to the occupied S  $p$ -valence states (or, alternatively, since the Fermi energy is fixed by the band edge of the majority  $4f$  states, the S  $p$ -states move up in energy with respect to the  $f$ -states). At  $U = 6.7$  eV this results in a  $p$ - $f$  energy separation of about 3 eV at the Fermi energy (direct band gap of 2.27 eV at  $\Gamma$ ), Fig. 5.2(d,h). This puts EuS<sub>2</sub> among the large band gap TMD semiconductors [193, 194]. On the other hand, increasing  $U$  drives the lower  $p$ -states into the region of majority  $f$ -states. In turn, this drives a strong hybridization between the  $p$ -,  $d$ - and  $f$ -states, which result in the orbital complexity and strong modifications that  $U$  brings to transport properties of the doped system which we discuss below. We conclude that the EuS<sub>2</sub> monolayer is a ferromagnetic semiconductor with a direct band gap, whose value depends on the choice of  $U$ . The studied system can be transformed into a half-metal by substituting Eu with, for example, Gd, forming an Eu<sub>1-x</sub>Gd<sub>x</sub>S<sub>2</sub> layer (see also the next chapter 6).

## 5.4 Anomalous Hall Effect in EuS<sub>2</sub>

Next, we proceed to investigate the AHE in the system. Here, we focus on the intrinsic Berry curvature contribution to the AHC tensor [195]. To compute the Berry curvature from maximally localized Wannier functions (MLWFs), we first construct a tight-binding Hamiltonian based on the MLWFs. Using the Wannier interpolation technique, we efficiently evaluate the  $xy$ -component of the anomalous Hall conductivity (AHC) as a Brillouin zone integral over a dense  $300 \times 300$  mesh of  $k$ -points. This calculation is performed according to the expressions provided in Eqs. 2.24 and 2.25. The results of our AHC calculations are summarized in Fig. 5.3(a) for  $U = 2.5$  eV and  $U = 6.7$  eV. We begin by discussing the case of  $U = 2.5$  eV. The energy dependence of the AHC for this value of  $U$  is shown as a dashed line in Fig. 5.3(a). The AHC is non-zero only in two narrow energy windows: between  $-0.6$  eV and  $0$  eV, corresponding to the upper region of majority  $f$ -states, and between  $-3$  eV and  $-2$  eV, corresponding to the region of occupied S  $p$ -states. The out-of-plane spin and orbital polarization of these states is shown in Figs. 5.4(a,b). We observe that the low lying  $p$ -states are slightly exchange split (by 50–100 meV), perfectly spin-polarized, and they display very strong orbital



**Fig. 5.3.:** Transport properties of monolayer  $\text{EuS}_2$  as a function of band filling. (a) Anomalous Hall conductivity (AHC)  $\sigma_{AH}$ , (b) spin Hall conductivity (SHC)  $\sigma_{SH}$ , and (c) orbital Hall conductivity (OHC)  $\sigma_{OH}$ , computed as a function of band filling in  $\text{EuS}_2$  for  $U = 2.5$  eV (red dashed line) and 6.7 eV (blue solid line). Vertical dashed lines in (c) are guides to the eye marking the position of energy at  $-1.2$ ,  $-0.9$ ,  $-0.6$  and  $-0.4$  eV. The influence of S-Eu hybridization at  $U = 6.7$  eV is clearly visible in the plot: while in the case of weak hybridization at  $U = 2.5$  eV the AHE and SHE in the system are suppressed, the combined effect of interacting S- $p$  and Eu- $f$  states on the overall magnitude and complex behavior of the AHE, SHE and OHE at  $U = 6.7$  eV just below the Fermi energy is drastic.

polarization, especially at the top and bottom of the group. This is consistent with the process of  $p_x \pm p_y$  orbital polarization, confirmed by the DOS analysis (not shown), giving rise to non-trivial Chern numbers and a non-vanishing AHC in the region of these states [196].

By inspecting the region of  $f$ -states in Fig. 5.4(a,b) closer, we observe a formation of relatively large gaps among the groups of  $f$ -states, separated by the combined effect of crystal field and spin-orbit interaction. The topological nature of these gaps is predominantly trivial, *i.e.* the quantized value of the AHC within these gaps is topologically trivial, except for the case of the gap at about  $-0.4$  eV, where

the AHC is quantized to a value of  $-1 \frac{e^2}{h}$  (*i.e.* having a Chern number of  $-1$ ). From the analysis of the orbital character of contributing states it is clear that this is the region where the overall change in the sign of orbital polarization of the states around the gap takes place. This change in the orbital polarization across the gap mediated by the spin-conserving part of spin-orbit interaction can be closely associated with the non-trivial AHC in this region [196]. At this point we would like to comment on the logic behind energy-resolved orbital polarization of the system for  $U = 2.5$  eV. For this correlation strength, the Eu- $f$  states are separated from the  $p$ -states of S atoms. The  $4f$  electrons are localized, and experience a non-spherical potential exhibiting the point group symmetry of the Eu atom embedded into EuS<sub>2</sub> monolayer. Therefore, the development of the orbital moments is a result of a competition between Coulomb interaction, summarized under Hunds second rule, and the crystal field. We notice that our results still resemble Hunds second and third rule, Fig. 5.4(a,b). In the lower half of the  $4f$ -majority states, the orbital moment is opposite to the spin moment, while for the upper half of the  $4f$  states the orbital moment is parallel to the spin moment, which results in a negligible orbital moment when summed over all majority  $4f$  states. For S- $4p$  states the situation is very different since the  $p$ -states are subject to a large band dispersion where crystal field effects dominate. For states along  $\Gamma$ -M the dispersion of the S  $p$ -states is rather small and Hunds third rule is obeyed since the orbital moments of the lower (upper) half of the bands is antiparallel (parallel) to the spin moments, but the strong crystal field associated with the band dispersion along the high-symmetry lines M-K- $\Gamma$  makes Hunds third rule inapplicable. This violation becomes even more prominent for larger values of the correlation strength.

The direct interaction between the  $p$ -states of S and the  $f$ -states of Eu, which occurs when the correlation strength is increased to  $U = 6.7$  eV, has a profound impact on the anomalous Hall effect (AHE). From the analysis of the DOS and the spin polarization of the states shown in Fig. 5.4(c), we observe that minority  $p$ -states are easily identifiable, as they do not hybridize at all with the  $f$ -states of opposite spin. In contrast, the hybridization of the majority  $p$ - and  $f$ -states is significant, as evidenced by strong changes in the position and dispersion of the bands, as well as the increased spread of Eu and S states over an energy range of nearly 1.5 eV in this spin channel. The resulting orbital complexity of hybridized states, visible in Fig. 5.4(c), has an amplifying effect on the overall magnitude of the anomalous Hall conductivity (AHC). In particular, values close to quantization are achieved for the lower bands around  $-1.2$  eV, which are related to a global gap present at that energy (crossed by minority-S states). Additionally, a wide plateau appears around  $-0.6$  eV, corresponding to the nontrivial gap between three upper and five

lower majority bands, separated by an "inactive" band at  $-0.55$  eV. Furthermore, a peak is observed at  $-0.4$  eV.

## 5.5 Spin Hall Effect in $\text{EuS}_2$

We now move on to the study of SHE and OHE in the system. While the SHE has been explicitly studied in group-VI dichalcogenides [171, 172], very recently, Canonico and co-workers investigated the orbital Hall effect in nonmagnetic H-phase 2D TMDs  $\text{MoS}_2$  and  $\text{WS}_2$  [94, 173], addressing especially a question of quantization of the OHC within the electronic energy gaps, and corresponding emergence of the quantum orbital Hall insulating phase. The study of the OHE is a subject of general interest since the magnitude of the OHC can be often dominant over that of the SHC, which suggests systems with large OHE as potential platform for novel concepts in orbitronics [62, 197].

Motivated by these considerations, we explore the possibility that  $\text{EuS}_2$  can host the spin Hall effect (SHE) and orbital Hall effect (OHE). Following the established methodology [42, 198], we calculate the SHC and OHC, which describe the generation of a transverse in-plane current of out-of-plane spin and orbital angular momentum, respectively, in response to an applied electric field. The SHC and OHC are computed using the Kubo formula, as given by Eqs. 2.26 and 2.27. While the results for the SHC are discussed in this section, the findings for the OHC will be presented in the next section.

We first look at the SHE, presenting the results of the calculations in Fig. 5.3(b). For the case of  $U = 2.5$  eV we find that for the  $f$ -states the SHC precisely follows the AHC as the band filling is varied. This is consistent with the picture of the spin angular momentum carried by the anomalous Hall current of fully spin-polarized  $f$ -bands in that energy region. The behavior of the SHC of  $S$   $p$ -states at lower energies is more complicated, however, the consistently negative sign of the SHC can be understood from the sign reversal both in net spin-polarization as well as in the AHC at the top and at the bottom of the  $S$ -states. Remarkably, an almost perfect one-to-one correspondence between the energy dependence of the SHC and AHC still persists for a more complex case of  $U = 6.7$  eV. This can be again understood taking into account pure spin character of  $f$ -states. The presence in that energy region of minority  $S$ -states, see Fig. 5.4(c-d), is the reason for a discrepancy in the shape of the SHC and AHC between  $-1.0$  and  $-0.6$  eV, which can be explained by taking into account a small negative spin Hall signal that the minority  $S$ -states bring with them into that energy region.



## 5.6 Orbital Hall effect in EuS<sub>2</sub>

Finally, we analyze the OHC in the system, which is calculated using Eq. 2.28 and Eq. 2.29. We present the results of our calculations of the OHC in Fig. 5.3(c). We first observe that in the “unhybridized” case of  $U = 2.5$  eV, the OHC of  $f$ -states is minimal, reaching in magnitude the SHC in the region around  $-0.4$  eV, where the AHE and SHE are present as well. Most remarkable is the gigantic OHE carried by the  $p$ -states of S atoms, positioned between  $-3$  and  $-2$  eV, Fig. 5.4(b). The OHC in this region is almost one order of magnitude larger than the SHC or OHC of the  $f$ -states for this value of  $U$ . By inspecting the correlation between the electronic structure, orbital polarization and orbital Hall conductivity, we can identify a region of energy between  $-2.4$  and  $-2.1$  eV as the dominant source of the OHE due to contributions coming from crossings among bands of different orbital character, clearly visible close to the M-point and the  $\Gamma$ -point in Fig. 5.4(b), as well as larger parts of the Brillouin zone where there is strong  $k$ -dependent exchange of orbital angular momentum among the oppositely-polarized bands, which is a direct consequence of  $k$ -dependent hybridization strength.

The fact that  $s$ ,  $p$ -electron systems in lower dimensions such as in thin films or at surfaces can give rise to strong orbital polarization and orbital Hall effect has been one of the most interesting developments in the field of orbitronics. In this context, it is known that even oxygen-based electronic structure can host prominent orbital polarization and current-induced orbital response, see *e.g.* Go *et al.* [199]. The large orbital Hall effect magnetism: The most remarkable thing here is that such  $sp$ -orbital magnetism in principle does not require strong spin-orbit interaction. The drawback of this comes with the fact that low spin-orbit interaction suppresses the anomalous and spin Hall effects, as it is clearly visible in Fig. 5.3(c).

Upon increasing the value of  $U$  to 6.7 eV the complicated  $p - d - f$  hybridization taking place among S and Eu atoms exerts a striking influence on the properties of the OHE of the  $p-f$  group of states below the Fermi energy. This is reflected in a three to four times larger magnitude of the OHC as compared to the SHC observed in the region of majority  $f$ -states, Fig. 5.3(c), and, in contrast to expectations from oversimplifying band-filling arguments applied in the past to nonmagnetic materials [41], a very non-trivial dependence of the OHC on the Fermi energy. At first, it is tempting to attribute the large computed OHC to S  $p$ -states, which promote a strong OHE already for smaller values of  $U$  without any need for aid from the side of  $f$ -orbitals. However, a closer analysis shows that this is not the case.

First, we take a look at the region of energy around  $-1.2$  eV, where the lowest of the peaks in the OHC appears [this energy value is indicated with dashed lines in Fig. 5.3(c) and Fig. 5.4(c-d)]. Here, the large peak in the OHC originates from an anticrossing among two strongly Eu-S hybridized bands [see also the corresponding DOS in Fig. 5.2(h)], positioned at that energy, where an exchange of the orbital polarization is particularly visible around the M-point. A very similar situation is encountered next to and directly at the M-point around  $E_F = -0.9$  eV, where the crucial role of the orbital angular momentum exchange and large values of the OHC displayed by the majority  $p$ -states become very apparent.

The key message of Fig. 5.3 is the illustration of the strong effect that the hybridization between  $p$  and  $f$  states has on transport properties of the system. For weaker correlation, while the AHE and SHE are quantized in a narrow region of energies which signifies the complexity and potential of  $f$ -states in driving non-trivial AHE and SHE, it is only upon infusing the  $f$ -states with the  $p$ -states for stronger correlation that makes the AHE and SHE in this region prominent. On the other hand, while the  $S$ -states exhibit a prominent OHE even at weaker correlation, this strong but uniform in sign effect acquires an energetic structure of so far unreported complexity while retaining its large magnitude in the region of  $f$ -states upon enhanced hybridization. On the other hand, this evolution cannot be understood based on the argument of DOS alone (as is the case with the intrinsic Berry curvature-driven Hall effects discussed in the next section): take a look for instance at the energy of  $0.6$  eV where the DOS varies quite strongly and where the AHC and SHC exhibit a wide plateau while the OHC vanishes.

## 5.7 Orbital Berry Curvature in EuS<sub>2</sub>

The distribution of the orbital Berry curvature of occupied states up to the corresponding Fermi energy, shown in Fig. 5.4(g-h), confirms this picture. From the  $k$ -space distribution of the orbital Berry curvature, we observe that large OHE at these energies originates from sharp circular features centered around  $K'$  points in the Brillouin zone.

Despite a sizable magnitude of the AHC and SHC in the region of energy between  $-0.7$  and  $-0.5$  eV [the value of  $-0.6$  eV is marked with dashed lines in Fig. 5.3(c) and Fig. 5.4(c-d)], the OHC is suppressed there, which is quite unexpected. The reason for this is the absence of the  $k$ -dependent exchange of the orbital polarization among pairs of bands, which appear in this energy window. And although there is a crossing of positively and negatively orbitally polarized bands around  $-0.55$  eV in



the vicinity of the K-point, no hybridization or exchange of orbital angular momentum takes place there. The corresponding orbital Berry curvature for this Fermi energy, shown in Fig. 5.4(f), is overall more than one order of magnitude smaller than for the previously considered cases. This solidifies the  $k$ -dependent orbital polarization exchange as a driving force behind large OHE observed in the studied system at lower energies.

The mechanism behind the large orbital Hall OHC near  $-0.4$  eV is distinct. In this case, the orbital exchange occurs around the M and K points, involving pairs of bands that are well-separated in energy—specifically, the second and third bands when counted from zero energy. This indicates that the exchange between different bands, rather than within the same band (as would occur at  $-0.5$  eV), is responsible for the significant orbital Hall effect in the system.

While this orbital exchange leads to moderate negative values of the orbital Berry curvature over large regions of reciprocal space near the K point, smaller regions around the M point, and particularly M', contribute substantially to the overall large OHC, as shown in Fig. 5.4(e). The negative sign of the OHC, which cannot be achieved by  $p$  or  $f$ -states alone (at lower  $U$  values), highlights the critical role of  $pf$  hybridization in driving the orbital Hall effect in  $\text{EuS}_2$ , and in rare earth dichalcogenides in general.

## 5.8 Conclusion

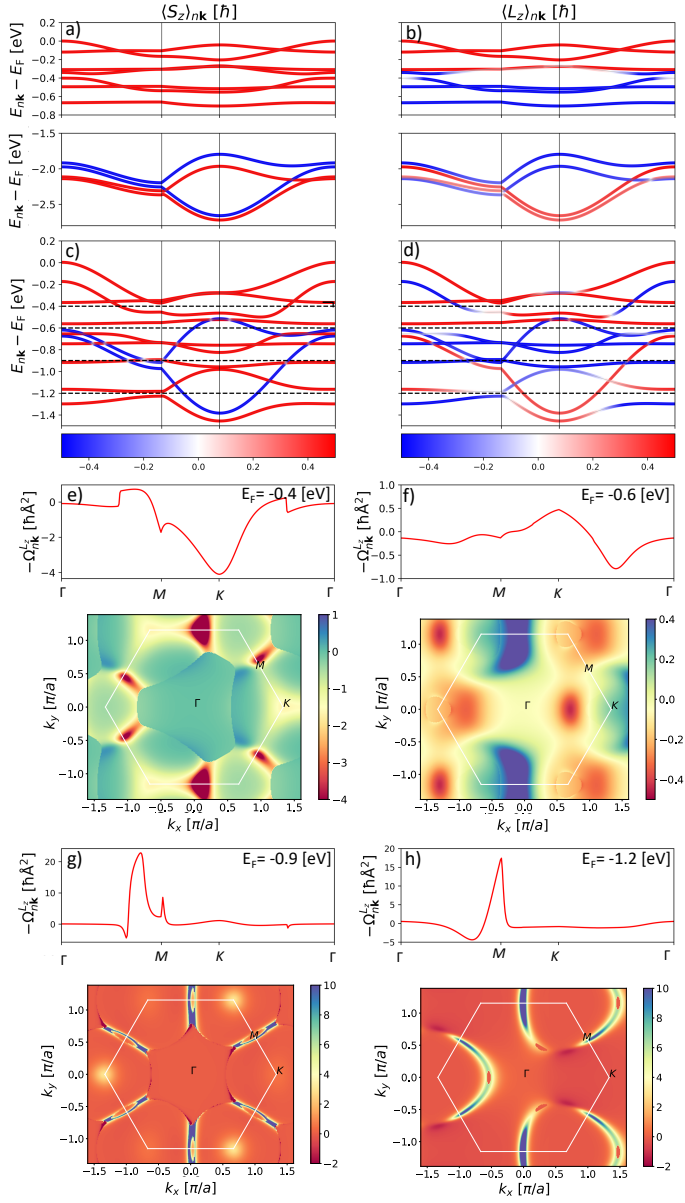
In this chapter, we made first steps in exploring the prospects of rare earth dichalcogenides as a source of transverse topological currents of charge, spin and orbital angular momenta. We chose  $\text{EuS}_2$  as a representative of this class of materials, and computed its structural, electronic, magnetic and transport properties from first principles. One of the strongest points of this type of system, besides the very prominent magnetism that  $f$  electrons exhibit, is the strong spin-orbit interaction mediated by the  $f$  states. This is a very robust and unique feature which is very difficult to achieve with other types of TMDs, where magnetism is much more fragile and the influence of the structure on the nature of exchange-split bands is much stronger.

We also found that this  $f$ -material gives rise to very strong Hall responses and hosts topologically non-trivial bands, despite strong correlation among electrons in the  $f$ -shell. We identified that the reason for this is a strong degree of  $pdf$  hybridization among the Eu and S atoms, enhanced by stronger correlation effects

owing to peculiar electronic structure of EuS<sub>2</sub>. Effectively, S-originated  $p$ -states and Eu  $f$ -states unite their efforts in enhancing each other's anomalous and spin Hall response. As a result of complex orbital hybridization among the latter states, the composite  $p - f$  group of states also exhibits a very large orbital Hall effect, which is dominant over the SHE by about a factor of three. We identify  $k$ -dependent orbital exchange among pairs of hybridized bands as the major source of the large orbital Hall effect. Our results not only promote rare-earth dichalcogenides as an exciting platform for spin and orbital physics, but also advance our understanding of complex orbital transport manifestations of correlated magnetic two-dimensional materials.

Concerning the feasibility of experimental observation of the studied system, we have to remark that several compounds of Eu with chalcogenides are experimentally known in bulk, at interfaces and in thin films *e.g.* based on EuS [30–34], EuSe<sub>2</sub> [35] and EuTe<sub>2</sub> [36]. We thus believe that thin two-dimensional layers of EuS<sub>2</sub> can be naturally produced using for example exfoliation or molecular beam epitaxy. On the other hand, the general conclusions of our work are valid for the entire class of two-dimensional rare-earth dichalcogenides, marking them as an exciting platform for spin and orbital physics, and advancing our understanding of complex orbital transport manifestations of correlated magnetic two-dimensional materials.

**Fig. 5.4.:** Evolution of the out-of-plane spin,  $\langle S_z \rangle_{n\mathbf{k}}$  (left panel: a,c), and orbital polarization,  $\langle L_z \rangle_{n\mathbf{k}}$  (right panel: b,d), of occupied states in EuS<sub>2</sub>. (a) The occupied spin-up  $f$ -states of Eu above  $-0.8$  eV (upper part), and S  $p$ -states in the region between  $-3$  and  $-1.5$  eV (lower part) for the case of  $U = 2.5$  eV. Note that both groups of displayed states are isolated in energy from other states. (c) shows the hybridized group of occupied  $f$  and  $p$ -states for the case of  $U = 6.7$  eV which reside above  $-1.5$  eV. (b,d): Analogous to (a,c). The value scales are indicated by the color bars at the bottom in units of  $\hbar$ . Horizontal dashed lines in (c-d) are guides to the eye marking the position of energy at  $-1.2$ ,  $-0.9$ ,  $-0.6$  and  $-0.4$  eV. (e-f) Distribution of the orbital Berry curvature along high symmetry lines for the indicated position of the Fermi energy, shown together with the distribution of the corresponding orbital Berry curvature in the Brillouin zone.



## Promoting $p$ -based Hall effects by $pdf$ hybridization and strain in rare-earth dichalcogenides

To build upon the intriguing results observed in  $\text{EuS}_2$ , we extend our investigation to a broader range of rare-earth dichalcogenides, aiming to explore further the role of hybridizations and the impact of strain on their transport properties. Specifically, we conduct a comprehensive first-principles study of Hall effects in rare-earth dichalcogenides, focusing on monolayers of the H-phase  $\text{EuX}_2$  and  $\text{GdX}_2$ , where  $\text{X} = \text{S}, \text{Se}, \text{and Te}$ . Our results reveal that all  $\text{EuX}_2$  and  $\text{GdX}_2$  systems exhibit high magnetic moments and wide bandgaps, positioning them as promising candidates for spintronic and orbitronic applications. We find that in  $\text{EuX}_2$ , the  $p$  and  $f$  states hybridize directly below the Fermi energy, while in  $\text{GdX}_2$ , the absence of  $f$  and  $d$  states at the Fermi level leads to a  $p$ -like spin-polarized electronic structure. This structure mediates  $p$ -based magnetotransport in the  $\text{GdX}_2$  systems. Notably, both  $\text{EuX}_2$  and  $\text{GdX}_2$  exhibit significant anomalous, spin, and orbital Hall conductivities. Furthermore, in  $\text{GdX}_2$ , we observe that the strength of electron correlations governs the relative positioning of the  $p$ ,  $d$ , and  $f$  states, as well as their hybridization. This has a profound impact on  $p$ -state polarization and the anomalous Hall effect, although it does not significantly influence the spin and orbital Hall effects. Moreover, we find that the application of strain can significantly modify the electronic structure of the monolayers, resulting in quantized charge, spin and orbital transport in  $\text{GdTe}_2$  via a strain-mediated orbital inversion mechanism taking place at the Fermi energy. Our findings suggest that rare-earth dichalcogenides hold promise as a platform for topological spintronics and orbitronics.

*The results presented in this chapter have already been published in Zeer et al., Phys. Rev. Research 6, 013095 (2024) [200].*

## 6.1 Introduction

In the previous chapter, we theoretically demonstrated that monolayer  $\text{EuS}_2$  exhibits intriguing electronic and magnetic properties, alongside large anomalous spin and orbital Hall effects [182]. These findings have motivated further investigation into the electronic structure and transport behavior of two-dimensional  $4f$  rare-earth dichalcogenides (REDs). In this chapter, we specifically focus on the physics of H-phase monolayers of  $\text{EuSe}_2$ ,  $\text{EuTe}_2$ ,  $\text{GdS}_2$ ,  $\text{GdSe}_2$ , and  $\text{GdTe}_2$ , utilizing first-principles calculations. The primary objective is to explore the potential of these materials as sources of significant charge, spin, and orbital currents, while also considering the impact of correlation effects.

Our results reveal that all of the considered REDs exhibit large magnetic moments and band gaps across a wide range of correlation strengths. For Eu-based REDs, we find that their properties are primarily governed by the direct interaction between  $p$ - and  $f$ -states below the Fermi energy. In contrast, for Gd-based REDs, the  $p$ -states, although well-separated from the  $f$ - and  $d$ -states, are spin-polarized by these states and play a crucial role in determining the electronic structure and transport properties. We demonstrate that significant charge, spin, and orbital responses occur in these materials when an electric field is applied, highlighting the tunability of their transport and electronic properties through correlations. Additionally, we identify strain as a powerful tool for inducing a topological Hall response in  $\text{GdTe}_2$ , which can transition into a Chern and quantum spin Hall insulator via an orbital Hall insulator phase at the Fermi energy. This demonstrates that the complex  $pdf$  hybridization in REDs can lead to purely  $p$ -based topological transport in time-reversal symmetry-broken  $f$ -based 2D phases. Our findings suggest that REDs hold significant potential for spintronics and magnetotransport applications.

*For convenience, in the following discussion, we will refer to the monolayers of  $\text{EuS}_2$ ,  $\text{EuSe}_2$ , and  $\text{EuTe}_2$  collectively as  $\text{EuX}_2$  ( $X = \text{S}, \text{Se}, \text{Te}$ ), and the monolayers of  $\text{GdS}_2$ ,  $\text{GdSe}_2$ , and  $\text{GdTe}_2$  as  $\text{GdX}_2$  ( $X = \text{S}, \text{Se}, \text{Te}$ ).*

## 6.2 Computational Details

We conducted our first-principles calculations using the density functional theory (DFT) code FLEUR [154], which implements the full-potential linearized augmented plane wave method [152]. The Perdew-Burke-Ernzerhof approximation [139] was used to account for exchange and correlation effects. The monolayer systems were

relaxed to obtain their optimal structures, both with and without the inclusion of the Coulomb interaction strength  $U$ , as shown in Table 7.1. The SOC effect was considered using the second-variation scheme in all calculations. We performed self-consistent calculations using a  $16 \times 16 \times 1$  Monkhorst-Pack grid in the first Brillouin zone. We found that all compounds favor ferromagnetic ground state by 13.3, 8.2, and 5.7 meV for  $\text{GdS}_2$ ,  $\text{GdSe}_2$ , and  $\text{GdTe}_2$ , respectively, and by 0.3, 0.62, and 2.5 meV for  $\text{EuS}_2$ ,  $\text{EuSe}_2$ , and  $\text{EuTe}_2$ , respectively. To construct maximally localized Wannier functions (MLWFs) from the Bloch wave functions obtained from the self-consistent DFT calculation, we utilized the Wannier90 package [161]. For the  $\text{EuX}_2$  systems, the MLWFs were constructed from the  $f$  and  $d$  orbitals of the Eu atom, and the  $p$  orbitals of the X atom. For the  $\text{GdX}_2$  systems, the MLWFs were constructed from the  $d$  orbitals of Gd and the  $p$  orbitals of the X atom, as the  $f$  state of Gd is located far away from the Fermi energy (see below). The maximum frozen window was set to be 2 eV higher than the Fermi energy for each system. A mesh of  $8 \times 8 \times 1$  k-points was employed to obtain 36 and 22 MLWFs for Eu and Gd systems, respectively. The matrix elements of spin and orbital angular momentum operators were first evaluated in the Bloch basis and then transformed into the MLWF basis.

To account for the effect of electronic correlations, we utilized the GGA+ $U$  method within the self-consistent DFT iteration. The Hubbard  $U$  correction was employed to address the strong on-site Coulomb repulsion between the electrons in the  $4f$ -shell of Eu and Gd. We selected the on-site Coulomb interaction strength  $U$  to be 0, 2.5, and 6.7 eV [191, 201], and the intra-atomic exchange interaction strength  $J$  to be 0.7 eV [191, 201, 202]. It is worth noting that a more accurate description of our systems could be achieved by using the widely accepted value of 6.7 eV for Eu/Gd. Nevertheless, we also performed calculations with a smaller value of  $U=2.5$  eV to gain a general understanding of the interplay between bonding and correlations in these systems.

## 6.3 Electronic Structure of the $\text{EuX}_2$ Monolayers

The computed structural and magnetic properties of  $\text{EuX}_2$  ( $X = \text{S, Se, Te}$ ) are presented in Table 7.1. We find that the local Eu magnetic moment in these systems is around  $6.95 \mu_B$ , being quite independent of the correlation strength and chemical composition. The local magnetic moment of chalcogen atoms was found to be small. When going from S to Te, we notice an expansion of the lattice, and an increase in the distance between the planes of Eu and X atoms, Table 7.1, which can be



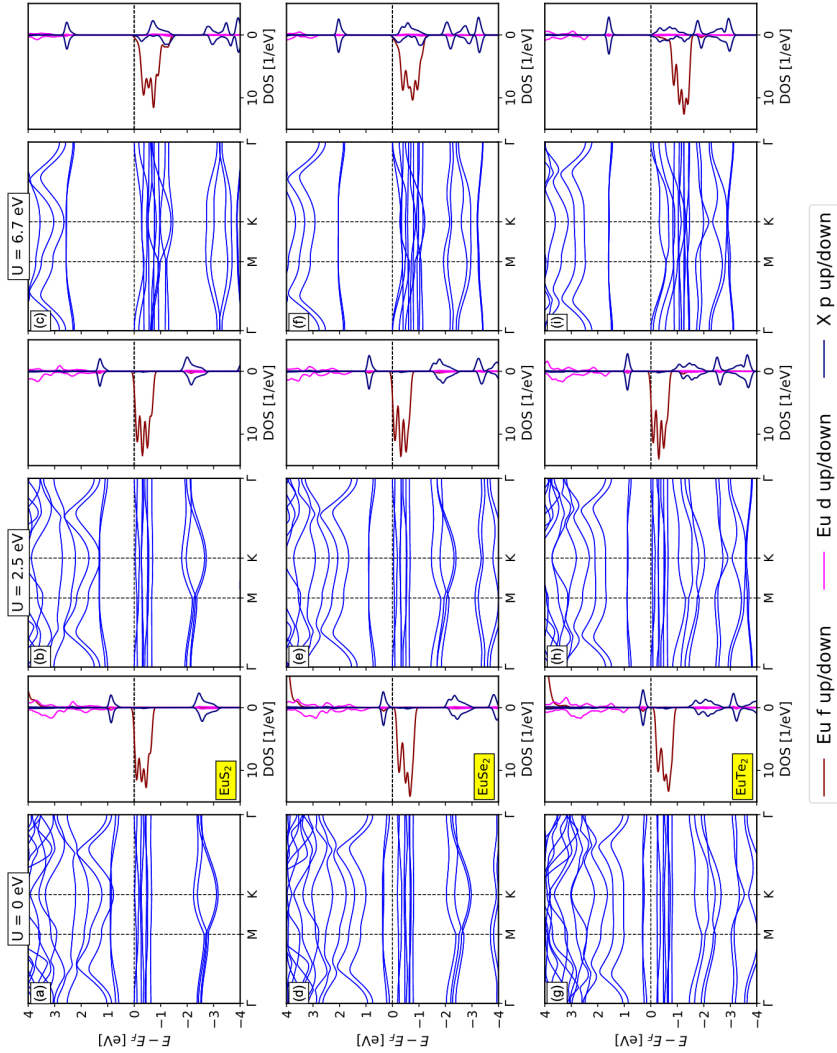
**Tab. 6.1.:** Relaxed atomic positions and magnetic properties of  $\text{Eu}/\text{GdX}_2$  compounds. Lattice constant ( $a$ ), distance between  $M$  and  $X$  atomic planes  $\Delta(M-X)$ , magnetic moments of  $M$  and  $X$  atoms, and band gap values are shown. The value of  $U$  is taken to be 6.7 eV.

| Materials         | Lattice Constant (Å) | $\Delta(M-X)$ (Å) | Magnetic Moment ( $M$ ) ( $\mu_B$ ) | Magnetic Moment ( $X$ ) ( $\mu_B$ ) | Band Gap (eV) |
|-------------------|----------------------|-------------------|-------------------------------------|-------------------------------------|---------------|
| $\text{EuS}_2$    | 4.616                | 1.103             | 6.82                                | -0.010                              | 0.59          |
| $\text{EuS}_2+U$  | 4.744                | 1.080             | 6.95                                | -0.028                              | 2.27          |
| $\text{EuSe}_2$   | 4.824                | 1.209             | 6.81                                | -0.005                              | 0.31          |
| $\text{EuSe}_2+U$ | 4.907                | 1.237             | 6.95                                | -0.024                              | 1.81          |
| $\text{EuTe}_2$   | 5.036                | 1.440             | 6.83                                | -0.009                              | 0.39          |
| $\text{EuTe}_2+U$ | 5.177                | 1.417             | 6.91                                | -0.026                              | 1.45          |
| $\text{GdS}_2$    | 4.138                | 1.388             | 6.86                                | -0.35                               | 0.82          |
| $\text{GdS}_2+U$  | 4.192                | 1.400             | 7.05                                | +0.32                               | 0.74          |
| $\text{GdSe}_2$   | 4.301                | 1.492             | 6.83                                | -0.37                               | 0.62          |
| $\text{GdSe}_2+U$ | 4.265                | 1.521             | 7.05                                | +0.33                               | 0.53          |
| $\text{GdTe}_2$   | 4.571                | 1.654             | 6.84                                | -0.32                               | 0.37          |
| $\text{GdTe}_2+U$ | 4.550                | 1.693             | 7.11                                | +0.27                               | 0.34          |

explained by an increasing spread of the chalcogen  $p$ -states with increasing the atomic number.

The computed band structures of  $\text{EuX}_2$  ( $X = \text{S}, \text{Se}, \text{Te}$ ) are shown in Fig. 6.1 (a)-(i), exhibiting a band gap at the Fermi energy for all systems. For  $U = 0$  eV the valence band maximum originates from the majority  $4f^\uparrow$  states of the Eu atom, while the conduction band bottom consists of the  $p$  states of chalcogenide atoms. Increasing the value of  $U$  from 0 to 6.7 eV leads to an increase in the band gap without a radical change in dispersion. For instance, the band gap increased from 0.59, 0.31 and 0.39 to 2.27, 1.81 and 1.45 eV for  $\text{EuS}_2$ ,  $\text{EuSe}_2$  and  $\text{EuTe}_2$  respectively, while the valence band maximum of  $\text{EuTe}_2$  acquires predominantly Te- $p$  character. Additionally, increasing  $U$  brings the majority of  $4f^\uparrow$ -states down in energy which results in strong direct hybridization between the  $4f^\uparrow$  Eu and the X  $p$  states. This hybridization has a strong impact on the transport properties, as discussed further. Overall, the basic ingredients of the electronic structure of  $\text{EuX}_2$  have been analyzed on the case of  $\text{EuS}_2$  in the section 5.3.

When comparing the three compounds with each other, several key differences can be observed. First of all, the relative distance between the valence  $f$ - and lower  $p$ -states decreases with increasing the atomic number when going from S to Te via Se. And while with increasing  $U$  the  $p$ - and  $f$ -states are ultimately brought into the



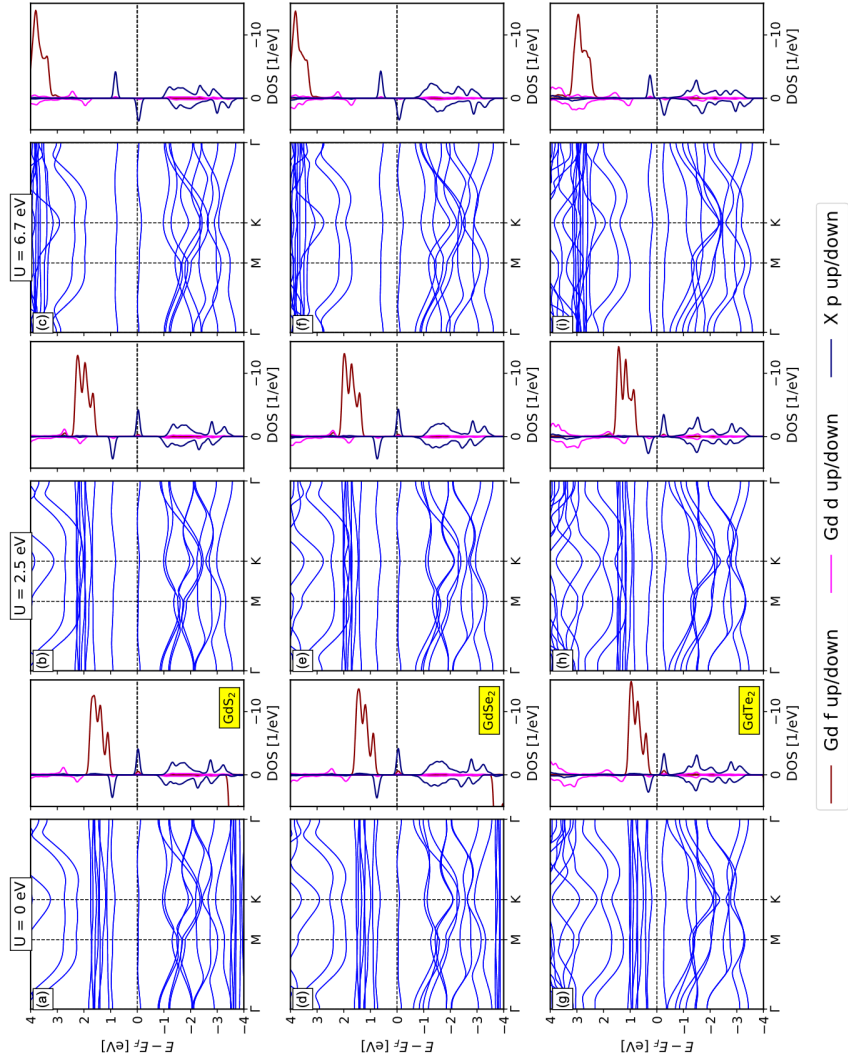
**Fig. 6.1.:** Electronic structure of the H-phase  $\text{EuX}_2$  monolayers as a function of Coulomb repulsion strength  $U$  of 0 (left column), 2.5 (middle column) and 6.7 eV (right column), as shown in (a)-(c), (d)-(f), and (g)-(i) for  $\text{EuS}_2$ ,  $\text{EuSe}_2$ , and  $\text{EuTe}_2$ , respectively. Left and right parts of each figure correspond to the band structure and spin-resolved density of states (DOS). The DOS of Eu- $f$ , Eu- $d$ , and X- $p$  states is shown in dark red, pink, and blue, respectively.

same region of energy, in case of  $\text{EuTe}_2$  the  $p$ -states “overshoot” the  $f$ -states, get promoted to the Fermi energy, and push Eu  $f$ -bands down in energy significantly. This can be explained by the overall reduction in repulsive interaction between

the lowest conduction and highest valence  $p$ -bands. In turn, this results in the reduction of the band gap when going from EuS<sub>2</sub> to EuTe<sub>2</sub>, see Table 7.1. Secondly, the small exchange splitting of chalcogen  $p$ -states, visible in the DOS, increases with increasing the atomic number. Since the  $p$ -magnetism in our systems arises due to  $pdf$  hybridization, larger spread of  $p$ -orbitals of Te as compared to that of S  $p$ -bands leads to stronger hybridization and correspondingly larger spin splitting of  $p$ -states. The variation in spin splitting has direct impact on the transport properties of  $p$ -states, as discussed further.

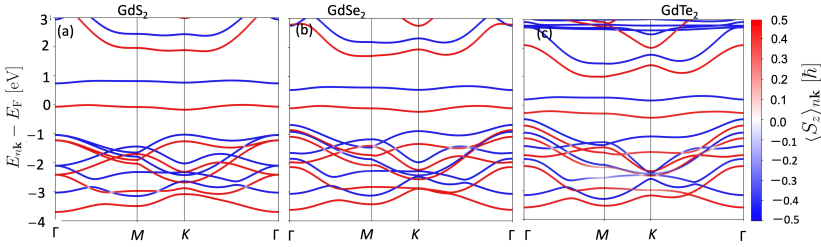
## 6.4 Electronic Structure of the GdX<sub>2</sub> Monolayers

We next discuss the electronic structure of GdX<sub>2</sub> compounds, shown in Fig. 6.2. We find that the maximum of the valence band of GdX<sub>2</sub> is located along the  $\Gamma$  – K path, while the minimum of the conduction band is positioned at the K point, resulting in an indirect, small band gap formed by the occupied and unoccupied  $p_z$ -states of X atom with opposite spin. Thus, all GdX<sub>2</sub> compounds exhibit a typical bipolar magnetic semiconducting feature as shown in Fig. 6.3, where the highest valence band and the lowest conduction band have an opposite spin orientation [203]. Such a feature is believed to offer significant advantages for the development of spintronics nanodevices [204]. We also investigate the effect of on-site Coulomb interaction  $U$  on the band gap of GdX<sub>2</sub>. At  $U = 0$  eV, the magnitude of the indirect band gaps of GdX<sub>2</sub> systems is 0.82, 0.62, and 0.37 eV for GdS<sub>2</sub>, GdSe<sub>2</sub>, and GdTe<sub>2</sub>, respectively. We observe that with increasing  $U$  the band gap changes only slightly, see Table 7.1. In contrast to Eu-based REDs considered above, the Gd majority (fully occupied) and minority (empty)  $f$ -states are positioned quite far away from the Fermi energy, and thus the properties around the Fermi energy are determined by the X  $p$ -states. The hybridization between these  $p$ -states and the  $d, f$ -states of Gd is evident from the DOS in the energy range from  $-4$  eV to  $-1$  eV. This hybridization is particularly strong near the Fermi level, where the  $p$ -orbitals of X atoms are strongly coupled with the  $d$ -orbitals of Gd. and it is largely responsible for the observed transport properties, as we shall see below. Key for magnetotransport properties of GdX<sub>2</sub> is the induced spin-splitting of the  $p$ -states. Around the Fermi energy, a sizeable spin splitting and spin-dependent occupation of the  $p_z$  states is largely responsible for an overall much larger moment of the X atom in GdX<sub>2</sub> as compared to the case of EuX<sub>2</sub>, see Table 7.1. Let us analyze in detail the evolution of the electronic structure and  $p$ -magnetism as we increase the effect of correlations. By increasing the value of  $U$  from 0 to 2.5 and then further to 6.7 eV, we observe a clear trend in the hybridization between the Gd- $f, d$  states and X- $p$  states, which is reflected



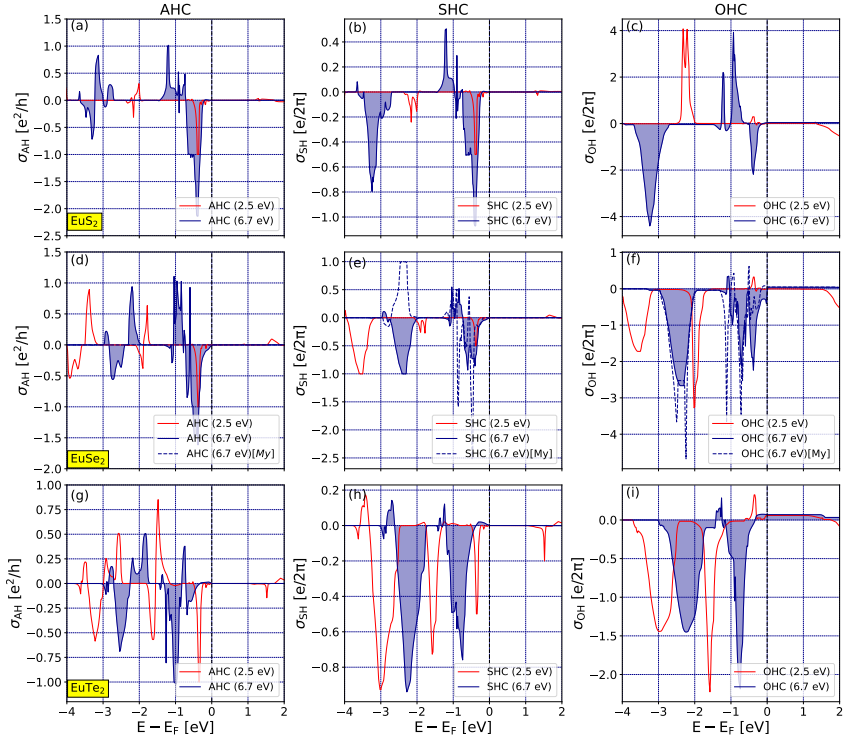
**Fig. 6.2.:** Electronic structure of the H-phase  $\text{GdX}_2$  monolayers as a function of Coulomb repulsion strength  $U$  of 0 (left column), 2.5 (middle column) and 6.7 eV (right column), as shown in (a)-(c), (d)-(f), and (g)-(i) for  $\text{GdS}_2$ ,  $\text{GdSe}_2$ , and  $\text{GdTe}_2$ , respectively. Left and right parts of each figure correspond to the band structure and spin-resolved DOS. The DOS of Gd- $f$ , Gd- $d$ , and X- $p$  states is shown in dark red, pink, and blue, respectively. Upon increasing  $U$  the minority/majority Gd- $f$  states shift up/down in energy and the minority/majority X  $p$ -states shift down/up. The  $pd$  hybridization between the Gd and X atoms below the Fermi energy transforms the separate group of  $p$ -states into a transport-active group of states.

in their energetic position. Specifically, we find that the majority( $\uparrow$ )/minority( $\downarrow$ ) DOS of the Gd- $f$  states moves down/up as we increase the  $U$ , and the  $p$ -states follow the same trend, Fig. 6.2(c,f,i). For smaller values of  $U$ , the  $p$ -states are spin-polarized with a moment opposite to that of Gd atom. This is the consequence of the  $p$ -states being positioned in the gap between strongly exchange-split  $f$ -states, which is best understood by looking at the  $p_z$ -states positioned right at the Fermi energy: starting from initially spin-degenerate  $p$ -states here, we realize that as a result of hybridization of majority  $p^\uparrow$ -states with the low  $f^\uparrow$ -states the former will move up in energy, while the minority  $p^\downarrow$ -states will move down in energy as a result of hybridization with high-lying  $f^\downarrow$ -states. This results in the occupation of the  $p^\downarrow$ -states at the Fermi energy, which also explains the opposite  $p$ -moment of X atoms. Moreover, upon increasing  $U$ , the  $f^\downarrow$ -states move higher in energy, and the



**Fig. 6.3.:** Illustration of the bipolar magnetic semiconducting nature of GdX<sub>2</sub> monolayers, where the highest valence band and the lowest conduction band display opposite spin orientations. Panels (a), (b), and (c) correspond to GdS<sub>2</sub>, GdSe<sub>2</sub>, and GdTe<sub>2</sub>, respectively, highlighting the unique spin-polarized electronic structures of these materials.

energetic position of the  $p$  states starts to be effected by the unoccupied exchange-split Gd  $d$ -states, lying higher than the  $p$ -states. This results in an increased  $pd$ -hybridization as visible also from the partial DOS in Fig. 6.2. Here, as a result of this hybridization,  $d^\uparrow$ -states push  $p^\uparrow$ -states much lower in energy than the  $d^\downarrow$ -states push down  $p^\downarrow$ -states, owing to the fact that  $d^\downarrow$  are situated further from the  $p$ -states than  $d^\uparrow$ -states. As a result, the polarization of  $p$ -states follows that of the Gd  $d$ -states. This promotes the occupation of  $p^\uparrow$  states and the corresponding X-moment switches its sign as  $U$  is increased. This behavior is reflected by deeper lying occupied  $p$ -states, as can be seen in the DOS. Our calculations thus underline a crucial influence of the subtle details of  $pdf$ -hybridization for the  $p$ -magnetism of GdX<sub>2</sub> compounds. While it is beyond the scope of the current work, a relevant challenge for future studies would be a detailed microscopic understanding of the effect that the  $pdf$ -hybridization has on orbitally- and atomically-resolved exchange interactions and their behavior with the correlation strength.



**Fig. 6.4.:** Transport properties of  $\text{EuX}_2$  as a function of band filling. Each row corresponds to a different system, with the first column representing the anomalous Hall conductivity (AHC,  $\sigma_{\text{AH}}$ ), the second column the spin Hall conductivity (SHC,  $\sigma_{\text{SH}}$ ), and the third column the orbital Hall conductivity (OHC,  $\sigma_{\text{OH}}$ ), computed for two values of  $U$ : 2.5 eV (red dashed line) and 6.7 eV (blue solid line). For  $\text{EuSe}_2$  blue dashed line shows the values for the case of in-plane magnetization along  $y$  (for  $U = 6.7$  eV).

## 6.5 Transport Properties of $\text{EuX}_2$

We investigate the transport properties of  $\text{EuX}_2$  and  $\text{GdX}_2$  compounds by computing the intrinsic Berry curvature contribution to the AHC, SHC, and OHC from the MLWF description. To do so, we construct a MLWF Hamiltonian and use the Wannier interpolation technique to calculate the  $xy$ -component of the AHC, SHC, and OHC as a Brillouin zone integral on a  $300 \times 300$  mesh of  $k$ -points. For the AHC, SHC, and OHC, we use the following equations: 2.24, 2.26, and 2.28. We present the results of our conductivity calculations in Fig. 6.4. We divide the figure into three columns: the first column presents AHC values, the second – SHC, and the third column – the values of the OHC. Within each panel, the band filling de-

pendence is shown for two values of  $U$ : 2.5 eV and 6.7 eV. Overall, all compounds exhibit significant values of the conductivities, and below we present a comparative analysis of the transport characteristics in relation to electronic structure features for each material.

For the transport properties of EuX<sub>2</sub> the  $p$ - $f$ -hybridization plays a crucial role, as discussed in previous work [182]. In both EuS<sub>2</sub> and EuSe<sub>2</sub> the increase in  $U$  drives the shift of the occupied  $p$ -states up in energy thus fostering a direct overlap and hybridization between  $p$ - and  $f$ -states right below the Fermi energy. This leads to a drastic enhancement of AHC, SHC and OHE in the region of energies above  $-1.5$  eV upon changing  $U$  from 2.5 to 6.7 eV, Fig. 6.4(a-f). Below  $-2$  eV the transport in EuS<sub>2</sub> and EuSe<sub>2</sub> is dominated by the spin-polarized  $p$ -states of X atoms, which carry significant AHC, SHC and OHC, and which move up in energy with increasing  $U$ .

In case of EuSe<sub>2</sub> for  $U = 6.7$  eV, Fig. 6.4(d-f), at the energy of  $-2.3$  eV the  $p_x, p_y$ -states form a global gap which can be characterized with a zero Chern number (*i.e.* vanishing AHC), but which exhibits quantized values of the SHC and OHC. This manifests the emergence of a combined time-reversal broken spin Hall insulating and orbital Hall insulating phase. Remarkably, the character of this topological gap can be tuned by bringing the magnetization to lie in the plane of the monolayer: in this case the AHC vanishes from symmetry, with the quantized value of the OHC remaining intact, but with the spin Chern number changing its sign, Fig. 6.4(d-f). At the same time the overall band filling dependence of SHC and OHC in EuSe<sub>2</sub> changes drastically upon changing the magnetization direction. Notably, among the compounds studied, EuSe<sub>2</sub> is the only system that exhibits a magnetization-direction tunable sharp peak in OHC precisely at the Fermi energy.

The case of EuTe<sub>2</sub>, Fig. 6.4(g-i), stands out quite distinctly when compared to EuS(Se)<sub>2</sub>. Indeed, upon changing the magnitude of  $U$  from 2.5 to 6.7 eV, the change in the band-filling dependence of the conductivities, at first sight, can be understood as a simple shift in energy. Upon inspection of the correlation between the peaks in the conductivities with evolution of the DOS discussed above, we realize that, as in the case of EuS(Se)<sub>2</sub>, at smaller value of  $U$  the overall behavior can be decomposed into two separate regimes: below  $-1$  eV, determined by  $p$ -states, and between  $-1$  eV and the Fermi energy, governed by the  $f$ -states. However, when the  $U$  is increased to 6.7 eV, the highest occupied  $p$ -states overshoot the  $f$ -states in EuTe<sub>2</sub>, with the latter moving down in energy into the region of  $[-1.5; -1.0]$  eV, keeping the direct hybridization with the  $p$ -states to a minimum.

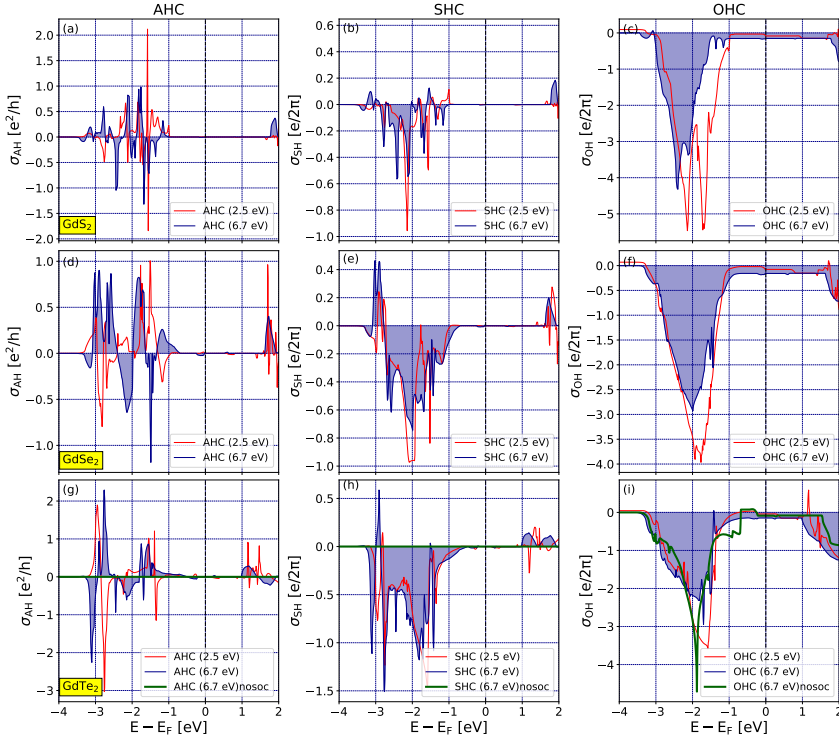
As a result of this, qualitatively, the contribution of the  $p$ -states simply shifts in energy with increasing  $U$ , and the peak due to  $f$ -states shifts down in energy, getting modified in the process by the interaction with a small fraction of  $p$ -states remaining below  $-1$  eV. For example, the  $f$ -peak in the AHC at  $-0.3$  eV [ $U = 2.5$  eV, Fig. 6.4(g)] moves to  $-1.25$  eV, the corresponding negative peak in the SHC [ $U = 2.5$  eV, Fig. 6.4(h)] is recognizable as a small positive peak at  $-1.25$  eV for  $U = 6.7$  eV, and a small positive OHC  $f$ -peak at  $-0.3$  eV [ $U = 2.5$  eV, Fig. 6.4(i)] moves to  $-1.25$  eV while keeping sign and magnitude. Overall, the  $p$ -contributions to transport remain by far dominant in magnitude.

## 6.6 Transport Properties of $\text{GdX}_2$

The transport properties of  $\text{GdX}_2$ , presented in Fig. 6.5(a-i), are distinctly different from  $\text{EuX}_2$ , but qualitatively similar among each other. Let us first look at the AHC, Fig. 6.5(a, d and g). A prominent AHC signal is observed in a wide range below the Fermi energy, and it can be associated with the  $p$ -states of X, considering that the occupied majority  $f$ -states are positioned very deep in energy, as discussed above. Not only for the AHC, but also for the SHC and OHC this range of “Hall-active” energies corresponds to the width of the  $p$ -band, with an exclusion of the  $p_z$ -states, spin-split across the Fermi energy, which do not contribute to the Hall effects. The fine structure of the AHC within the  $p$ -band differs among the members of the  $\text{GdX}_2$  family, however, the influence of  $U$  on the AHC can be understood from the changes in the spin-resolved character of the  $p$ -states discussed above. Indeed, increasing the  $U$  from 2.5 to 6.7 eV interchanges the position of  $p^\uparrow$  and  $p^\downarrow$  states, which results in the reversal of the  $p$  spin-polarization. In a simple picture, this should result in a change in the sign of the AHC at a given energy – the qualitative validity of this prediction can be indeed checked in Fig. 6.5(a, d and g).

The effect of  $U$  is much weaker on the SHC, Fig. 6.5(b, e and h). Unlike for the case of  $\text{EuX}_2$ , the increase in  $U$  does not result in significant changes of energy positions of the  $p$ -states below the Fermi energy, while the slight rearrangements of the states within the  $p$ -band influence the fine structure of the SHC, with an overall SHC magnitude increasing and the distribution becoming more uniform with increasing the atomic number of the X atom. This may be attributed to two factors: (i) a slightly increasing  $p$ -band width and correspondingly stronger spin-orbit splittings of the states with increasing the atomic number, which drive a stronger SHC; (ii) the two separate groups of  $p$ -states which separately provide contributions to the SHC and OHC at different energies of  $\text{EuX}_2$  are coming together within the same energy region for  $\text{GdX}_2$ , contributing by a wide plateau of the conductivities. Unlike the





**Fig. 6.5.:** Transport properties of  $\text{GdX}_2$  as a function of band filling. Each row corresponds to a different system, with the first column representing the anomalous Hall conductivity (AHC,  $\sigma_{\text{AH}}$ ), the second column the spin Hall conductivity (SHC,  $\sigma_{\text{SH}}$ ), and the third column the orbital Hall conductivity (OHC,  $\sigma_{\text{OH}}$ ), computed for two values of  $U$ : 2.5 eV (red dashed line) and 6.7 eV (blue solid line). For the case of  $\text{GdTe}_2$  thick green lines show the values computed without spin-orbit coupling (for  $U = 6.7$  eV).

AHC, the overall sign of the SHC stays predominantly negative as a function of band filling and  $U$ .

This is even more pronounced in the case of OHC for all  $\text{GdX}_2$  materials: within the considered range of energies we predict only a negative sign of the OHC, Fig. 6.5(c, f and i). In comparison to the SHC behavior, and even more so when compared to the AHC, the shape of the OHC dependence on band filling seems almost universal. One reason for such universality is certainly the fact that the position of the  $p$ -states and their orbital composition remains frozen when varying the X element – the effect is further enhanced by the fact that the primary OHC roots in the electronic structure of the  $p$ -states already without spin-orbit interaction, which demolishes the effect

of atomic-number dependent band splittings via SOC, on the orbital Hall effect. To demonstrate this clearly, for the case of  $\text{GdTe}_2$ , we compute the conductivities without spin-orbit, finding that while the AHC and SHC expectedly turn to zero, the OHC remains influenced very little by such a drastic change in the electronic structure. Of course,  $X$ -dependent variation in the OHC can be also observed – note for example the double peak structure of the OHC for  $\text{GdS}_2$  and its evolution with  $U$  which can be attributed to the energy shift of the anticrossing at  $K$  from  $-2$  eV [Fig. 6.2(b)] to  $-2.5$  eV [Fig. 6.2(c)] – but this occurs on a much smaller scale than that of the characteristic peak OHC magnitude of the order of  $2$  to  $4 e^2/h$ .

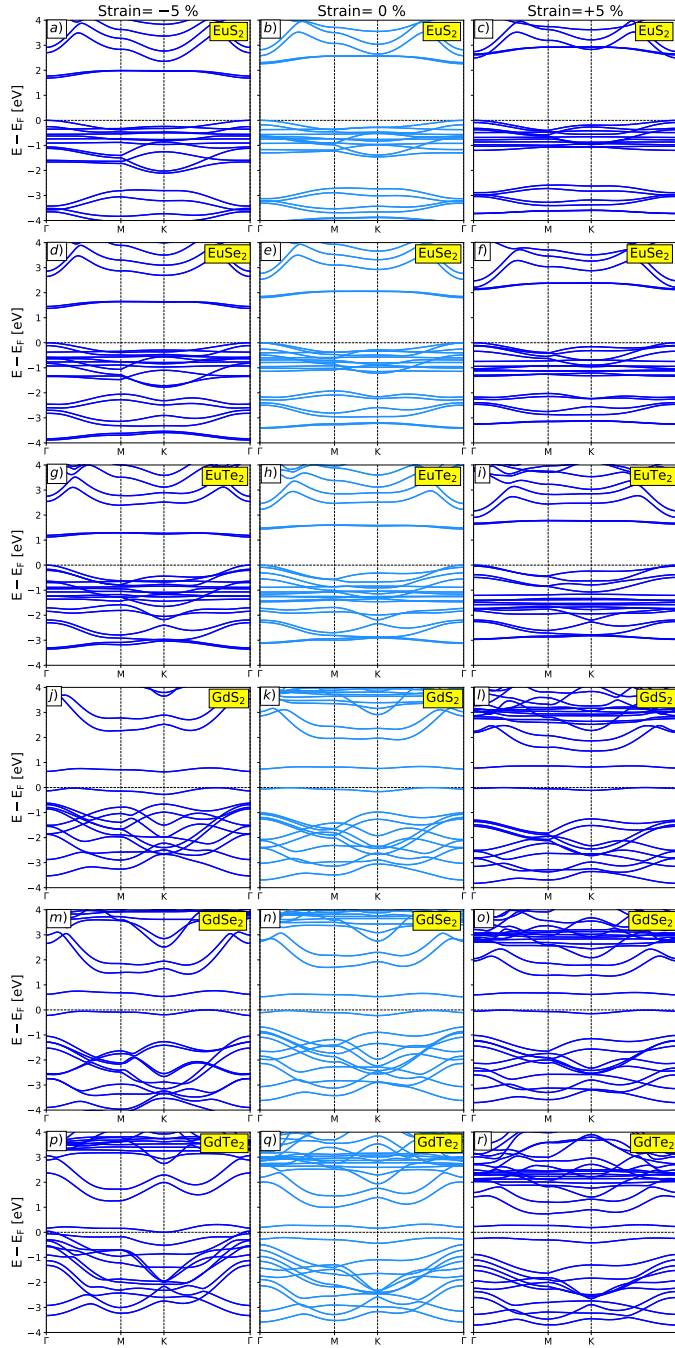
## 6.7 Tuning Electronic Structure by Strain

Next, in our calculations we apply a bi-axial tensile and compressive strain of  $\pm 5\%$ , relax the atomic positions and compute the electronic structure of  $\text{EuX}_2$  and  $\text{GdX}_2$ , presenting the evolution of band structures with strain along  $K$ - $\Gamma$  in Fig. 6.6. We observe that despite the influence of strain on the exact values of the magnetic anisotropy energy, the magnetic moment for all systems remains basically unaffected in size and orientation.

In  $\text{EuX}_2$ , Fig. 6.6(a-i), strain exerts a strong influence on the relative energetic positions of the  $p$ - and  $f$ -states: with increasing tensile strain the  $p$ -states generally become less dispersive and move higher in energy, while at the same time the occupied  $f$ -states move down in energy. This reduces the hybridization and overlap of lowest valence  $p$ -states with the  $f$ -bands, has a strong impact on the dominant orbital character of the states around the Fermi energy as the lattice is expanded, and increases the bandgap from 2.27, 1.81 and 1.45 eV at equilibrium to 2.52, 2.13 and 1.65 eV at  $+5\%$  tensile strain for  $\text{EuS}_2$ ,  $\text{EuSe}_2$  and  $\text{EuTe}_2$ , respectively. Such behavior might be desirable for optoelectronic applications where spin- and orbitally-resolved details of coupling of optical pulse to electronic states may be tuned by strain.

In case of  $\text{GdX}_2$ , Fig. 6.6(j-r), the key features of the electronic structure remain intact in the considered strain range: for the majority of the cases, the central group of half-filled exchange-split  $p_z$ -states positioned right at the Fermi energy remains well-separated from the unoccupied  $d$ -states and occupied  $p_{x,y}$ -bands. This is despite the fact that with increasing tensile strain the  $d$ -states approach the  $p_z$ -group, while the  $p_{x,y}$ -bands come close to the Fermi energy under compressive strain. This means that the profoundly metallic behavior with tunable orbital character at the Fermi energy may be achieved  $\text{GdS}_2$  and  $\text{GdSe}_2$  under very large strain. It

is only for the case of  $\text{GdTe}_2$  that at  $-5\%$  strain the  $p_{x,y}$ -states reach the Fermi energy which leads to an intricate hybridization with the  $p_z$ -states and significant modifications in the electronic structure. We consider this case in detail below.



**Fig. 6.6.:** Evolution of the band structure of  $\text{EuX}_2$  (a-i) and  $\text{GdX}_2$  (j-r) under +5% tensile strain (right panel) and -5% compressive strain (left panel) in comparison to the case in equilibrium, shown in the middle in faded blue.

## 6.8 Quantized Anomalous, Spin and Orbital Hall Effect in Strained GdTe<sub>2</sub>

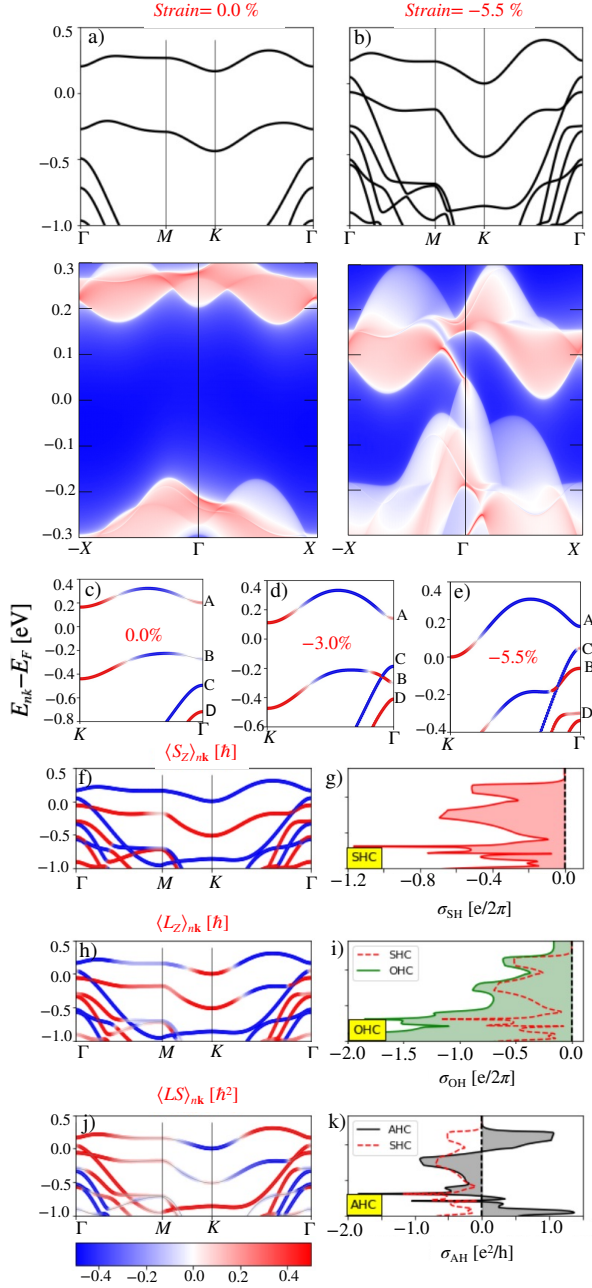
The closing of the bandgap in GdTe<sub>2</sub> upon  $-5\%$  strain with simultaneous presence of magnetization opens the possibility to implement a quantum anomalous Hall phase after a band inversion which opens a global band gap at the Fermi energy for a slightly larger compression of  $-5.5\%$ , see Fig. 6.7(a,b). Our results show that compressive strain shifts the occupied states towards the Fermi energy and induces changes in their spectral shape. At  $-5.5\%$  strain we observe quantized and plateau-like Hall conductivities with values of  $\sigma^{AH} \approx 1 \frac{e^2}{h}$ ,  $\sigma^{SH} \approx -1 \frac{2e^2}{h}$  and  $\sigma^{OH} \approx 1 \frac{2e^2}{h}$ , within the global band gap at the Fermi level, see Fig. 6.7(g,i,k). The quantization of the Hall conductance comes with the associated edge states upon strain with a single chiral edge mode visible at  $\Gamma$ , while the unstrained spectrum stays gapped. The computation of edge states for a zigzag terminated monolayer was conducted using the Green function method as implemented in WannierTools [205, 206].

To scrutinize the nature of Hall transport properties, we take a closer look at the behavior and correlation between the three types of Hall conductivity, shown in relation to the state-dependent out-of-plane spin polarization,  $S_{zn\mathbf{k}}$ , orbital out-of-plane polarization  $L_{zn\mathbf{k}}$  and spin-orbit polarization, defined as an average of the spin-orbit part of the Kohn-Sham Hamiltonian,  $\langle \mathbf{L}\mathbf{S}_{n\mathbf{k}} \rangle$ , shown respectively in Fig. 6.7(f), (h) and (j). From inspecting the spin-polarization of the states, Fig. 6.7(f), we observe that despite the presence of spin-orbit coupling, the states remain purely spin-polarized almost everywhere, which leads us to a conclusion that the effect of spin-flip part of spin-orbit interaction on Hall effects is negligible, and that it is primarily the orbital intermixing and hybridization which drive the Hall response. Indeed, the distribution of orbital polarization and its interchange among the bands appears to be much more complex, Fig. 6.7(h). Correspondingly, following the mechanism of orbital inversion, present already without SOC, a strong orbital Hall effect in the vicinity of the Fermi energy of strained GdTe<sub>2</sub> arises, Fig. 6.7(i). In turn, upon including spin-orbit into consideration, this gives rise to the SHC, shown in Fig. 6.7(g).

According to an early model-based theory [189], the SHC and OHC at a given energy are related to each other by the averaged spin-orbit polarization at that energy. And while it is clear that such a relation cannot be true in general, it seems to qualitatively hold in considered case. Indeed, from looking at the OHC and SHC plotted together in Fig. 6.7(i), we conclude that while the general features in both quantities agree very well, a drop in SHC magnitude at lower energies can

be attributed to a suppression of the spin-orbit polarization by the bands with negative  $\langle \mathbf{LS}_{n\mathbf{k}} \rangle$  below  $-0.3$  eV, Fig. 6.7(j), with this quantity being predominantly positive above this energy. In turn, given the suppression of spin-flip spin-orbit, the two-channel model of spin Hall and anomalous Hall effect, implying that the two types of conductivities are related by the spin-polarization of the states, also clearly works in our case. When plotted together in Fig. 6.7(j), the AHC and SHC exhibit a clear correlation in magnitude and features, and the changes in sign of the AHC can be understood from the changes in the spin-polarization of the states (shown in Fig. 6.7(f)), which carry the corresponding Berry curvatures.

Our Berry curvature analysis in  $k$ -space is shown in Appendix B (Fig. B.1) indicates that the origin of the topological properties around the Fermi energy comes from interaction among the bands around the  $\Gamma$ -point, and thus we look closer at the band evolution taking place around  $\Gamma$  in response to strain, Fig. 6.7(c-e). At equilibrium, Fig. 6.7(c), we see two weakly dispersing around the Fermi energy bands, A and B, with two lower bands C and D separated from band B by a couple of hundreds of meV. Given the predominantly spin-conserving nature of spin-orbit interaction at the Fermi energy, among these four participating bands, the pairs of bands of the same spin, A and C (majority), and B and D (minority), are allowed to hybridize, and the degree of their hybridization increases upon compression of the lattice, as it is already visible for the case of  $-3\%$  strain (Fig. 6.7(d)), as the bands C and D move up in energy. At the latter strain the hybridization is not strong enough to cause an orbital inversion at  $\Gamma$ , and the Chern number in the gap remains zero. However, as the bands are brought closer to each other for larger strain, the orbital exchange among bands takes place, Fig. 6.7(e): the reversal in sign of orbital polarization at  $\Gamma$  among the bands A and C is clearly visible, while the orbital polarization of band B is increased at the expense of band D losing it. This orbital inversion process not only explains the rise and quantization of Hall effects in the gap at the Fermi energy, but also can be named responsible for enhanced conductivities in the energy range in between the band edges of B and D bands at the  $\Gamma$ -point around  $-0.2$  eV. The fact that the AHC would be quantized in that gap given the absence of other “contaminating” metallic bands is supported by the observation of a distinct edge state around  $\Gamma$  between  $-0.3$  and  $-0.2$  eV, Fig. 6.7(b).



**Fig. 6.7.:** Electronic structure and transport properties of GdTe<sub>2</sub>. Band structure is shown for the monolayer and for the ribbon without (a) and with (b) compressive strain of  $-5.5\%$ . The color range from blue to red represents increasing value of local DOS at one edge of the ribbon. Topological edge states are clearly visible in the strained case around the  $\Gamma$  point. (c-e) Shown is the evolution of the bands around the Fermi energy with strain. A, B, C and D mark the bands of interest, colored by the value of the orbital polarization, mediating band inversion and quantum anomalous Hall effect. (f)-(g) show the spin expectation value and spin Hall conductivity, (e)-(f) show the orbital expectation value and orbital Hall conductivity, and (g)-(h) show the spin-orbit polarization with anomalous Hall conductivity, as a function of energy. In (i) and (k) dashed line stands for the distribution of the SHC.

## 6.9 Conclusion

Driven by the intricate interplay of *pdf* hybridization, chemical composition, correlation effects, and strain, we have identified two promising candidates  $\text{EuSe}_2$  and  $\text{GdTe}_2$  where the electronic structure may give rise to quantization phenomena and exotic edge states. In  $\text{GdTe}_2$ , strain induces hybridization between the  $p_{x,y}$  and  $p_z$  states at the Fermi energy, facilitating an orbital inversion and exchange process. This leads to the quantization of anomalous, spin, and orbital Hall conductivities. Transport measurements on strained  $\text{GdTe}_2$  for example, after deposition on a suitable insulating substrate could directly confirm the topological nature of the gap.

In contrast,  $\text{EuSe}_2$  presents a topologically non-trivial gap well below the Fermi energy, which can be probed using spectroscopy techniques such as ARPES [207] to confirm the gap's presence and its influence on edge-state physics [208]. Interestingly, while the anomalous Hall effect (AHE) within the gap vanishes regardless of the magnetization direction, the quantized spin Hall conductivity (SHC) and orbital Hall conductivity (OHC) remain sensitive to magnetization changes. This behavior is highly unusual. Previous studies have attributed variations in the Chern number in 2D magnets to the formation of "mixed Weyl points" in the combined space of magnetization direction and the  $k$ -vector, which also contribute to enhanced spin-orbit torques [209, 210]. Our findings suggest the potential to extend this "mixed" viewpoint to encompass the intertwined spin and orbital channels with  $k$ -space geometry in 2D magnets, opening a pathway for exploring Berry-phase-driven spin-orbit and orbital torques. These torques could either be hidden [59] or enhanced by the breaking of inversion symmetry in structures such as Janus monolayers [56, 211–214].







# Orbital topology, orbital torques, and orbital pumping in two-dimensional rare-earth dichalcogenides

The design of spin-orbit torque properties in two-dimensional (2D) materials presents one of the challenges of modern spintronics. In this context, 2D layers involving rare-earth ions – which give rise to robust magnetism, exhibit pronounced orbital polarization of the states, and carry strong spin-orbit interaction – hold particular promise. Here, we investigate ferromagnetic Janus H-phase monolayers of 4*f*-Eu rare-earth dichalcogenides EuSP, EuSSe, and EuSCl using first-principles calculations. We demonstrate that all compounds exhibit significant spin-orbit torques which originate predominantly in the colossal orbital response to current of the Eu *f*-electrons. Moreover, we demonstrate that the corresponding orbital torques can be used to drive strong in-plane currents of orbital angular momentum with non-trivial direction of polarization. Our findings promote *f*-based 2D materials as a promising platform for spin-orbit torque applications, and motivate further research on educated design of orbital properties for orbitronics in two dimensions.

*The results presented in this chapter have already been published in Zeer et al., npj Computational Materials volume 11, 305 (2025) [215].*

## 7.1 Introduction

Spin-orbit torque design in two-dimensional materials represents one of the actively developing areas of 2D magnetism [216]. Following the continuity equations for the redistribution of angular momentum, it has been suggested that in purely 2D systems the spin-orbit torque should be entirely attributed to the orbital contribution [106], while it was found that in 2D layers of finite thickness the spin and orbital nature of the torque can be controlled by the magnetization direction [59]. One particular class of 2D materials – Janus H-phase – has attracted significant attention

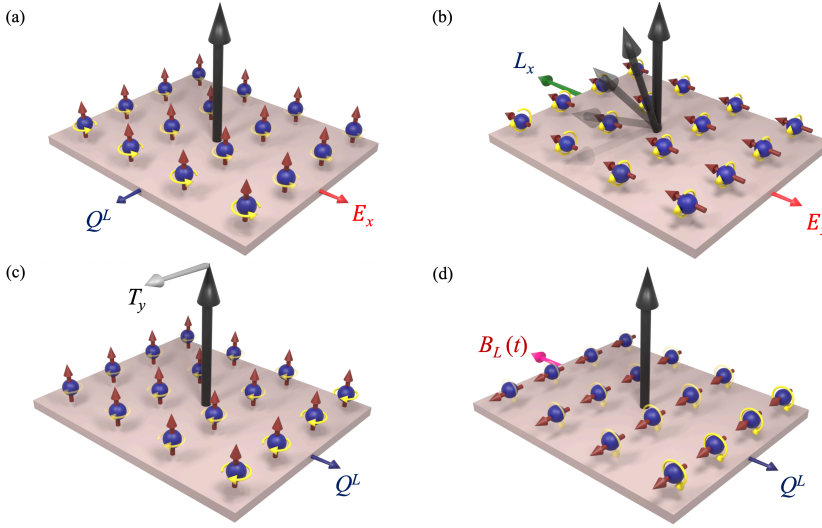


recently [217, 218]. The latter structure has been predicted and experimentally realized in nonmagnetic TMDs MoSSe and WSSe [219–221]. Low symmetry of these materials promotes such phenomena as the spin Hall effect [222] and topological insulators [223, 224]. The Janus layers were found to possess a high level of electrical conductivity, mechanical strength [225, 226], and thermal stability [227], making them a promising material class for applications in electronics, energy storage, and catalysis [228, 229]. The Janus structure has also attracted significant research in the context of spin-orbit torque switching [230].

In this work, taking as an example low-symmetry rare-earth dichalcogenides of Janus type, which combine strong magnetism with large SOC and orbital polarization, we explore the role of orbital magnetism in mediating spin-orbit torque and orbital pumping in 2D magnets based on  $f$ -electrons. First of all, we demonstrate that the current-induced torque acting on the  $f$ -states can reach an order of magnitude observed in most optimal representatives of the 2D magnets [231, 232]. We find that the  $f$ -torque is purely *orbital* in nature, that is, it is directly proportional to the current-induced local orbital moment of rare-earth atoms, Fig. 7.1(b). Second, we predict that igniting magnetization dynamics by external means will result in the effect of two-dimensional *orbital pumping*, with the pumped in-plane currents of angular momentum dominated by the orbital contribution, Fig. 7.1(c). Moreover, our prediction is that, due to the low symmetry of the structure, the response of pumped current is non-trivial in terms of direction and polarization, which may find direct applications in spintronic architectures based on an in-plane spin and OAM injection. Finally, we uncover the origin of the orbital pumping in the effect of *orbital-to-orbital-current conversion* mechanism, Fig. 7.1(d), where a perturbation of the Hamiltonian by the time-dependent orbital field can fully replace the dynamical term in predicting the response, thus postulating an equivalence between the torque and OAM operators within the subspace of  $f$ -electrons. At the end of the manuscript, we discuss the implications of our findings in the discipline of 2D magnetism and spintronics research and experimental methods to confirm our predictions.

## 7.2 Computational Details

We performed our density functional theory (DFT) calculations of Janus H-phase monolayers of EuSX ( $X = \text{P, Se, Cl}$ ) using the full-potential linearized augmented plane wave method (FLAPW), as implemented in the Jülich DFT code FLEUR [190], with the Perdew-Burke-Ernzerhof [139] parametrization of the exchange-correlation potential. The structural optimization was carried out using the FLEUR code with



**Fig. 7.1.:** Orbital torque and orbital pumping in 2D Janus dichalcogenides. (a) In 2D magnetic dichalcogenides (magnetization out of the plane along  $z$ , indicated with a black arrow), application of an electric field  $E_x$  along  $x$  (red arrow) leads to generation of a current of orbital angular momentum  $Q^L$  along  $y$ , polarized along  $z$  (small red arrows and yellow circular arrows) via the mechanism of the *orbital Hall effect* (OHE). (b) At the same time, due to broken  $z$ -reflection symmetry in Janus geometry, the electric field will give rise to a current-induced orbital polarization  $L_x$  (green arrow, as well as small red arrows and yellow circular arrows), which mediates an *orbital torque* on the magnetization along  $y$  (depicted with fading arrows). (c) In a reciprocal process, perturbing the magnetization with a torque  $T_y$  along  $y$  (grey arrow) will result in a generation of an orbital current along  $x$  polarized along  $y$  and  $z$  (shown with small yellow and red arrows), which constitutes the effect of *in-plane orbital pumping*. (d) For rare-earth dichalcogenides, the effect of orbital pumping is equivalent to the effect of *orbital-to-orbital-current conversion*, where the orbital current arises in response to an increasing linearly in time orbital field  $B_L(t)$  applied along  $x$  (pink arrow). While the OHE can be used to generate orbital currents in 2D materials, the effect of orbital pumping mediated by orbital torque can be used to achieve orbital current generation by magnetization dynamics in 2D geometry.

DFT+ $U$  method. For the self-consistent calculations, a  $16 \times 16$   $k$ -point mesh was used to sample the first Brillouin zone. All calculations included spin-orbit coupling (SOC) self-consistently within the second variation scheme. To account for the effect of strongly correlated electrons of the highly localized  $4f$ -electrons of Eu, the GGA+ $U$  method was applied within a self-consistent DFT cycle. The on-site Coulomb and Hund exchange parameters were set to  $U = 6.7$  eV and  $J = 0.7$  eV, which are values commonly used for the treatment of  $f$  electrons in chemical elements with half-filled  $4f$  shells such as Eu and Gd [191, 233]. We varied the

quantization axis of the Janus H-phase monolayer EuSX and associated spin-orbit interaction of the Kohn-Sham wavefunctions within the self-consistent iterations. The electric field  $E$  was defined along the  $x$ -axis, and the quantization axis varied from out-of-plane to in-plane. Table 7.1 summarizes the relaxed structure lattice parameters, magnetic moments of Eu, S, and X atoms, and the band gaps calculated using the GGA+ $U$  method. Regarding the computational parameters, we set the muffin-tin radii to  $2.80 a_0$  for Eu and 1.69, 1.84 and  $2.35 a_0$  for S, and 2.16, 2.35 and  $2.95 a_0$  for X, where  $a_0$  is the Bohr radius. The cut-off for the plane-wave basis functions was chosen as  $K_{\max} = 3.7, 4.3$  and  $3.9 a_0^{-1}$  for the wave functions and  $G_{\max} = 11.1, 12.9$  and  $11.7 a_0^{-1}$  for the charge density and potential for EuSP, EuSSe and EuSCL, respectively. The upper limit of angular momentum inside of the muffin-tin sphere was set to  $l_{\max} = 10$  for Eu and  $l_{\max} = 6, 8$  and  $10$  for S and 8, 8 and  $10$  for X. The magnetic anisotropy energy of EuSP, EuSSe and EuSCL was estimated to be 0.73, 0.047 and 2.341 meV, respectively, favoring an out-of-plane direction of magnetization.

To assess the stability and synthesizability of the studied materials, we performed phonon-based dynamical stability analyses. The calculated phonon dispersions, presented in Appendix C (Fig. C.1), show no imaginary frequencies and therefore confirm the structural stability of these systems.

From DFT calculations, we extracted 36 maximally localized Wannier functions (MLWFs) by using Eu- $d$ , Eu- $f$  and S,X- $p$  orbitals as initial projections, as these orbitals dominate the band structure in a wide energy window around the Fermi energy, with the frozen window maximum set to 2 eV above the Fermi energy. The electronic band structure was compared to the band structure obtained by constructing MLWFs in the FLAPW formalism [192] and the open-source wannier90 [234]. Based on the tight-binding Wannier Hamiltonian constructed from the Wannier functions, we computed the transport properties on a  $500 \times 500$  interpolation  $k$ -mesh. The anomalous Hall conductivity (AHC), spin Hall conductivity (SHC), and orbital Hall conductivity (OHC) were calculated using the Kubo formula and Wannier interpolation technique.

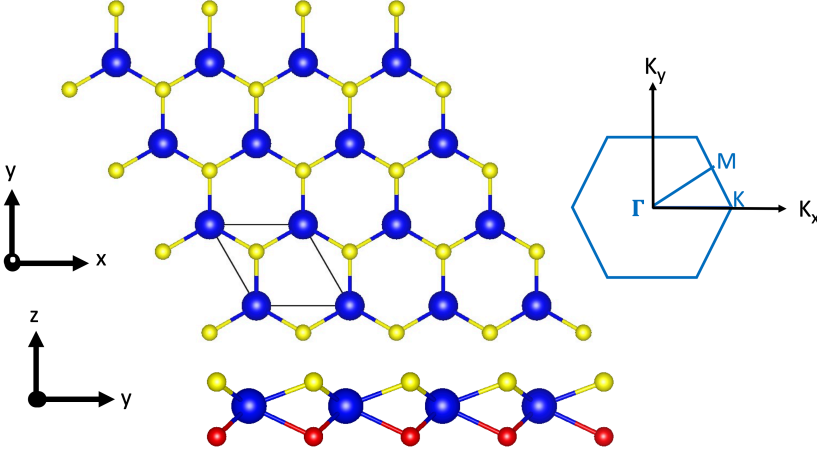
## 7.3 Electronic and Magnetic Properties of EuSX

We start by analyzing evolution of the electronic structure and magnetic properties when increasing the number of electrons in the systems by going from EuSP, to EuSSe, and EuSCL. The bandstructures and spin-resolved density of states (DOS) for EuSX monolayers computed with SOC are shown in Fig. 7.3. Starting with the

case of EuSP, Fig. 7.3(a-b) we find that the system is a metal with states at the Fermi energy dominated by  $p$ -orbitals of P, and Eu- $f$  states located much lower in energy. While the bandstructures look very similar for the cases with magnetization along  $x$  ( $M_x$ , not shown) and  $y$  ( $M_y$ ), the bandstructure for magnetized along  $z$  ( $M_z$ ) EuSP is noticeably different, especially in the region of  $f$ -states of heavy Eu atoms, where significant gaps among the  $f$ -bands are visible. Remarkably, the  $f$ -state of Eu and the  $p$ -state of S or P atoms are always separated in energy irrespective of the magnetization direction. In contrast, the  $p$ -states of S and P atoms exhibit a strong hybridization, particularly between  $-3$  eV and  $-4$  eV. On the other hand, the system exhibits a large magnetic moment irrespective of the magnetization directions. For instance, when the magnetization is out-of-plane, Eu atom carries about  $6.83 \mu_B$ , while P and S atoms exhibit small magnetic moments of  $-0.34$  and  $-0.12 \mu_B$ , respectively. Remarkably, in  $M_z$  case EuSP possesses an orbital moment on Eu of  $-0.047 \mu_B$ , which is the largest among all considered materials.

Turning to EuSSe, Fig. 7.3(c-d), we observe that it is a semiconductor with a large band gap of 1.84 eV, with the valence band maximum (VBM) and the conduction band minimum (CBM) both located at the  $\Gamma$  point. The VBM is dominated by the  $4f$  majority states of the Eu atom, while the CBM originates from the  $p$  states of S/P atoms. Additionally, EuSSe has the highest magnetic moment of  $6.95 \mu_B$  among all EuSX layers, and strong hybridization between the majority  $f$ -state of Eu and  $p$  states of S and Se atoms near the Fermi energy. Most remarkably, by altering the magnetization from  $M_z$  to  $M_x$  or  $M_y$ , the 4  $p$ -bands in the region from  $-3.5$  to  $-2.5$  eV become degenerate, Fig. 7.3(d), creating a nodal line around  $\Gamma$ , while forming a global bandgap of 125 meV for  $M_z$ , as shown in Fig. 7.3(c). This non-trivial gap between the two upper and two lower majority  $p$ -bands leads to quantized spin and orbital Hall conductivity, as will be discussed later in next section.

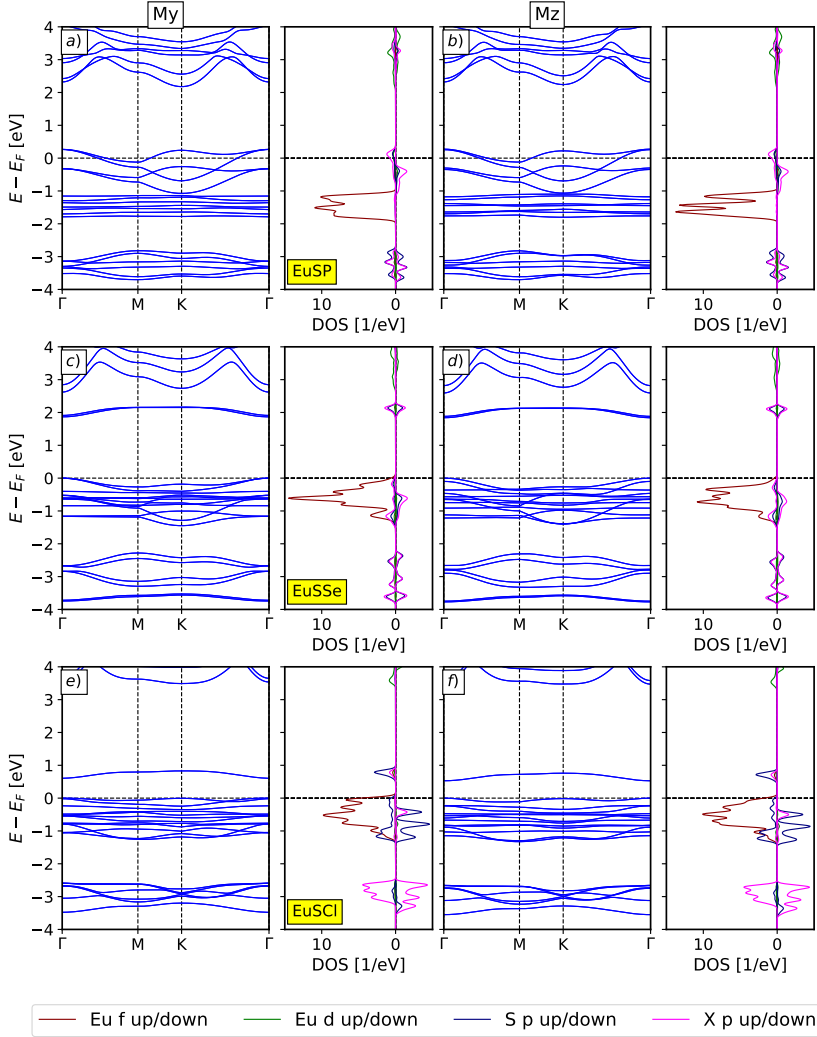
Finally, we discuss the the electronic structure of the EuSCl monolayer, shown in Fig. 7.3(e-f). This system has a direct semiconducting band gap of 0.534 eV at  $\Gamma$ , which is generated by the flat band of majority  $4f$ -state of the Eu atom and the minority  $3p$  of the Cl atom. It also exhibits a large magnetic moment of  $6.88 \mu_B$ , and the largest magnetic moment of the S atom among all layers of  $-0.64 \mu_B$ , which can be explained by the partial occupation of spin-split  $p$ -bands. On the other hand, this system shows strong hybridization between the  $4f$ -state of Eu and the  $p$ -state of Cl atoms as compared to S atoms, and a weak hybridization between  $p$ -states of S and Cl, which can be confirmed by inspecting the DOS in Fig. 7.3(e-f). In addition, the band structure near Fermi energy shows no (avoided) crossing



**Fig. 7.2.:** Top and side views of the Janus H-phase monolayer EuSX ( $X = \text{P, Se, and Cl}$ ) are shown. The dark blue balls represent Eu atoms, and the yellow/red balls represent S and X atoms, respectively, with the axes shown with black arrows. The first Brillouin zone is shown on the right.

between the  $f$ -state of Eu and  $p$  state of Cl or S. In contrast, in the case of EuSSe, the  $4f$ -state and  $3p$ -state of S and Se strongly hybridize and the bands cross each other. We demonstrated that the addition of electrons to EuSX will increase the degree of hybridization between the different configurations of EuSX, resulting in a more stable hybridized state. This is due to the increased number of electrons available for hybridization, which makes the hybridized state more energetically favourable. We have found that the  $4f$ -state of Eu in EuSP does not hybridize with the  $p$ -state of S and P atoms, but it does strongly hybridize with the  $p$ -state of both S and Se atoms in EuSSe, and with the  $p$ -state of the Cl atom in EuSCL.

To conclude this point, we have shown that Janus H-phase monolayer EuSX is a promising candidate for spintronics applications due to their large magnetic moments and tunable hybridization between the  $f$ -state of Eu and the  $p$ -states of S and X atoms. For instance, EuSP has a large magnetic moment and lacks hybridization between the  $f$ -state of Eu and the  $p$ -state of S or P atoms. Furthermore, the system exhibits a metallic phase with a band structure that is sensitive to the direction of the magnetization axis, which could be used to control spin-dependent transport properties. Additionally, EuSSe and EuSCL have large band gaps, high magnetic moments, and strong hybridization between the majority  $f$ -state of Eu and  $p$ -states of S and Se atoms near the Fermi energy. Their large band gaps and high magnetic moments make them suitable for use in spin-filtering devices, while



**Fig. 7.3.:** Electronic structure of H-phase EuSX (X=P, Se and Cl) for the direction of the magnetization along  $y$  and  $z$ , denoted as  $M_y$  and  $M_z$ , respectively. In (a)-(b), (c)-(d) and (e)-(f) the band structures and the corresponding spin-resolved density of states (DOS) are shown. The left and right parts of the DOS correspond to the majority and minority spin, respectively. The DOS of Eu- $f$ , Eu- $d$ , S- $p$  and X- $p$  states is shown with dark red, green, blue and pink, respectively. While in case of EuSP the  $p$  states dominate the Fermi energy, the strongest  $p$ - $f$  hybridization around the Fermi energy is achieved for EuSSe, with  $p$ -states going further down in energy in E.



**Tab. 7.1.:** Optimal lattice parameters and magnetic properties of EuSX for out-of-plane magnetization: lattice constant ( $a$ ), buckling height between Eu-S/X atomic planes  $\Delta(\text{Eu} - \text{S/X})$ , spin magnetic moments ( $S$ ) of Eu and S/X atoms, orbital magnetic moment ( $L$ ) of Eu, and the band gap.

| Materials  | Lattice<br>(Å) | $\Delta(\text{Eu-S})$<br>(Å) | $\Delta(\text{Eu-X})$<br>(Å) | $S_{\text{Eu}}$<br>( $\mu_B$ ) | $S_{\text{S}}$<br>( $\mu_B$ ) | $S_{\text{X}}$<br>( $\mu_B$ ) | $L_{\text{Eu}}$<br>( $\mu_B$ ) | Band<br>Gap<br>(eV) |
|------------|----------------|------------------------------|------------------------------|--------------------------------|-------------------------------|-------------------------------|--------------------------------|---------------------|
| EuSP+ $U$  | 4.789          | 1.136                        | 1.008                        | 6.829                          | −0.337                        | −0.120                        | −0.047                         | 0.002               |
| EuSSe+ $U$ | 4.785          | 1.029                        | 1.307                        | 6.951                          | −0.022                        | −0.024                        | −0.007                         | 1.847               |
| EuSCL+ $U$ | 4.479          | 1.315                        | 1.571                        | 6.875                          | −0.638                        | −0.192                        | −0.035                         | 0.534               |

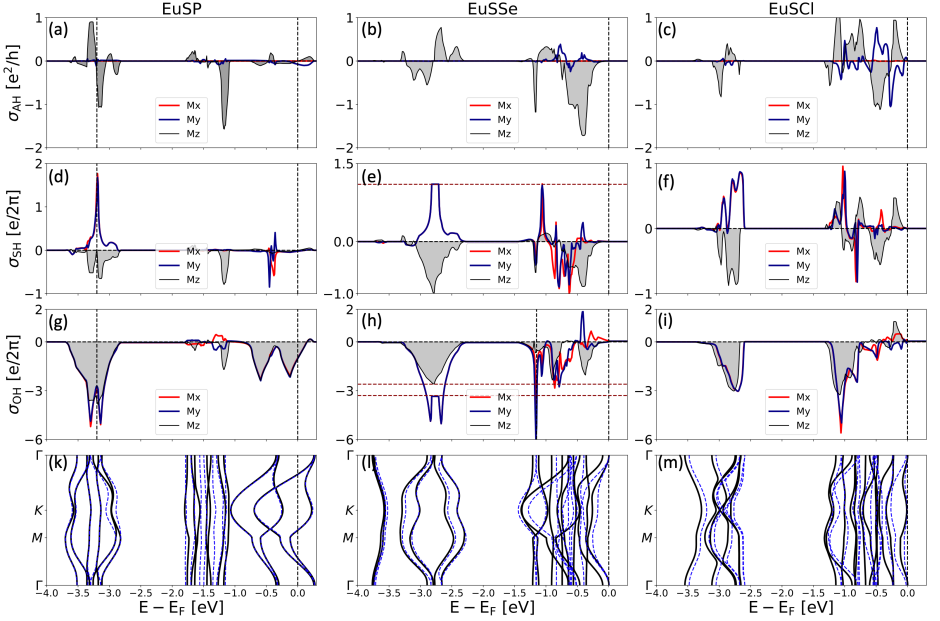
their strong hybridization near the Fermi energy makes them suitable for use in spin-polarized transport applications. Moreover, the EuSX Janus structure makes them ideal candidates for use in van der Waals heterostructures, allowing for further exploration of their potential applications.

## 7.4 Transport Properties of EuSX

We present the results of the effect of additional electrons on the transport properties as we progress from EuSP to EuSSe and EuSCL, as illustrated in Fig. 7.4, while considering the impact of changing the magnetization axis. Each row in the figure corresponds to data for a specific system as a function of band filling, with different quantization axes ( $M_x$ ,  $M_y$ , and  $M_z$ ) represented by red, blue, and gray areas, respectively. The AHC, SHC, and OHC are displayed in the first, second, and third columns, respectively. While these systems exhibit several remarkable findings, we focus on key points in this discussion, as the primary objective of this chapter is to highlight the spin-orbit torque in these materials.

In the case of the EuSP system, the electronic states are categorized into three distinct energy regions: the hybridized  $p_y \pm p_z$  states of S and P atoms, occurring at  $\approx (-3.7 \text{ to } -2.8 \text{ eV})$ ; the  $4f$  states of Eu, situated at  $\approx (-1.8 \text{ to } -1.1 \text{ eV})$ ; and the  $p_y$  states of S and P atoms, appearing at  $\approx (-1 \text{ to } 0.3 \text{ eV})$ . These regions are shown in Fig.7.3(a) and (b), with their respective AHC, SHC, and OHC conductivities shown in Fig.7.4(a), (b), and (c).

The hybridized  $p$ -states of S and P atoms in the energy range of  $-3.7 \text{ to } -2.8 \text{ eV}$ , for the out-of-plane magnetization axis ( $M_z$ , gray area), exhibit an AHC conductivity of approximately  $\pm 1 \frac{e^2}{h}$ . For Quantum Hall-based conductivity, the band structure of EuSP is effectively gapped in the intervals  $-3.317 \text{ to } -3.259 \text{ eV}$  and  $-3.177 \text{ to }$



**Fig. 7.4.:** Transport properties of monolayer EuSP, EuSSe, and EuSCl as a function of band filling. Panels (a-c) show the anomalous Hall conductivity (AHC,  $\sigma_{\text{AH}}$ ), spin Hall conductivity (SHC,  $\sigma_{\text{SH}}$ ), and orbital Hall conductivity (OHC,  $\sigma_{\text{OH}}$ ) for EuSP; panels (d-f) show the same for EuSSe; and panels (g-i) for EuSCl. These plots highlight the differences between in-plane magnetization ( $M_x$  red and  $M_y$  blue lines) and out-of-plane magnetization ( $M_z$ , shaded grey areas). Panels (k-m) present the corresponding band structures for each system under out-of-plane magnetization (solid black lines) and in-plane magnetization (dashed blue lines), illustrating the influence of magnetization orientation on the electronic structure for (k) EuSP, (l) EuSSe, and (m) EuSCl.

−3.120 eV, resulting in quantized SHC and OHC with plateau-like conductivity within these "insulating" regions. In the intermediate region, where the bands approach each other, the AHC inverts and vanishes precisely at the avoided crossings as shown in Fig.7.4(a). In the same energy region, the SHC exhibits a dip, deviating from its (half)-quantized values above and below the crossing points. The OHC also decreases slightly but remains the only surviving component at these energies (−3.2 eV). For in-plane magnetizations ( $M_x$  and  $M_y$ ), the AHC shows small peaks near the energy at which it vanishes, while the SHC exhibits a sharp peak, with a value of  $\sigma_{\text{SH}} \approx +2 \frac{e}{2\pi}$ , accompanied by a sign change. Similarly, the OHC for  $M_x$  and  $M_y$  follows a comparable trend, displaying two peaks on either side of the avoided crossing region. The most remarkable finding for EuSP is the significant OHC at the Fermi energy ( $\sigma_{\text{OH}} \approx -1 \frac{e}{2\pi}$ ), which remains consistent across all

quantization axes, while the AHC and SHC are comparatively small. While for the Eu  $4f$  states, spanning the range  $-1.8$  to  $-1.1$  eV and energetically separated from the  $p$ -states, produce quantized and plateau-like conductivities for the AHC, SHC, and OHC. These features are particularly evident between the second and third  $f$ -bands from the Fermi energy, as shown in Fig. 7.4(a), (d), and (g).

We now turn our attention to the results for the EuSSe monolayer as shown in Fig.7.4(d), (e), and (f). In this system, the AHC, SHC, and OHC can be divided into two distinct regions: the first, spanning  $-3.2$  to  $-2.2$  eV, where the  $p$ -states of S and Se exhibit strong hybridization (see Fig.7.3(c) and (d)); and the second, below the Fermi energy, involving the hybridization of Eu  $f$ -states with the  $p$ -states of S and Se atoms. For the AHC under out-of-plane magnetization ( $M_z$ ) within the energy interval of  $-3.2$  to  $-2.2$  eV, the behavior resembles that of EuS<sub>2</sub> and EuSe<sub>2</sub>, as shown in Fig. 6.4. However, the global gap between the four bands is narrower than in EuSe<sub>2</sub> but wider than in EuS<sub>2</sub>. This topological gap is characterized by a zero AHC, while the SHC and OHC exhibit quantized, plateau-like behavior. When the magnetization axis is changed to an in-plane orientation, thereby altering the quantization axis, a global energy gap of 120 meV is formed. Within this gap, both SHC and OHC maintain quantized plateaus with a plateau-like behavior, whereas the AHC remains entirely suppressed. A notable observation within this gap is that the SHC changes sign, consistent with the behavior shown in Fig.6.4(e). Meanwhile, the OHC plateau increases from  $\approx -2.6$  to  $-3.4 \frac{e}{2\pi}$ , as indicated by the horizontal dashed red lines in Fig.6.4(h). This behavior is confirmed by the spin and orbital Berry curvatures, as illustrated in Fig. 6.4. This enhancement occurs precisely in the energy range where the AHC is entirely suppressed across the four bands under in-plane magnetization.

On the other hand, a sharp peak in the OHC is observed for in-plane magnetizations  $M_x$  and  $M_y$ , as shown by the dashed line in Fig.7.4(h). This peak corresponds to the last two bands near the Fermi energy. Upon changing the magnetization from out-of-plane ( $M_z$ ) to in-plane, these two bands move together from the  $M$ -point to the  $\Gamma$ -point, exhibiting band degeneracy, as illustrated in Fig.7.3(c) and (d). Notably, at this energy ( $-1.1$  eV), the OHC is zero for out-of-plane ( $M_z$ ) magnetization.

Finally, we discuss the EuSCl system, focusing on the energy region dominated by the  $p$ -states of S and Cl atoms. Changing the magnetization axis results in only a slight upward shift of the band structure in energy, without forming band crossings as shown in Fig.7.3(e) and (f). This behavior indicates that the SHC and OHC conductivities are relatively similar for both in-plane and out-of-plane

magnetization, although the SHC changes sign. Notably, when the SHC drops to zero, the OHC transitions to a larger magnitude. For out-of-plane magnetization, the AHC transitions from negative to positive across this energy range. In the region of hybridization between Eu  $f$ -states and the  $p$ -states of S and Cl, the system exhibits nearly quantized values of AHC, SHC, and OHC near the Fermi energy for out-of-plane magnetization, centered around  $-0.2$  eV. These quantized conductivities,  $\sigma_{\text{AH}} \approx +1 \frac{e^2}{h}$  and  $\sigma_{\text{OH}} \approx +1 \frac{e}{2\pi}$ , correspond to the valence band maximum associated with a global gap present in this region.

### 7.4.1 General Features of Transport Properties in EuSX

The AHC, SHC, and OHC in these materials exhibit several common characteristics which can be categorized as follows:

- The SHC and OHC of EuSX for in-plane magnetization directions ( $M_x$  and  $M_y$ ) are similar at low energy due to their origin in purely  $p$ -states but exhibit slight differences near the Fermi energy as the result of Eu- $f$  state.
- Adding more electrons in the progression from EuSP to EuSSe and EuSCl enhances the strong hybridization among  $pdf$  states, leading to particularly striking behavior of AHC, SHC, and OHC near the Fermi energy. Notably, the OHC shows a larger magnitude as compared to the SHC as a function of band filling.
- The SHC reverses sign when comparing out-of-plane and in-plane configurations, whereas the OHC maintains a consistent sign in both cases.
- Changing the magnetization axis from out-of-plane to in-plane induces band degeneracy, which results in a pronounced peak in the OHC.
- The AHC in the in-plane configuration within the region of  $p$ -states is relatively trivial, whereas this region displays significant signals for SHC and OHC.

Overall the band structures reveal how in-plane and out-of-plane magnetizations impact electronic topology. In-plane magnetization enhances band crossings, while out-of-plane magnetization stabilizes gaps, reflecting distinct anisotropies. These insights are crucial for designing devices that exploit Hall conductivities for data



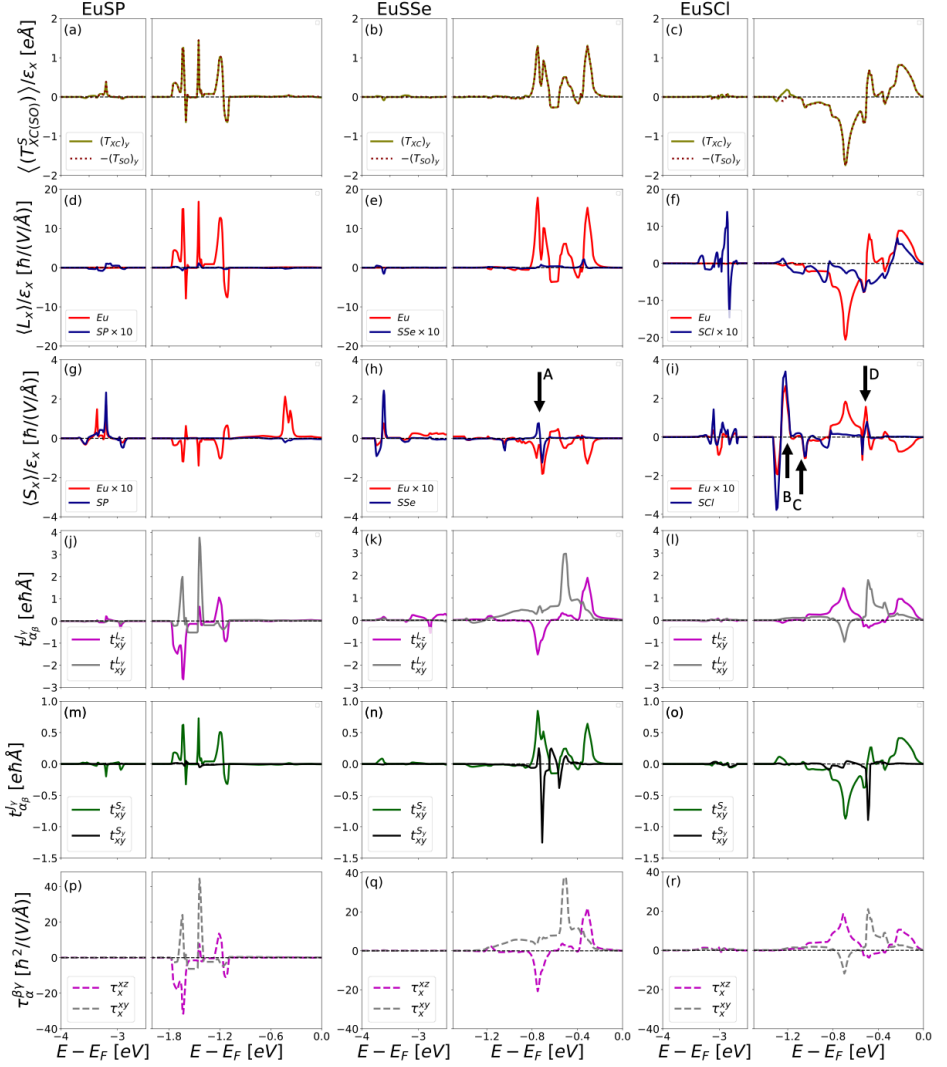
storage and processing. The tunability of these properties through magnetization orientation, strain, or external fields offers pathways to enhance functionality.

## 7.5 Orbital Torque in EuSX

The geometry of EuSX is of Janus type, where the structural inversion symmetry and the reflection symmetry with respect to  $z$  ( $\mathcal{M}_z$ ) are naturally broken, which allows for the effect of spin-orbit torque for the out of plane magnetization. We present the results of our calculations for the only non-vanishing  $y$ -component of the overall exchange torque (*i.e.*, the non-vanishing  $t_{yx}$  component of the torque tensor) and the orbital torque in Fig. 7.5(ac) for the case of out-of-plane magnetization, as a function of band filling, using Eq. (2.37). We set the temperature  $T = 300$  K. The broadening parameter  $\eta$  effectively describes the degree of disorder, which also help numerical convergence. We set  $\eta = 25$  meV in all the calculations shown below. In the following, for brevity, we skip the explicit reference to the distribution function in the summation, referring thus to the zero temperature limit expressions for computed quantities.

First, we find that the overall magnitude of the exchange torque is very large, almost reaching  $-2$  eÅ, far exceeding that in transition metal heterostructures [235]. This is comparable to the magnitude exhibited by some representative 2D materials [22, 210, 236–238]. For EuS<sub>2</sub>Cl, large values of the torque can be achieved in direct proximity of the Fermi level, which underlines the promise of this particular material for spin-orbit torque applications. Expectedly, the total summed over all atoms exchange torque in these systems is equivalent to the orbital torque, which is also apparent from the fact that it is very difficult to distinguish the curves for the orbital and exchange torques in the whole interval of energies, Fig. 7.5(a-c).

By relating the regions in energy, where the exchange torque is non-zero, to the band-resolved contributions shown in Fig. 7.6(a-c), we identify the largest torque with the region of  $4f$ -states, observing that a much smaller torque also arises in the region of  $p$ -states below the  $f$ -region (for EuSP). Correspondingly, we identify Eu atoms as contributing predominantly to the orbital “ $f$ ”-torque, although at some energies within the  $f$ -band of EuS<sub>2</sub>Cl the contributions of S and Cl atoms are also non-zero (not shown), owing to the  $pdf$  hybridization taking place in that region of energy. The origin of the torque in near band degeneracies becomes apparent after a careful analysis of band-resolved contributions: for example, while the two flat  $p$ -bands positioned at about  $-3.2$  eV in EuSP contribute only at the points where they cross the more dispersive bands, the latter provide a contribution over



**Fig. 7.5.: Orbital torque and orbital pumping in EuSX.** (a-c) Band-filling dependence of the  $y$ -component of spin-orbital ( $\langle T_{SO}^S \rangle$ ) and exchange ( $\langle T_{XC}^S \rangle$ ) torque normalized to the strength of an electric field  $E_x$  applied along  $x$ . The band-filling of the  $x$ -component of the normalized current-induced OAM ( $\langle L_x \rangle$ , d-e) and spin ( $\langle S_x \rangle$ , g-h), resolved into Eu and SX contributions, is shown for comparison. In (j-l), the distribution of orbital currents arising in response to the exchange torque along  $y$ , as given by tensor components  $t_{xy}^{L_z}$  and  $t_{xy}^{L_y}$  defined by Eq. 2.42, is shown. In (m-o), the respective tensors are shown for the case of spin current. The band filling dependence of the orbital-to-orbital-current conversion strength, as given by tensors  $\tau_x^{xz}$  and  $\tau_x^{xy}$  defined by Eq. 2.43, is shown for comparison in (p-r). The case of EuSP, EuSSe, and EuSCL corresponds to the left, middle, and right columns, respectively.

a larger part of the  $\mathbf{k}$ -path as they are nearly degenerate around  $-3$  eV. The roles of these two groups of  $p$ -states are interchanged in EuSSe, with flat bands being nearly degenerate resulting in a small but finite torque, while the more dispersive bands are split off from each other almost everywhere giving a vanishing torque signal.

Secondly, we clearly identify the orbital torque as by far the main source of the exchange torque in studied systems. This is apparent from that fact that it is close to impossible to distinguish the curves for the orbital and exchange torques for the energies within the  $f$ -band, Fig. 7.5(a-c). The only exclusion can be seen around  $-3.4$  and  $-0.5$  eV for EuSP, below  $-3.0$  eV for EuSSe and around  $-3.0$  and  $-1.2$  eV for EuSCl, *i.e.* within the  $p$ -band of S and X atoms. Correspondingly, we identify Eu atoms as contributing predominantly to the orbital “ $f$ ”-torque, although at some energies within the  $f$ -band of EuSCl the contributions of S and Cl atoms are also non-zero (not shown), owing to the  $pdf$  hybridization taking place in that region of energy. On the other hand, the small exchange “ $p$ ”-torque on small moments developed by S and X atoms has strong flux contributions, and thus can be to a large extent attributed to the spin current across the layer, arising as the electric field is applied.

### 7.5.1 Non-equilibrium Spin and Orbital Accumulation Induced by an External Electric Field

Next, to gain further insight into the origins of the torque, we analyze the non-equilibrium spin and orbital accumulation induced by an external electric field and calculated according to

$$\frac{\mathbf{J}}{e\hbar E_x} = \sum_{\substack{m\mathbf{k} > E_F \\ n\mathbf{k} < E_F}} \text{Im} \left[ \frac{\langle u_{n\mathbf{k}} | \hat{\mathbf{J}} | u_{m\mathbf{k}} \rangle \langle u_{m\mathbf{k}} | v_x | u_{n\mathbf{k}} \rangle}{N_{\mathbf{k}}(\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}} + i\eta)^2} \right], \quad (7.1)$$

where  $\hat{\mathbf{J}}$  stands either for the operator of spin or orbital angular momentum,  $\hat{\mathbf{S}}$  or  $\hat{\mathbf{L}}$ . The results are presented in Fig. 7.5(d-i) for the only non-vanishing components  $L_x$  and  $S_x$ , decomposed into Eu and SX contributions. The exchange  $f$ -torque observed in the monolayers of EuSX systems, as shown in Fig. 7.5, can be primarily attributed to orbital physics, which clearly manifests in a remarkable one-to-one correspondence in the band filling behavior of  $L_x$  on Eu atoms and  $T_{\text{XC}}$  in the region of  $f$ -states. This observation highlights the key role played by nonequilibrium OAM generation in mediating the overall torque in 2D limit. Note, that the  $L_x$  of SX atoms is non-vanishing only for the case of EuSCl, where it is still an order of

magnitude smaller than that on Eu atoms. In particular, for EuSCl, the non-equilibrium orbital magnetism is of purely  $p$ -origin in the  $p$ -region of states below  $-3$  eV, see also Fig. 7.6(f). The impact of the  $p$ -states in the  $f$ -region on the Eu- $L_x$  is also significant for EuSCl, as the behavior of  $L_x$  and the torque for this material is distinctly different from the other two materials – something, which can be clearly observed also in Fig. 7.6(d-f).

The analysis of current-induced spin polarization,  $S_x$ , in Fig. 7.5(g-i) confirms the origin of the non-equilibrium spin density on Eu atoms in the current-induced orbital magnetization by spin-orbit coupling: although the magnitude of Eu  $S_x$  is about two orders of magnitude smaller than that of  $L_x$ , the close correlation between the two quantities in the  $f$ -region is obvious. At the end, it is the  $L_x$ -induced  $S_x$  which is responsible for non-vanishing  $T_{XC}$  by the virtue of the matrix elements of the exchange torque  $\mathcal{T}_{XC} = \frac{1}{i\hbar}[\hat{\mathbf{S}}, H_{XC}] = -\mu_B \hat{\mathbf{S}} \times \mathbf{V}_{XC}$  (see section 2.8.1), which are expressed in terms of the operator of spin and exchange field [235]. It is important to realize, however, that although the operator of OAM does not participate in the exchange torque directly, its correlation with  $T_{XC}$  is much stronger: note for example that the response of  $S_x$  on Eu atoms is driven predominantly by the hybridization with the SX atoms at the energies indicated by small black arrows (marked as A, B, C and D) in Fig. 7.5(h-i). The corresponding features in Eu- $S_x$  are not reflected in  $T_{XC}$  – an observation that can be attributed to the fact that the overlap of the SX-induced spin density  $S_x(\mathbf{r})$  with strongly localized  $\mathbf{V}_{XC}(\mathbf{r})$  is small, with the former being inconsistent in symmetry with the angular expansion of the  $f$ -electron driven exchange field. On the other hand, the local orbital response  $L_x$  on Eu atoms is very localized as well, and it stems predominantly from the  $f$ -states which ensures the locality and suitable symmetry of the SOC-induced spin density. Our findings thus establish the local current-induced orbital angular momentum as the key factor in mediating spin-orbit torques in  $f$ -systems.

Remarkably, the latter points do not contribute to the exchange torque via  $T_{XC}^{\mathbf{S}} = (-2 \mu_B / \hbar) \mathbf{S} \times \mathbf{V}_{XC}$ , which is expressed in terms of the operator of spin and exchange field [235]. This effectively results in an observation that the current-induced orbital magnetization correlates with the exchange torque much stronger than the non-equilibrium spin density, despite the fact that the operator of OAM does not participate in  $T_{XC}^{\mathbf{S}}$  explicitly. This can be attributed to the fact that the overlap of the induced spin density  $S_x(\mathbf{r})$  at chalcogen atoms S and X with strongly localized  $\mathbf{V}_{XC}(\mathbf{r})$  is small, with the former being inconsistent in symmetry with the angular expansion of the  $f$ -electron driven exchange field. On the other hand, the local orbital response  $\langle L_x \rangle$  on Eu atoms is very localized as well, and it stems predomi-



nantly from the  $f$ -states which ensures the locality and suitable symmetry of the SOC-induced spin density. Our findings thus establish the local current-induced OAM as the key factor in mediating spin-orbit torques in  $f$ -systems.

## 7.6 Orbital Pumping in EuSX

Next, we focus on the spin and orbital current generation by magnetization dynamics using eq 2.42. In this context, one can also ask a question whether the time-dependent magnetization can give rise to the non-vanishing spin and orbital currents, arising in addition to the currents of charge. In the past, Kubo linear response formalism has been used to compute the effect of spin pumping – *i.e.* spin current driven by magnetization dynamics – into the substrate by a deposited thin ferromagnet, *e.g.* [111], with the effect being extensively studied experimentally [239]. In a similar geometry the twin effect of orbital pumping has been also recently observed [240]. In contrast to the previous works where the currents of spin and OAM propagate into the bulk of the film, here, we consider a situation where the corresponding currents are lying in the plane of our EuSX systems, see Fig. 7.1(c), constituting thereby the effect of *in-plane orbital pumping*.

For our compounds, the symmetry dictates that for the case of the torque along  $y$  ( $\beta = y$ ) the only non-vanishing components are  $t_{xy}^{J_z}$ ,  $t_{xy}^{J_y}$ , and  $t_{yy}^{J_x} = t_{xy}^{J_y}$ , while for the torque along  $x$  ( $\beta = x$ ) the non-vanishing components are  $t_{yx}^{J_z} = -t_{xy}^{J_z}$ ,  $t_{yx}^{J_x} = -t_{xy}^{J_y}$ , and  $t_{xx}^{J_x} = -t_{yy}^{J_y}$ . In other words, when a torque is applied on the magnetization, this results in a generation of two types of currents of angular momentum:

- (i) A current which is polarized along the direction of the magnetization and propagates together with the pumped flow of charge;
- (ii) An in-plane current of angular momentum with an in-plane propagation-direction-dependent polarization, whose properties can be controlled by the direction of the applied torque.

It is important to realize that were our atoms arranged in a square lattice with both mirror symmetries  $\mathcal{M}_x$  and  $\mathcal{M}_y$  present, only one, conventional, component  $t_{xy}^{J_z}$  would survive, driving the current of type (i). The complexity of the pumped currents of angular momentum is thus a direct consequence of the complex structural symmetry of considered Janus rare-earth dichalcogenides.

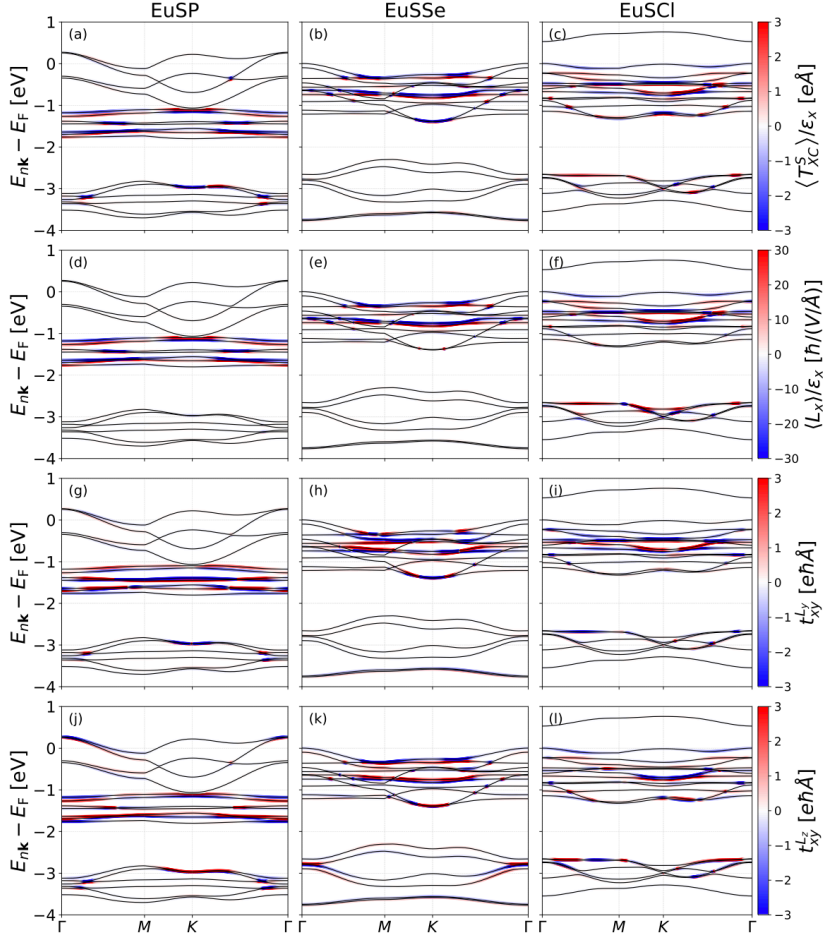
The tensor components  $t_{xy}^{S_z}$  and  $t_{xy}^{S_y}$  for the case of spin, and  $t_{xy}^{L_z}$  and  $t_{xy}^{L_y}$  for the case of orbital channel are shown for EuSX in Fig. 7.5(j-l) and Fig. 7.5(m-o), respectively, as a function of band filling. By first looking at the pumped spin currents, we immediately realize that for all compounds, in case of the  $z$ -polarization, the spin

current in the  $f$ -region is directly proportional to the generated by  $E_x$  distribution of  $\langle T_{XC}^S \rangle$  and  $\langle L_x \rangle$ , and not to the corresponding values of  $\langle S_x \rangle$ , Fig. 7.5(a-i). This is rooted in the fact that in the ground state the localized  $f$ -states, on which the torque predominantly operates, are almost pure eigenstates of  $\langle S_z \rangle$ , which results in proportionality of  $Q_x^{S_z}$  to  $\langle T_{XC}^S \rangle$ . On the other hand, for similar reasons, the  $Q_y^{S_y}$  is vanishing almost everywhere except for the spin-flip points where the spin-up and spin-down bands of SX and Eu atoms meet (observe for example vanishing  $t_{xy}^{S_y}$  for EuSP where  $f$ -states are well-separated from SP-states).

The behavior of the pumped orbital currents is, however, very different. We observe that, qualitatively, in the  $f$ -region the  $z$ -polarized orbital current can be reconstructed from the behavior of the  $\langle T_{XC}^S \rangle$  and  $\langle L_x \rangle$  in the lower (negative  $\langle L_z \rangle$ ) and upper (positive  $\langle L_z \rangle$ )  $f$ -band, qualitatively multiplied with the sign of  $\langle L_z \rangle$  (not shown), while being suppressed in the middle of the  $f$ -band, where the transition from positive to negative  $\langle L_z \rangle$  occurs. In an effective picture, one can thus perceive pumped orbital current  $Q_x^{L_z}$  as an orbital filtering effect, where the pumped charge current gets orbitally polarized along  $z$  as it is generated by an application of a torque. This picture has to be taken with caution, however, since it is the off-diagonal matrix elements of OAM operator which ultimately participate in the Eq. 2.42, and which do not necessarily reflect the orbital polarization of the states in the ground state.

When inspecting the in-plane polarized OAM current, we find that it is peaking in the middle of the  $f$ -band, where  $z$ -polarized current is suppressed. In contrast to  $Q_x^{S_y}$ ,  $Q_x^{L_y}$  is spread over wider regions of energy, owing to a higher complexity in the orbital composition of the bands which allows for richer coupling among the states with different values of OAM. On the other hand, when looking at the  $k$ -space behavior of the two types of orbital currents, shown in Fig. 7.6(g-l), we find two clear distinctions: first,  $Q_x^{L_y}$  is originating from narrower regions in the reciprocal space, and second,  $Q_x^{L_z}$  exhibits variations in sign when going along a given band, while  $Q_x^{L_z}$  is much more uniform when the  $k$ -point is varied within the regions of strong contributions.

To assess the sensitivity of our results to the choice of the Hubbard  $U$  and Hund's exchange  $J$  parameters in our DFT+ $U$  calculations. In fact, in previous chapters 5 and 6, we have carefully investigated the impact of the Coulomb repulsion parameter on the electronic structure and transport properties. Despite the fact that the value of  $U$  of about 7 eV is known to provide the best description of Eu-based materials in terms of their electronic structure [191, 233], we did to ensure the



**Fig. 7.6.: Anatomy of orbital torques and orbital pumping in  $k$ -space.** For each compound, the  $k$ -resolved distribution is shown for (a-c) exchange torque  $\langle T_{XC}^S \rangle$  normalized to the field  $E_x$ , (d-f) current induced orbital moment  $\langle L_x \rangle$  normalized to the field  $E_x$ , and pumped by the torque along  $y$  orbital current density along  $x$  polarized along  $y$  ( $t_{xy}^{L_y}$ , g-i), and along  $z$  ( $t_{xy}^{L_z}$ , j-l). For all cases, the colored circles represent the expectation value of the plotted quantity with the color code shown on the left.

robustness of our findings by systematically testing a range of  $U$  values. Our key results—specifically the magnitude and qualitative behavior of the orbital torque, as well as the spin and orbital pumping—remained consistent across this range.

### 7.6.1 Orbital to Orbital-Current Conversion

Above, we have observed that for  $f$ -electrons the effect of the current-induced torque on the exchange field is equivalent to the effect of current-induced orbital magnetization. Therefore, it seems rewarding to explore whether the same holds true for the inverse process: *i.e.*, whether the pumping of the OAM currents by the torque is equivalent to an OAM current arising in response to an induced orbital magnetization.

The calculations for the  $\tau_x^{xz}$  and  $\tau_x^{xy}$  components corresponding to the generation of  $y$ - or  $z$ -polarized OAM currents along  $x$  in response to the orbital field applied along  $x$  are shown in Fig. 7.5(pr) and are calculated using Eq. (2.43). From this data we make a remarkable observation: as far as pumped orbital currents in the region of the  $f$ -states are concerned, the effect of the magnetization dynamics represented by a torque along  $y$  is *quantitatively* completely equivalent to the effect of  $L_x$  via the application of a time-dependent orbital field, since the two types of tensors exhibit an almost identical to each other behavior in the region of corresponding energies. Conceptually, this is similar to the picture of the origin of the inverse spin Edelstein effect in a linearly increasing magnetic field acting on spin [112]. The effect of the OAM current generation by inverse orbital torque that we uncover here can be correspondingly qualitatively considered as an intrinsic version of the inverse orbital Rashba effect for the orbital current, see Fig. 7.1.



## 7.7 Discussion and Conclusion

Our study provides unique insights into the structural, electronic, magnetic, and transport properties of Janus H-phase monolayers of  $4f$ -Eu rare-earth dichalcogenides. The observation of substantial magnetic moments, distinct electronic hybridization channels, and non-trivial response to an applied current highlights the potential of these materials for spintronic device applications. The understanding gained from this research opens new avenues for designing and optimizing orbitronics devices based on  $4f$ -Eu rare-earth dichalcogenides, thus contributing to the broader knowledge of electronic and transport properties of 2D magnetic materials.

One of our key findings is observation of pronouncedly orbital character of the response to the electrical current which is mediated by  $f$ -electrons. The close correlation between the orbital magnetism and torques in  $f$ -systems suggest that the orbital torque properties can be engineered by carefully crafting the crystal field around rare-earth atoms due to surrounding cage of  $sp$ -atoms. The latter is susceptible to strong changes in 2D geometry which signifies a powerful channel for spin-orbit torque design. The found correlation between the orbital magnetism and torque within the  $f$ -shell is so strong that the torque operator acting on  $f$ -states can be replaced by the local OAM operator within the same sub-shell in the linear response expressions for the response properties. We attribute this to the properties of the exchange field of  $f$ -atoms, which is closely interlinked to the strong orbital polarization of the  $f$ -states by very large spin-orbit interaction. Since the properties of the  $f$ -states, their orbital polarization and degree of localization are very sensitive to the degree of correlation of valence electrons, one possible way to tune the response properties in considered classes of systems lies in alloying with other rare-earth or transition-metals.

One of the most remarkable properties of our systems that we uncovered is the effect of in-plane orbital pumping, *i.e.* pumping of currents of OAM by magnetization dynamics. Intuitively, one would expect that this effect can be attributed to the OAM polarized along the magnetization direction and carried by the pumped current of charge. However, we find this picture to be too naive to reflect the complexity that we observe in the magnitude, direction and polarization of the generated current. We speculate that this complexity may be exploited for the purpose of orbital current injection into a planar adjacent material, with a controllable polarization of the current. We predict that this is a generic effect which occurs in 2D magnetic materials of Janus type owing to their lowered crystal symmetry. While we leave

the exploration of exact microscopic mechanisms for the orbital pumping to future work, we make the first step in this direction by discovering that orbital pumping realized by  $f$ -electrons can be considered as an effective orbital-to-orbital-current conversion, where changing in time induced orbital polarization relaxes via a generation of an orbital current. Our findings open new possibilities for generating diverse flavors of orbital currents in 2D materials which go beyond the protocol of the orbital Hall effect. We thus declare that the peculiarities of 2D geometry combined with the specifics of  $f$ -electron magnetism may provide as robust and efficient platform for orbital current generation.





## 8.1 Summary

This thesis investigates spinorbitronic and topological properties of two-dimensional dichalcogenides, with a particular focus on rare-earth dichalcogenides (REDs). The study builds on the structural framework of conventional transition-metal dichalcogenides (TMDs), in which the transition-metal atoms are replaced by rare-earth elements. The analysis focuses on systems such as  $\text{EuX}_2$  and  $\text{GdX}_2$  ( $\text{X} = \text{S}, \text{Se}, \text{Te}$ ), as well as Janus H-phase monolayers including  $\text{EuSP}$ ,  $\text{EuSSe}$ , and  $\text{EuSCl}$ . It represents one of the first steps toward understanding how these materials, with their special electronic, magnetic, and structural properties, can be used for spintronics and orbitronics applications. We combined first-principles calculations using the FLEUR Jülich code, Wannier interpolation methods, and the Kubo formula to calculate key transport properties like anomalous Hall conductivity (AHC), spin Hall conductivity (SHC), and orbital Hall conductivity (OHC), and to study orbital torques.

The idea behind this work was to explore how combining  $f$ -electrons with chalcogenide atoms in two-dimensional materials can lead to new magnetic and transport properties.

In Chapter 5, we extended the well-known advantages of TMDs such as their exotic charge, spin, and orbital transport properties to the new field of rare-earth dichalcogenides. First, we focused on the H-phase  $\text{EuS}_2$  monolayer as a starting point and investigated its structure, electronic properties, magnetism, and transport behavior using *first-principles* methods. Our results showed that  $\text{EuS}_2$  becomes a half-metal under doping, with a large magnetic moment of  $6.95 \mu_B$  per unit cell and a large direct band gap. We also found that H-phase  $\text{EuS}_2$  exhibits a strong AHC, while the SHC closely follows it. Most importantly, however, the system exhibits a very large OHC that strongly depends on the strength of electronic correlations, as tuned by the Hubbard  $U$  parameter.





In Chapter 6, we expanded the study to monolayers of  $\text{EuX}_2$  and  $\text{GdX}_2$  ( $X = \text{S, Se, Te}$ ) in the H-phase. We found that these systems have strong magnetism and Hall transport effects. Interestingly, the OHC still appears even without spin-orbit coupling, suggesting it is a fundamental feature of these materials. We also found that in  $\text{GdX}_2$  compounds, electronic correlations control the energy levels and hybridization of  $p$ ,  $d$ , and  $f$  states, which affects the polarization of  $p$ -orbitals and the anomalous Hall effect. Applying strain to these materials leads to important changes: in  $\text{GdTe}_2$ , it causes orbital inversion near the Fermi energy, which leads to quantized charge, spin, and orbital transport—a clear indication of topological behavior.

In Chapter 7, we introduced new ideas related to orbital torque and orbital pumping in REDs. We discovered a strong orbital torque caused by  $f$ -electrons and proposed a new mechanism, called orbital-to-orbital current conversion, where a time-changing orbital angular momentum (OAM) creates an orbital current. This mechanism is especially important in Janus-type REDs ( $\text{EuSP}$ ,  $\text{EuSSe}$ ,  $\text{EuSCl}$ ), which have strong spin-orbit effects and broken inversion symmetry. The in-plane orbital pumping observed in these materials offers a new and efficient way to generate and control orbital currents, with possible advantages over traditional spintronic devices.

In conclusion, this thesis makes strong contributions to the growing fields of spin-orbitronics and orbitronics by showing the important roles of  $f$ -electron physics,  $pdf$  hybridization, and strain control in 2D materials. Our results show that REDs are excellent candidates for studying complex orbital transport and topological properties in two-dimensional magnetic systems.

Besides the fundamental discoveries, the results point to real applications. REDs could be used to develop new types of spintronic and orbitronic devices, low-energy memory storage, and even quantum computing technologies. We hope these results will encourage more theoretical and experimental work to explore the rich physics and practical potential of rare-earth-based 2D materials.

## 8.2 Outlook

This thesis opens several exciting directions for future research.

First, the strong spin-orbital interactions we found in H-phase  $\text{EuSe}_2$  monolayers suggest the existence of "mixed Weyl points," where spin and orbital channels are

intertwined with the  $k$ -space geometry and magnetization direction. Studying these features could bring new advances in spin-orbit torque physics and in understanding topological transport in low-dimensional systems. Another important direction is building heterostructures that combine REDs with TMDs. These layered structures offer more ways to control electronic, magnetic, and topological properties, helping to design flexible and tunable 2D materials for future quantum and spintronic devices. In particular, looking at the magnetic behavior at the interface could reveal how interfacial effects change magnetization direction, offering new ways to engineer spin and orbital textures and to control magnetic anisotropy at the nanoscale. Furthermore, studying how spin, orbital, and orbital quadrupole currents interact in REDs [241] may uncover new topological phases, special transport behaviors, and new techniques for generating and controlling orbital currents. Following these paths will deepen our understanding of correlated electrons in two-dimensional materials and will speed up the development of future orbitronic and spintronic technologies.

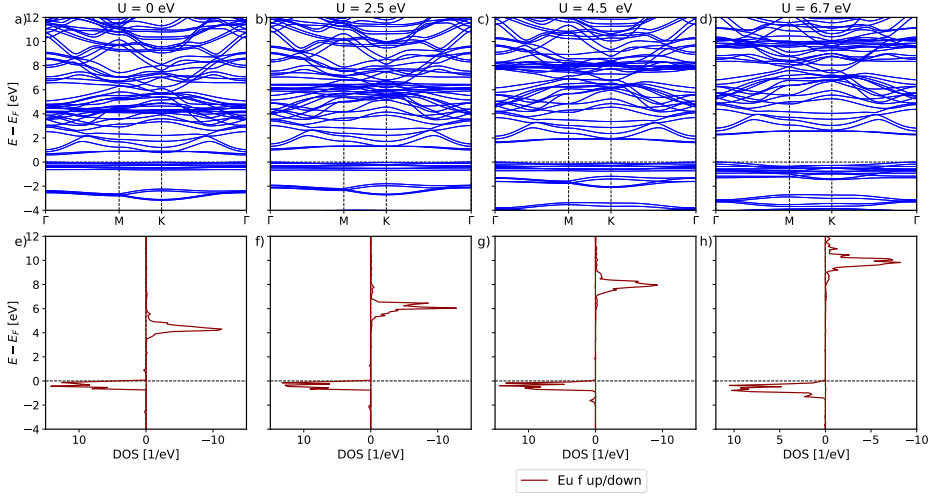
Rare-earth dichalcogenides stand out as a unique and powerful platform for pushing forward research in orbitronics, spintronics, and quantum information science.





## Appendix Chapter 5

# A



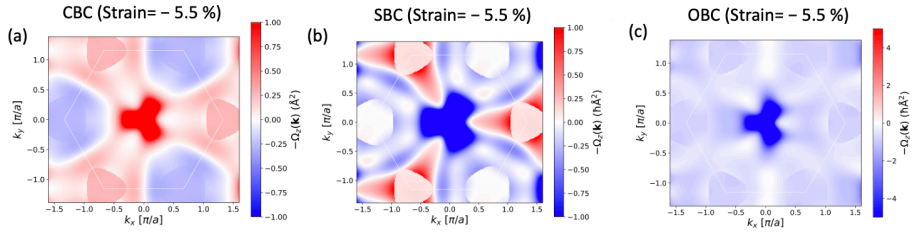
**Fig. A.1.:** Band structure of the  $\text{EuS}_2$  over an extended energy range, and DOS of 4f states for both majority and minority spins, as a function of the Hubbard parameter  $U$



## Appendix Chapter 6

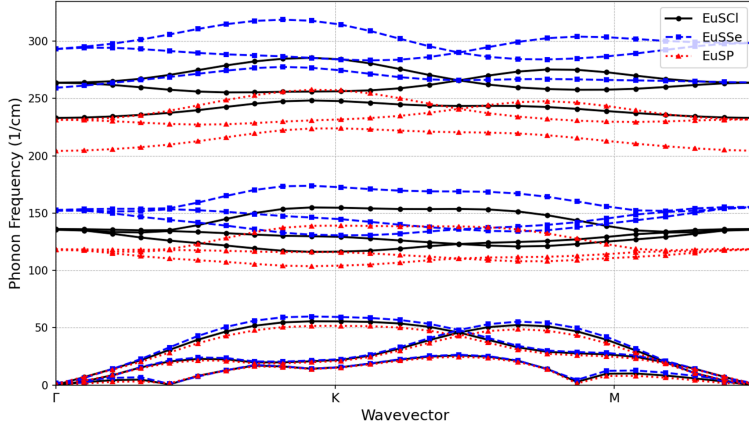
# B

To assess the stability of the strained  $-5.5\%$  GdTe<sub>2</sub> structure, we computed the formation energy ( $E_f$ ), which denotes the released energy during the formation of compounds from their respective elements. The formation energy of the monolayer GdTe<sub>2</sub> under a  $-5.5\%$  strain is determined by the equation  $E_f = E_{Bulk}(\text{Gd}) + 2E_{Bulk}(\text{Te}) - E_{Strained}(\text{GdTe}_2)$ , where  $E_{Bulk}(\text{Gd})$  and  $2E_{Bulk}(\text{Te})$  represent the total energy of the equilibrium states of bulk Gd, bulk Te and  $E_{Strained}(\text{GdTe}_2)$ , respectively. The calculated formation energy for the monolayer under a  $-5.5\%$  strain constitutes a very large value of 2.21 eV, indicating that the system would be still stable at this strain. In addition, a recent work reports phonon calculations for GdS<sub>2</sub> and GdSe<sub>2</sub> [242], suggesting the stability of these monolayers. According to this work, these monolayers could be synthesized like GdGe<sub>2</sub> monolayers [243].



**Fig. B.1.:** (a), (b), and (c) represent the charge, spin, and orbital Berry curvature, respectively



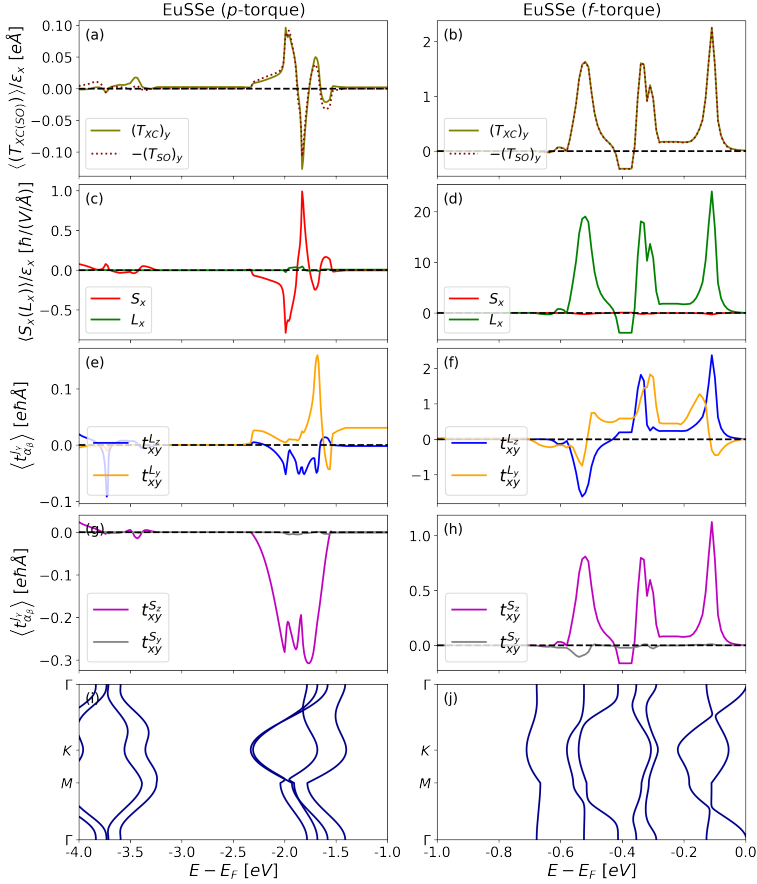


**Fig. C.1.1:** Phonon band structures of EuSP, EuSSe, and EuSbI. The absence of imaginary frequencies confirms the dynamical stability of all EuSX monolayers.

As a concrete example of the sensitivity of our results to the choice of the Hubbard  $U$  and Hunds exchange  $J$  parameters in our DFT+ $U$  calculation, here we present additional calculations for EuSSe using a smaller value of  $U = 3.5$  eV. While these results are not included in the main text, they show that the orbital torque and spin orbital pumping retain their characteristic features, further confirming the insensitivity of our conclusions to the choice of  $U$ . For instance, the magnitude of the orbital torque exhibits two distinct regions as a function of band filling. One region, associated with the  $p$ -state contributions (shown in the left panel of Fig. C.2), displays a relatively small torque, with a maximum value around  $-0.13 e \cdot \text{\AA}$ . This behavior is consistent with the corresponding region in Fig. 7.5 of the Chapter 7. In contrast, the second region, dominated by contributions from the localized  $f$ -states, reveals a torque magnitude that is 10–20 times larger than that of the  $p$ -state region, reaching values comparable to those obtained with  $U = 6.7$  eV (Fig. 7.5 b of the Chapter 7).



These observations emphasize the robustness of our findings: the  $f$ -state contributions are central to achieving pronounced orbital torque in this class of materials, and our conclusions remain valid across a reasonable range of  $U$  values.



**Fig. C.2.:** Calculated orbital torque and spin/orbital pumping for EuSSe using a reduced Hubbard interaction of  $U = 3.5$  eV. The results show that both the magnitude and qualitative behavior of the orbital torque and pumping signals are consistent with those obtained using the conventional value of  $U = 6.7$  eV. Two distinct regions are observed in the torque behavior as a function of band filling. In the low-filling region, where  $p$ -state contributions dominate (left panel), the torque reaches a relatively small maximum of approximately  $-0.13 e \cdot \text{\AA}$ , consistent with Fig. 7.5b of the Chapter 7. In the high-filling region, strong contributions from localized  $f$ -states lead to a significantly enhanced torque 10–20 times larger comparable in magnitude to the  $f$ -dominated region in Fig. 7.5 b of the Chapter 7 of the main text. These findings confirm the robustness of the orbital torque and pumping mechanisms with respect to the choice of  $U$ , and further emphasize the central role of  $f$ -state physics in driving the observed behavior.

## D.1 Journal articles related to this thesis

- **Mahmoud Zeer**, Dongwook Go, Mathias Kläui, Wulf Wulfhekel, Stefan Blügel, Yuriy Mokrousov, "**Orbital torques and orbital pumping in two-dimensional rare-earth dichalcogenides**", *npj Computational Materials*, 11, 305, (2025).
- **Zhigi Chen**, Runhan Li, Yingxi Bai, Ning Mao, **Mahmoud Zeer**, Dongwook Go, Ying Dai, Baibiao Huang, Yuriy Mokrousov, Chengwang Niu, "**Topology-Engineered Orbital Hall Effect in Two-Dimensional Ferromagnets**", *Nano Letters*, 24(16), 4826-4833, (2024).
- **Mahmoud Zeer**, Dongwook Go, Peter Schmitz, Tom Saunderson, Hao Wang, Jamal Ghabboun, Stefan Blügel, Wulf Wulfhekel, Yuriy Mokrousov, "**Promoting  $p$ -based Hall effects by  $p$ - $d$ - $f$  hybridization in Gd-based dichalcogenides**", *Physical Review Research*, 6, 013095, (2024).
- **Mahmoud Zeer**, Dongwook Go, Johanna P. Carbone, Tom G. Saunderson, Matthias Redies, Mathias Kläui, Jamal Ghabboun, Wulf Wulfhekel, Stefan Blügel, Yuriy Mokrousov, "**Spin and orbital transport in rare-earth dichalcogenides: The case of  $\text{EuS}_2$** ", *Physical Review Materials*, 6, 074004, (2022).
- **Luká Nádvořík**, Oliver Gueckstock, Lukas Braun, Chengwang Niu, Joachim Gräfe, Gunther Richter, Gisela Schütz, Hidenori Takagi, **Mahmoud Zeer**, Tom S. Seifert, Peter Kubařík, Avanindra K. Pandeya, Abdelmadjid Anane, Heejun Yang, Amílcar Bedoya-Pinto, Stuart S. P. Parkin, Martin Wolf, Yuriy Mokrousov, Hiroyuki Nakamura, Tobias Kampfrath, "**Terahertz Spin-to-Charge Current Conversion in Stacks of Ferromagnets and the Transition-Metal Dichalcogenide  $\text{NbSe}_2$** ", *Advanced Materials Interfaces*, 9(36), 2201675, (2022).

## D.2 Journal articles in preparation

- **Zhiqi Chen**, Xiaorong Zou, **Mahmoud Zeer**, Ying Dai, Baibiao Huang, Yuriy Mokrousov, Chengwang Niu, "**Quantum orbital-to-spin conversion by ferroelectric topological switch**", *Nano Letters*, 25(48), 1681716823 (2025).
- **Lishu Zhang**, **Mahmoud Zeer**, Dongwook Go, Theodoros Adamantopoulos, Peter Schmitz, Stefan Blügel, Yuriy Mokrousov, "**Orbital and spin Hall effect in Rh-doped altermagnetic RuO<sub>2</sub>**", *To be submitted*, (2025).

## Some Personal Reflections



- *A simple model is comfortable for understanding, but it does not contain the full truth.*
- *The mind is a miniature fabric of the universe, sharing its greatness and boundlessness.*
- *Love needs no reason, and hate seeks no fault.*
- *In the beginning and in the end, the human being is a creature of contradictions, most adept at claiming ideals.*
- *No one has the right to claim a pure past; even snow, if it remains on the ground long enough, becomes tainted.*
- *No matter how fierce the flames grow, they cannot burn a fresh green branch.*
- *Justice blossoms only in the garden of strength.*
- *Although life may appear unfair, no one ultimately receives less than they deserve.*

*by Mahmoud Zeer*





# Bibliography

- [1] Mark H Kryder and Chang Soo Kim. “After Hard Drives What Comes Next?” In: *IEEE Transactions on Magnetics* 45.10 (2009), pp. 3406–3413 (cit. on p. 3).
- [2] Shazia Hassan, Mamoon Asghar, et al. “Limitation of Silicon Based Computation and Future Prospects”. In: *2010 Second International Conference on Communication Software and Networks*. IEEE. 2010, pp. 559–561 (cit. on p. 3).
- [3] Youdi Gu, Zhenyi Zheng, Lanxin Jia, et al. “Ferroelectric Control of Spin-Orbitronics”. In: *Advanced Functional Materials* (2024), p. 2406444 (cit. on p. 3).
- [4] Aurelien Manchon, Hyun Cheol Koo, Junsaku Nitta, Sergey M Frolov, and Rembert A Duine. “New Perspectives for Rashba Spin-Orbit Coupling”. In: *Nature Materials* 14.9 (2015), pp. 871–882 (cit. on pp. 3, 24).
- [5] Kostya S Novoselov, Andre K Geim, Sergei V Morozov, et al. “Electric Field Effect in Atomically Thin Carbon Films”. In: *Science* 306.5696 (2004), pp. 666–669 (cit. on p. 3).
- [6] Deep Jariwala, Vinod K Sangwan, Lincoln J Lauhon, Tobin J Marks, and Mark C Hersam. “Emerging Device Applications for Semiconducting Two-Dimensional Transition Metal Dichalcogenides”. In: *ACS Nano* 8.2 (2014), pp. 1102–1120 (cit. on p. 3).
- [7] Ruitao Lv, Joshua A Robinson, Raymond E Schaak, et al. “Transition Metal Dichalcogenides and Beyond: Synthesis, Properties, and Applications of Single- and Few-Layer Nanosheets”. In: *Accounts of Chemical Research* 48.1 (2015), pp. 56–64 (cit. on p. 3).
- [8] Alexander V Kolobov and Junji Tominaga. *Two-Dimensional Transition-Metal Dichalcogenides*. Vol. 239. Springer, 2016 (cit. on p. 3).
- [9] Lei Liu, Jewook Park, David A Siegel, et al. “Heteroepitaxial Growth of Two-Dimensional Hexagonal Boron Nitride Templated by Graphene Edges”. In: *Science* 343.6167 (2014), pp. 163–167 (cit. on p. 3).
- [10] MB Tahir, Nisar Fatima, Urooj Fatima, and M Sagir. “A Review on the 2D Black Phosphorus Materials for Energy Applications”. In: *Inorganic Chemistry Communications* 124 (2021), p. 108242 (cit. on p. 3).
- [11] Sheneve Z Butler, Shawna M Hollen, Linyou Cao, et al. “Progress, Challenges, and Opportunities in Two-Dimensional Materials Beyond Graphene”. In: *ACS Nano* 7.4 (2013), pp. 2898–2926 (cit. on p. 3).



- [12] Vigneshwaran Shanmugam, Rhoda Afriyie Mensah, Karthik Babu, et al. “A Review of the Synthesis, Properties, and Applications of 2D Materials”. In: *Particle & Particle Systems Characterization* 39.6 (2022), p. 2200031 (cit. on p. 3).
- [13] Yumei Jing, Baoze Liu, Xukun Zhu, et al. “Tunable Electronic Structure of two-Dimensional Transition Metal Chalcogenides for Optoelectronic Applications”. In: *Nanophotonics* 9.7 (2020), pp. 1675–1694 (cit. on p. 3).
- [14] Qing Hua Wang, Kourosh Kalantar-Zadeh, Andras Kis, Jonathan N Coleman, and Michael S Strano. “Electronics and Optoelectronics of two-Dimensional Transition Metal Dichalcogenides”. In: *Nature Nanotechnology* 7.11 (2012), pp. 699–712 (cit. on pp. 3, 63).
- [15] Bevin Huang, Genevieve Clark, Efrén Navarro-Moratalla, et al. “Layer-Dependent Ferromagnetism in a van der Waals Crystal down to the Monolayer Limit”. In: *Nature* 546.7657 (June 2017), pp. 270–273 (cit. on p. 3).
- [16] Yujun Deng, Yijun Yu, Yichen Song, et al. “Gate-Tunable Room-Temperature Ferromagnetism in Two-Dimensional  $\text{Fe}_3\text{GeTe}_2$ ”. In: *Nature* 563.7729 (Oct. 2018), pp. 94–99 (cit. on p. 3).
- [17] Cheng Gong, Lin Li, Zhenglu Li, et al. “Discovery of Intrinsic Ferromagnetism in Two-Dimensional van der Waals Crystals”. In: *Nature* 546.7657 (Apr. 2017), pp. 265–269 (cit. on p. 3).
- [18] Fengfeng Zhu, Lichuan Zhang, Xiao Wang, et al. “Topological Magnon Insulators in Two-Dimensional van der Waals Ferromagnets  $\text{CrSiTe}_3$  and  $\text{CrGeTe}_3$  : Toward Intrinsic Gap-Tunability”. In: *Science Advances* 7.37 (2021), eabi7532 (cit. on p. 3).
- [19] Hui Pan. “Magnetic and Electronic Evolutions of Hydrogenated  $\text{VTe}_2$  Monolayer under Tension”. In: *Scientific Reports* 4 (2014), p. 7524 (cit. on p. 4).
- [20] Yandong Ma, Ying Dai, Meng Guo, et al. “Evidence of the Existence of Magnetism in Pristine  $\text{VX}_2$  Monolayers ( $\text{X}=\text{S}, \text{Se}$ ) and their Strain-Induced Tunable Magnetic Properties”. In: *ACS Nano* 6.2 (2012). PMID: 22264067, pp. 1695–1701 (cit. on pp. 4, 72).
- [21] Wei Chen, Jian-min Zhang, Yao-zhuang Nie, Qing-lin Xia, and Guang-hua Guo. “Electronic Structure and Magnetism of  $\text{MTe}_2$  ( $\text{M}=\text{Ti}, \text{V}, \text{Cr}, \text{Mn}, \text{Fe}, \text{Co}$  and  $\text{Ni}$ )”. In: *Journal of Magnetism and Magnetic Materials* 508 (2020), p. 166878 (cit. on p. 4).
- [22] Hailong Wang, Keng-Yuan Meng, Pengxiang Zhang, et al. “Large Spin-Orbit Torque Observed in Epitaxial  $\text{SrIrO}_3$  Thin Films”. In: *Applied Physics Letters* 114.23 (2019), p. 232406 (cit. on pp. 4, 114).
- [23] H. Y. Lv, W. J. Lu, D. F. Shao, Y. Liu, and Y. P. Sun. “Strain-Controlled Switch between Ferromagnetism and Antiferromagnetism in  $1\text{T-CrX}_2$  ( $\text{X}=\text{Se}, \text{Te}$ ) Monolayers”. In: *Physical Review B* 92 (21 Dec. 2015), p. 214419 (cit. on p. 4).
- [24] C. Ataca, H. ahin, and S. Ciraci. “Stable, Single-Layer  $\text{MX}_2$  Transition-Metal Oxides and Dichalcogenides in a Honeycomb-Like Structure”. In: *The Journal of Physical Chemistry C* 116.16 (2012), pp. 8983–8999. eprint: <https://doi.org/10.1021/jp212558p> (cit. on pp. 4, 72).

- [25] Min Kan, Subash Adhikari, and Qiang Sun. “Ferromagnetism in  $\text{MnX}_2$  ( $\text{X} = \text{s, se}$ ) Monolayers”. In: *Physical Chemistry Chemical Physics* 16.10 (2014), pp. 4990–4994 (cit. on pp. 4, 72).
- [26] Bing Wang, Xiwen Zhang, Yehui Zhang, et al. “Prediction of a Two-Dimensional High-TC f-Electron Ferromagnetic Semiconductor”. In: *Materials Horizons* 7.6 (2020), pp. 1623–1630 (cit. on pp. 4, 33).
- [27] Weiqi Liu, Junwei Tong, Li Deng, et al. “Two-dimensional Ferromagnetic Semiconductors of Rare-Earth Monolayer  $\text{GdX}_2$  ( $\text{X} = \text{Cl, Br, I}$ ) with Large Perpendicular Magnetic Anisotropy and High Curie Temperature”. In: *Materials Today Physics* 21 (2021), p. 100514 (cit. on pp. 4, 33).
- [28] Kang Sheng, Hong-Kuan Yuan, and Zhi-Yong Wang. “Monolayer Gadolinium Halides,  $\text{GdX}_2$  ( $\text{X} = \text{F, Cl, Br}$ ): Intrinsic Ferrovalley Materials with Spontaneous Spin and Valley Polarizations”. In: *Physical Chemistry Chemical Physics* 24.6 (2022), pp. 3865–3874 (cit. on pp. 4, 33).
- [29] Haipeng You, Yang Zhang, Jun Chen, et al. “Peierls Transition Driven Ferroelasticity in the Two-Dimensional  $d$ – $f$  Hybrid Magnets”. In: *Physical Review B* 103 (16 Apr. 2021), p. L161408 (cit. on pp. 4, 33).
- [30] Spiridon D. Pappas, Panagiotis Pouloupoulos, Björn Lewitz, et al. “Direct Evidence for Significant Spin-Polarization of EuS in Co/EuS Multilayers at Room Temperature”. In: *Scientific Reports* 3 (2013), p. 1333 (cit. on pp. 4, 81).
- [31] P. Pouloupoulos, A. Goschew, V. Kapaklis, et al. “Induced Spin-Polarization of EuS at Room Temperature in Ni/EuS Multilayers”. In: *Applied Physics Letters* 104.11 (2014), p. 112411 (cit. on pp. 4, 81).
- [32] Sebastian M. J. Beer, Arbresha Muriqi, Patrick Lindner, et al. “Ferromagnetic Europium Eulfide Thin Films: Influence of Precursors on Magneto-Optical Properties”. In: *Chemistry of Materials* 34.1 (2022), pp. 152–164. eprint: <https://doi.org/10.1021/acs.chemmater.1c02958> (cit. on pp. 4, 81).
- [33] C. J. P. Smits, A. T. Filip, J. T. Kohlhepp, et al. “Magnetic and Structural Properties of EuS for Magnetic Tunnel Junction Barriers”. In: *Journal of Applied Physics* 95.11 (2004), pp. 7405–7407 (cit. on pp. 4, 81).
- [34] A. T. Filip, P. LeClair, C. J. P. Smits, et al. “Spin-Injection Device Based on EuS Magnetic Tunnel Barriers”. In: *Applied Physics Letters* 81.10 (2002), pp. 1815–1817 (cit. on pp. 4, 81).
- [35] Jerry A. Cowen, P. Michlin, Jennifer Kraus, et al. “ $\text{EuSe}_2$ : A novel Antiferromagnetic Rare-Earth Polychalcogenide”. In: *Journal of Applied Physics* 85 (1999), pp. 5381–5383 (cit. on pp. 4, 81).
- [36] Junjie Yin, Changwei Wu, Lisi Li, et al. “Large Negative Magnetoresistance in the Antiferromagnetic Rare-Earth Dichalcogenide  $\text{EuTe}_2$ ”. In: *Physical Review Materials* 4 (1 Jan. 2020), p. 013405 (cit. on pp. 4, 81).
- [37] Qiming Shao, Peng Li, Luqiao Liu, et al. “Roadmap of Spin–Orbit Torques”. In: *IEEE Transactions on Magnetics* 57.7 (2021), pp. 1–39 (cit. on pp. 4, 24).





- [38] D Jo, D Go, GM Choi, and HW Lee. “Spintronics Meets Orbitronics: Emergence of Orbital Angular Momentum in Solids. npj Spintronics 2”. In: (2024) (cit. on p. 4).
- [39] Ping Wang, Feng Chen, Yuhe Yang, et al. “Orbitronics: Mechanisms, Materials and Devices”. In: *Advanced Electronic Materials* (2024), p. 2400554 (cit. on p. 4).
- [40] Yong Xu, Fan Zhang, Albert Fert, et al. “Orbitronics: Light-Induced Orbital Currents in Ni Studied by Terahertz Emission Experiments”. In: *Nature Communications* 15.1 (2024), p. 2043 (cit. on p. 4).
- [41] T. Tanaka, H. Kontani, M. Naito, et al. “Intrinsic Spin Hall Effect and Orbital Hall Effect in 4d and 5d Transition Metals”. In: *Physical Review B* 77 (16 Apr. 2008), p. 165117 (cit. on pp. 4, 21, 69, 78).
- [42] Dongwook Go, Daegeun Jo, Changyoung Kim, and Hyun-Woo Lee. “Intrinsic Spin and Orbital Hall Effects from Orbital Texture”. In: *Physical Review Letters* 121 (8 Aug. 2018), p. 086602 (cit. on pp. 4, 20–23, 69, 77).
- [43] Dongwook Go and Hyun-Woo Lee. “Orbital torque: Torque Generation by Orbital Current Injection”. In: *Physical review research* 2.1 (2020), p. 013177 (cit. on pp. 4, 5, 25, 28).
- [44] Hiroki Hayashi, Daegeun Jo, Dongwook Go, et al. “Observation of Long-Range Orbital Transport and Giant Orbital Torque”. In: *Communications Physics* 6.1 (2023), p. 32 (cit. on pp. 4, 29).
- [45] Dongwook Go, Daegeun Jo, Kyoung-Whan Kim, et al. “Long-Range Orbital Torque by Momentum-Space Hotspots”. In: *Physical review letters* 130.24 (2023), p. 246701 (cit. on pp. 4, 22).
- [46] Dongjoon Lee, Dongwook Go, Hyeon-Jong Park, et al. “Orbital Torque in Magnetic Bilayers”. In: *Nature Communications* 12.1 (2021), p. 6710 (cit. on p. 4).
- [47] Pietro Gambardella and Ioan Mihai Miron. “Current-Induced Spin–Orbit Torques”. In: *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences* 369.1948 (2011), pp. 3175–3197 (cit. on p. 4).
- [48] B. Andrei Bernevig, Taylor L. Hughes, and Shou-Cheng Zhang. “Orbitronics: The Intrinsic Orbital Current in *p*-Doped Silicon”. In: *Physical Review Letters* 95 (6 Aug. 2005), p. 066601 (cit. on pp. 5, 69).
- [49] Sayantika Bhowal and S. Satpathy. “Intrinsic Orbital and Spin Hall Effects in Monolayer Transition Metal Dichalcogenides”. In: *Physical Review B* 102 (3 July 2020), p. 035409 (cit. on p. 5).
- [50] Sayantika Bhowal and S. Satpathy. “Intrinsic Orbital Moment and Prediction of a Large Orbital Hall Effect in Two-Dimensional Transition Metal Dichalcogenides”. In: *Physical Review B* 101 (12 Mar. 2020), p. 121112 (cit. on p. 5).
- [51] Tarik P Cysne, Marcio Costa, Luis M Canonico, et al. “Disentangling Orbital and Valley Hall Effects in Bilayers of Transition Metal Dichalcogenides”. In: *Physical Review Letters* 126.5 (2021), p. 056601 (cit. on pp. 5, 7, 21, 51, 52, 54, 55, 63, 69).

- [52] I. V. Tokatly. “Orbital Momentum Hall Effect in  $p$ -Doped Graphane”. In: *Physical Review B* 82 (16 Oct. 2010), p. 161404 (cit. on p. 5).
- [53] Sayantika Bhowal and Giovanni Vignale. “Orbital Hall Effect as an Alternative to Valley Hall Effect in Gapped Graphene”. In: *Physical Review B* 103 (19 May 2021), p. 195309 (cit. on p. 5).
- [54] Sayantika Bhowal and Sashi Satpathy. “Intrinsic Orbital and Spin Hall Effects in Monolayer Transition Metal Dichalcogenides”. In: *Physical Review B* 102.3 (2020), p. 035409 (cit. on pp. 5, 23, 63).
- [55] Rajagopalan Ramaswamy, Jong Min Lee, Kaiming Cai, and Hyunsoo Yang. “Recent Advances in Spin-Orbit Torques: Moving Towards Device Applications”. In: *Applied Physics Reviews* 5.3 (2018), p. 031107 (cit. on p. 5).
- [56] Idris Smaili, Slimane Laref, Jose H Garcia, et al. “Janus Monolayers of Magnetic Transition Metal Dichalcogenides as an all-in-one Platform for Spin-Orbit Torque”. In: *Physical Review B* 104.10 (2021), p. 104415 (cit. on pp. 5, 101).
- [57] Shilei Ding, Andrew Ross, Dongwook Go, et al. “Harnessing Orbital-to-Spin Conversion of Interfacial Orbital Currents for Efficient Spin-Orbit Torques”. In: *Physical Review Letters* 125.17 (2020), p. 177201 (cit. on p. 5).
- [58] Shilei Ding, Zhongyu Liang, Dongwook Go, et al. “Observation of the Orbital Rashba-Edelstein Magnetoresistance”. In: *Physical Review Letters* 128.6 (2022), p. 067201 (cit. on p. 5).
- [59] Tom G Saunderson, Dongwook Go, Stefan Blügel, Mathias Kläui, and Yuriy Mokrousov. “Hidden Interplay of Current-Induced Spin and Orbital Torques in Bulk Fe<sub>3</sub>GeTe<sub>2</sub>”. In: *Physical Review Research* 4.4 (2022), p. L042022 (cit. on pp. 5, 101, 103).
- [60] Soogil Lee, Min-Gu Kang, Dongwook Go, et al. “Efficient Conversion of Orbital Hall Current to Spin Current for Spin-Orbit Torque Switching”. In: *Communications Physics* 4 (2021), pp. 1–6 (cit. on p. 5).
- [61] Dongwook Go, Frank Freimuth, Jan-Philipp Hanke, et al. “Theory of Current-Induced Angular Momentum Transfer Dynamics in Spin-Orbit Coupled Systems”. In: *Physical Review Research* 2 (3 Sept. 2020), p. 033401 (cit. on p. 5).
- [62] Dongwook Go, Daegun Jo, Tenghua Gao, et al. “Orbital Rashba Effect in a Surface-Oxidized Cu Film”. In: *Physical Review B* 103 (12 Mar. 2021), p. L121113 (cit. on pp. 5, 77).
- [63] Di Xiao, Ming-Che Chang, and Qian Niu. “Berry Phase Effects on Electronic Properties”. In: *Reviews of Modern Physics* 82.3 (2010), pp. 1959–2007 (cit. on pp. 6, 12, 16).
- [64] Robert M White, Robert M White, and Bradford Bayne. *Quantum Theory of Magnetism*. Vol. 1. Springer, 1983 (cit. on p. 9).
- [65] Marie-Catherine Desjonquères, Cyrille Barreteau, Gabriel Autes, and Daniel Spanjaard. “Orbital Contribution to the Magnetic Properties of Iron as a Function of Dimensionality”. In: *Physical Review B Condensed Matter and Materials Physics* 76.2 (2007), p. 024412 (cit. on p. 9).



- [66] AR Mackintosh and OK Andersen. *Electrons at the Fermi Surface*. 1980, p. 149 (cit. on p. 12).
- [67] DD Koelling and BN Harmon. “A Technique for Relativistic Spin-Polarised Calculations”. In: *Journal of Physics C: Solid State Physics* 10.16 (1977), p. 3107 (cit. on p. 12).
- [68] Naoto Nagaosa, Jairo Sinova, Shigeki Onoda, Allan H MacDonald, and Nai Phuan Ong. “Anomalous Hall Effect”. In: *Reviews of modern physics* 82.2 (2010), p. 1539 (cit. on pp. 12, 17, 19).
- [69] Michael Victor Berry. “Quantal Phase Factors Accompanying Adiabatic Changes”. In: *Proceedings of the Royal Society of London. A. Mathematical and Physical Sciences* 392.1802 (1984), pp. 45–57 (cit. on p. 12).
- [70] Alfred Shapere and Frank Wilczek. *Geometric Phases in Physics*. Vol. 5. World scientific, 1989 (cit. on p. 13).
- [71] Arno Bohm, Ali Mostafazadeh, Hiroyasu Koizumi, Qian Niu, and Josef Zwanziger. *The Geometric Phase in Quantum Systems: Foundations, Mathematical Concepts, and Applications in Molecular and Condensed Matter Physics*. Springer Science & Business Media, 2013 (cit. on p. 13).
- [72] Jan-Philipp Hanke. “Topological Properties of Complex Magnets from an Advanced ab-initio Wannier Description”. PhD thesis. Dissertation, RWTH Aachen University, 2018, 2018 (cit. on pp. 14, 15).
- [73] Yuriy Mokrousov and Frank Freimuth. *Geometric Phases and Topological Effects*. 2014. arXiv: 1407.2847 [cond-mat.mes-hall] (cit. on p. 16).
- [74] Edwin Herbert Hall. “XVIII. On the Rotational Coefficient in Nickel and Cobalt”. In: *The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science* 12.74 (1881), pp. 157–172 (cit. on pp. 17, 22).
- [75] Yugui Yao, Leonard Kleinman, AH MacDonald, et al. “First principles Calculation of Anomalous Hall Conductivity in Ferromagnetic bcc Fe”. In: *Physical review letters* 92.3 (2004), p. 037204 (cit. on pp. 18, 22).
- [76] Mikhail I Dyakonov. “Possibility of Orienting Electron Spins with Current”. In: *JETP Lett. USSR* 13 (1971), p. 467 (cit. on p. 18).
- [77] Mikhail I Dyakonov and VI Perel. “Current-Induced Spin Orientation of Electrons in Semiconductors”. In: *Physics Letters A* 35.6 (1971), pp. 459–460 (cit. on p. 18).
- [78] JE Hirsch. “Spin Hall Effect”. In: *Physical Review Letters* 83.9 (1999), p. 1834 (cit. on p. 18).
- [79] Takashi Kimura, Y Otani, T Sato, S Takahashi, and S Maekawa. “Room-Temperature Reversible Spin Hall Effect”. In: *Physical Review Letters* 98.15 (2007), p. 156601 (cit. on p. 19).
- [80] Sergio O Valenzuela and M Tinkham. “Direct Electronic Measurement of the Spin Hall Effect”. In: *Nature* 442.7099 (2006), pp. 176–179 (cit. on p. 19).

- [81] Laurent Vila, Takashi Kimura, and YoshiChika Otani. “Evolution of the Spin Hall Effect in Pt Nanowires: Size and Temperature Effects”. In: *Physical Review Letters* 99.22 (2007), p. 226604 (cit. on p. 19).
- [82] Takeshi Seki, Yu Hasegawa, Seiji Mitani, et al. “Giant Spin Hall Effect in Perpendicularly Spin-Polarized FePt/Au Devices”. In: *Nature Materials* 7.2 (2008), pp. 125–129 (cit. on p. 19).
- [83] J Smit. “The Spontaneous Hall Effect in Ferromagnetics I”. In: *Physica* 21.6-10 (1955), pp. 877–887 (cit. on p. 19).
- [84] Jan Smit. “The Spontaneous Hall Effect in Ferromagnetics II”. In: *Physica* 24.1-5 (1958), pp. 39–51 (cit. on p. 19).
- [85] Jairo Sinova, Dimitrie Culcer, Qian Niu, et al. “Universal Intrinsic Spin Hall Effect”. In: *Physical Review Letters* 92.12 (2004), p. 126603 (cit. on p. 19).
- [86] NA Sinitsyn. “Semiclassical Theories of the Anomalous Hall Effect”. In: *Journal of Physics: Condensed Matter* 20.2 (2007), p. 023201 (cit. on p. 19).
- [87] T Jungwirth, Qian Niu, and AH MacDonald. “Anomalous Hall Effect in Ferromagnetic Semiconductors”. In: *Physical review letters* 88.20 (2002), p. 207208 (cit. on p. 19).
- [88] Tarik P. Cysne, Sayantika Bhowal, Giovanni Vignale, and Tatiana G. Rappoport. “Orbital Hall Effect in Bilayer Transition Metal Dichalcogenides: From the intratomic Approximation to the Bloch States Orbital Magnetic Moment Approach”. In: *Physical Review B* 105 (19 May 2022), p. 195421 (cit. on pp. 21, 69).
- [89] Xinjie Wang, David Vanderbilt, Jonathan R Yates, and Ivo Souza. “Fermi-Surface Calculation of the Anomalous Hall Conductivity”. In: *Physical Review B Condensed Matter and Materials Physics* 76.19 (2007), p. 195109 (cit. on p. 22).
- [90] Guang-Yu Guo, Shuichi Murakami, T-W Chen, and Naoto Nagaosa. “Intrinsic Spin Hall Effect in Platinum: First-Principles Calculations”. In: *Physical review letters* 100.9 (2008), p. 096401 (cit. on p. 22).
- [91] Ahmet Avsar, Jong Hak Lee, Gavin Kok Wai Koon, and Barbaros Özyilmaz. “Enhanced Spin–Orbit Coupling in Dilute Fluorinated Graphene”. In: *2D Materials* 2.4 (2015), p. 044009 (cit. on p. 22).
- [92] Jayakumar Balakrishnan, Gavin Kok Wai Koon, Ahmet Avsar, et al. “Giant Spin Hall Effect in Graphene Grown by Chemical Vapour Deposition”. In: *Nature Communications* 5.1 (2014), p. 4748 (cit. on p. 22).
- [93] Young-Gwan Choi, Daegun Jo, Kyung-Hun Ko, et al. “Observation of the Orbital Hall Effect in a Light Metal Ti”. In: *Nature* 619.7968 (2023), pp. 52–56 (cit. on pp. 22, 23).
- [94] Luis M Canonico, Tarik P Cysne, Alejandro Molina-Sanchez, RB Muniz, and Tatiana G Rappoport. “Orbital Hall Insulating Phase in Transition Metal Dichalcogenide Monolayers”. In: *Physical Review B* 101.16 (2020), p. 161409 (cit. on pp. 22, 54, 63, 69, 77).



- [95] Dongwook Go, Daegeun Jo, Hyun-Woo Lee, Mathias Kläui, and Yuriy Mokrousov. “Orbitronics: Orbital Currents in Solids”. In: *Europhysics Letters* 135.3 (2021), p. 37001 (cit. on p. 23).
- [96] Sayantika Bhowal and Sashi Satpathy. “Intrinsic Orbital Moment and Prediction of a Large Orbital Hall Effect in Two-Dimensional Transition Metal Dichalcogenides”. In: *Physical Review B* 101.12 (2020), p. 121112 (cit. on p. 24).
- [97] Daniel C Ralph and Mark D Stiles. “Spin Transfer Torques”. In: *Journal of Magnetism and Magnetic Materials* 320.7 (2008), pp. 1190–1216 (cit. on p. 24).
- [98] Cheng Song, Ruiqi Zhang, Liyang Liao, et al. “Spin-Orbit Torques: Materials, Mechanisms, Performances, and Potential Applications”. In: *Progress in Materials Science* 118 (2021), p. 100761 (cit. on p. 24).
- [99] Mikhail I Dyakonov and VI Perel. “Current-Induced Spin Orientation of Electrons in Semiconductors”. In: *Physics Letters A* 35.6 (1971), pp. 459–460 (cit. on p. 24).
- [100] Victor M Edelstein. “Spin Polarization of Conduction Electrons Induced by Electric Current in Two-Dimensional Asymmetric Electron Systems”. In: *Solid State Communications* 73.3 (1990), pp. 233–235 (cit. on p. 24).
- [101] Daegeun Jo, Dongwook Go, and Hyun-Woo Lee. “Gigantic Intrinsic Orbital Hall effects in weakly Spin-Orbit Coupled Metals”. In: *Physical Review B* 98.21 (2018), p. 214405 (cit. on p. 24, 69).
- [102] Thomas L Gilbert. “A Phenomenological Theory of Damping in Ferromagnetic Materials”. In: *IEEE transactions on magnetics* 40.6 (2004), pp. 3443–3449 (cit. on p. 25).
- [103] Aurélien Manchon and Shufeng Zhang. “Theory of spin torque due to spin-orbit coupling”. In: *Physical Review B* 79.9 (2009), p. 094422 (cit. on p. 25).
- [104] Shilei Ding, Andrew Ross, Dongwook Go, et al. “Harnessing Orbital-to-Spin Conversion of Interfacial Orbital Currents for Efficient Spin-Orbit Torques”. In: *Physical review letters* 125 (17 Oct. 2020), p. 177201 (cit. on p. 26).
- [105] Shilei Ding, Zhongyu Liang, Dongwook Go, et al. “Observation of the Orbital Rashba-Edelstein Magnetoresistance”. In: *Physical review letters* 128 (6 Feb. 2022), p. 067201 (cit. on p. 26).
- [106] Dongwook Go, Frank Freimuth, Jan-Philipp Hanke, et al. “Theory of Current-Induced Angular Momentum Transfer Dynamics in Spin-Orbit Coupled Systems”. In: *Physical Review Research* 2 (3 Sept. 2020), p. 033401 (cit. on pp. 26, 49, 103).
- [107] Dongwook Go, Kazuya Ando, Armando Pezo, et al. “Orbital Pumping by Magnetization Dynamics in Ferromagnets”. In: *Physical Review B* 111 (14 Apr. 2025), p. L140409 (cit. on p. 29).
- [108] Seungyun Han, Hye-Won Ko, Jung Hyun Oh, et al. “Orbital Pumping Incorporating Both Orbital Angular Momentum and Position”. In: *Physical review letters* 134 (3 Jan. 2025), p. 036305 (cit. on p. 29).

- [109] Paul M Haney and Mark D Stiles. “Current-Induced Torques in the Presence of Spin-Orbit Coupling”. In: *Physical Review Letters* 105.12 (2010), p. 126602 (cit. on p. 29).
- [110] Dongwook Go, Kazuya Ando, Armando Pezo, et al. “Orbital pumping by magnetization dynamics in ferromagnets”. In: *Physical Review B* 111.14 (2025), p. L140409 (cit. on p. 29).
- [111] Frank Freimuth, Stefan Blügel, and Yuriy Mokrousov. “Direct and Inverse Spin-Orbit Torques”. In: *Physical Review B* 92.6 (2015), p. 064415 (cit. on pp. 29, 118).
- [112] Ka Shen, Giovanni Vignale, and Roberto Raimondi. “Microscopic Theory of the Inverse Edelstein Effect”. In: *Physical Review Letters* 112.9 (2014), p. 096601 (cit. on pp. 31, 121).
- [113] Frank Freimuth, Stefan Blügel, and Yuriy Mokrousov. “Charge Pumping Driven by the Laser-Induced Dynamics of the Exchange Splitting”. In: *Physical Review B* 95 (9 Mar. 2017), p. 094434 (cit. on p. 31).
- [114] Yi Zhang, Tay-Rong Chang, Bo Zhou, et al. “Direct Observation of the Transition from Indirect to Direct Bandgap in Atomically Thin Epitaxial MoSe<sub>2</sub>”. In: *Nature Nanotechnology* 9.2 (2014), pp. 111–115 (cit. on p. 31).
- [115] Kin Fai Mak, Changgu Lee, James Hone, Jie Shan, and Tony F Heinz. “Atomically Thin MoS<sub>2</sub>: A New Direct-Gap Semiconductor”. In: *Physical Review Letters* 105.13 (2010), p. 136805 (cit. on p. 31).
- [116] Humberto R Gutiérrez, Nestor Perea-López, Ana Laura Elías, et al. “Extraordinary Room-Temperature Photoluminescence in Triangular WS<sub>2</sub> Monolayers”. In: *Nano Letters* 13.8 (2013), pp. 3447–3454 (cit. on pp. 31, 51).
- [117] Hui Zhang, Li-Min Liu, and Woon-Ming Lau. “Dimension-Dependent Phase Transition and Magnetic Properties of VS<sub>2</sub>”. In: *Journal of Materials Chemistry A* 1.36 (2013), pp. 10821–10828 (cit. on p. 31).
- [118] N David Mermin and Herbert Wagner. “Absence of Ferromagnetism or Antiferromagnetism in one-or Two-Dimensional Isotropic Heisenberg Models”. In: *Physical Review Letters* 17.22 (1966), p. 1133 (cit. on p. 32).
- [119] Jose L Lado and Joaquín Fernández-Rossier. “On the Origin of Magnetic Anisotropy in Two Dimensional CrI<sub>3</sub>”. In: *2D Materials* 4.3 (2017), p. 035002 (cit. on p. 32).
- [120] Bevin Huang, Genevieve Clark, Efrén Navarro-Moratalla, et al. “Layer-Dependent Ferromagnetism in a van der Waals Crystal down to the Monolayer Limit”. In: *Nature* 546.7657 (2017), pp. 270–273 (cit. on p. 32).
- [121] Haiyu Wang, Yingjie Liu, Peichen Wu, et al. “Above Room-Temperature Ferromagnetism in Wafer-Scale Two-Dimensional van der Waals Fe<sub>3</sub>GeTe<sub>2</sub> Tailored by a Topological Insulator”. In: *ACS Nano* 14.8 (2020), pp. 10045–10053 (cit. on p. 32).
- [122] Thomas Olsen. “Magnetic Anisotropy and Exchange Interactions of Two-Dimensional FePS<sub>3</sub>, NiPS<sub>3</sub> and MnPS<sub>3</sub> from First Principles Calculations”. In: *Journal of Physics D: Applied Physics* 54.31 (2021), p. 314001 (cit. on p. 32).



- [123] Wei Yu, Jing Li, Tun Seng Herng, et al. “Chemically Exfoliated VSe2 Monolayers with Room-Temperature Ferromagnetism”. In: *Advanced Materials* 31.40 (2019), p. 1903779 (cit. on p. 32).
- [124] Ping Kwan Johnny Wong, Wen Zhang, Fabio Bussolotti, et al. “Evidence of Spin Frustration in a Vanadium Diselenide Monolayer Magnet”. In: *Advanced Materials* 31.23 (2019), p. 1901185 (cit. on p. 32).
- [125] E. Schrödinger. “An Undulatory Theory of the Mechanics of Atoms and Molecules”. In: *Physical Review* 28 (6 Dec. 1926), pp. 1049–1070 (cit. on p. 35).
- [126] Paul Adrien Maurice Dirac and Ralph Howard Fowler. “The Quantum Theory of the Electron”. In: *Proceedings of the Royal Society of London. Series A, Containing Papers of a Mathematical and Physical Character* 117.778 (1928), pp. 610–624 (cit. on p. 35).
- [127] Gregor Michalíček. “Extending the Precision and Efficiency of the All-Electron Full-Potential Linearized Augmented Plane-Wave Density-Functional Theory Method”. englisch. PhD thesis. Jülich: Zugleich: Dissertation, Rheinisch-Westfälische Technische Hochschule Aachen, 2014, 2015, 195 Seiten (cit. on pp. 35, 45).
- [128] Richard M. Martin. *Electronic structure: Basic Theory and Practical Methods*. Cambridge University Press, 2004 (cit. on p. 35).
- [129] Jan-Philipp Hanke. “Topological Properties of Complex Magnets from an Advanced ab-initio Wannier Description”. englisch. PhD thesis. Jülich: Zugleich: Dissertation, Rheinisch-Westfälische Technische Hochschule Aachen, 2018, 2018, XI, 173 Seiten (cit. on p. 35).
- [130] Markus Betzinger. “Orbital-Dependent Exchange-Correlation Functionals in Density-Functional Theory Realized by the FLAPW Method”. englisch. PhD thesis. Jülich: Zugleich: Dissertation, Rheinisch-Westfälische Technische Hochschule Aachen, 2011, 2013, VI, 173 Seiten (cit. on p. 35).
- [131] Philipp Kurz. “Non-Collinear Magnetism at Surfaces and in Ultrathin Films [E-Book]”. englisch. PhD thesis. Jülich: Zugleich: Dissertation, Rheinisch-Westfälische Technische Hochschule Aachen, 2000, 2000, 1 Online-Ressource (IV, 207 Seiten) (cit. on p. 35).
- [132] Matthias Redies. “High-Performance Computing Approach to Hybrid Functionals in the all-electron DFT Code FLEUR”. Veröffentlicht auf dem Publikationsserver der RWTH Aachen University; Dissertation, RWTH Aachen University, 2022. Dissertation. Aachen: RWTH Aachen University, 2022, 1 Online-Ressource : Illustrationen (cit. on p. 35).
- [133] Johanna Paulina Carbone. “Rare-Earth Atoms on Two-Dimensional Materials: *ab initio* investigation of magnetic properties”. Druckausgabe: 2024. - Onlineausgabe: 2024. - Auch veröffentlicht auf dem Publikationsserver der RWTH Aachen University 2024; Dissertation, RWTH Aachen University, 2023. Dissertation. Jülich: RWTH Aachen University, 2024, 1 Online-Ressource : Illustrationen (cit. on p. 35).

- [134] M. Born and R. Oppenheimer. “Zur Quantentheorie der Molekeln”. In: *Annalen der Physik* 389.20 (1927), pp. 457–484. eprint: <https://onlinelibrary.wiley.com/doi/pdf/10.1002/andp.19273892002> (cit. on p. 36).
- [135] P. Hohenberg and W. Kohn. “Inhomogeneous Electron Gas”. In: *Physical Review* 136 (3B Nov. 1964), B864–B871 (cit. on p. 37).
- [136] W. Kohn and L. J. Sham. “Self-Consistent Equations Including Exchange and Correlation Effects”. In: *Physical Review* 140 (4A Nov. 1965), A1133–A1138 (cit. on pp. 38, 40).
- [137] Robert O Jones. “Density Functional Theory: Its Origins, Rise to Prominence, and Future”. In: *Reviews of modern physics* 87.3 (2015), p. 897 (cit. on p. 40).
- [138] LJ Sham, PM Marcus, JF Janak, and AR Williams. “Computational Methods in Band Theory”. In: *Plenum (New York)* (1971), p. 458 (cit. on p. 40).
- [139] John P Perdew, Kieron Burke, and Matthias Ernzerhof. “Generalized Gradient Approximation Made Simple”. In: *Physical review letters* 77.18 (1996), p. 3865 (cit. on pp. 40, 41, 52, 71, 84, 104).
- [140] J. P. Perdew and Alex Zunger. “Self-Interaction Correction to Density-Functional Approximations for Many-Electron Systems”. In: *Physical Review B* 23 (10 May 1981), pp. 5048–5079 (cit. on p. 41).
- [141] Ulf Von Barth and Lars Hedin. “A local Exchange-Correlation Potential for the Spin Polarized Case. i”. In: *Journal of Physics C: Solid State Physics* 5.13 (1972), p. 1629 (cit. on p. 41).
- [142] Burak Himmetoglu, Andrea Floris, Stefano De Gironcoli, and Matteo Cococcioni. “Hubbard-Corrected DFT Energy Functionals: The LDA+ U Description of Correlated Systems”. In: *International Journal of Quantum Chemistry* 114.1 (2014), pp. 14–49 (cit. on p. 43).
- [143] John P Perdew. “Density Functional Theory and the Band Gap Problem”. In: *International Journal of Quantum Chemistry* 28.S19 (1985), pp. 497–523 (cit. on p. 43).
- [144] A Rohrbach, J Hafner, and G Kresse. “Electronic Correlation Effects in Transition-Metal Sulfides”. In: *Journal of Physics: Condensed Matter* 15.6 (2003), p. 979 (cit. on p. 43).
- [145] AM Oles and G Stollhoff. “Correlation Effects in Ferromagnetism of Transition Metals”. In: *Physical Review B* 29.1 (1984), p. 314 (cit. on p. 43).
- [146] John Hubbard. “Electron Correlations in Narrow Energy Bands-IV. The Atomic Representation”. In: *Proceedings of the Royal Society of London. Series A. Mathematical and Physical Sciences* 285.1403 (1965), pp. 542–560 (cit. on p. 44).
- [147] AG Petukhov, II Mazin, Liviu Chioncel, and AI Lichtenstein. “Correlated Metals and the LDA+ U Method”. In: *Physical Review B* 67.15 (2003), p. 153106 (cit. on p. 44).





- [148] Sergei L Dudarev, Gianluigi A Botton, Sergey Y Savrasov, CJ Humphreys, and Adrian P Sutton. “Electron-Energy-Loss Spectra and the Structural Stability of Nickel Oxide: An LSDA+ U Study”. In: *Physical Review B* 57.3 (1998), p. 1505 (cit. on p. 44).
- [149] IV Solovyev, PH Dederichs, and VI Anisimov. “Corrected Atomic Limit in the Local-Density Approximation and the Electronic Structure of d Impurities in Rb”. In: *Physical Review B* 50.23 (1994), p. 16861 (cit. on p. 44).
- [150] Stefan Blügel and Gustav Bihlmayer. “Full-Potential Linearized Augmented Planewave Method”. In: *Computational nanoscience: do it yourself* 31 (2006), pp. 85–129 (cit. on p. 45).
- [151] David J Singh and Lars Nordstrom. *Planewaves, Pseudopotentials, and the LAPW method*. Springer Science & Business Media, 2006 (cit. on p. 45).
- [152] E. Wimmer, H. Krakauer, M. Weinert, and A. J. Freeman. “Full-potential Self-Consistent Linearized-Augmented-Plane-Wave Method for Calculating the Electronic Structure of Molecules and Surfaces: O<sub>2</sub> Molecule”. In: *Physical Review B* 24 (2 July 1981), pp. 864–875 (cit. on pp. 45, 52, 71, 84).
- [153] O Krogh Andersen. “Linear Methods in Band Theory”. In: *Physical Review B* 12.8 (1975), p. 3060 (cit. on p. 46).
- [154] Daniel Wortmann, Gregor Michalick, Nadjib Baadji, et al. *FLEUR*. Zenodo. May 2023 (cit. on pp. 46, 49, 50, 52, 84).
- [155] Gregory H Wannier. “The Structure of Electronic Excitation Levels in Insulating Crystals”. In: *Physical Review* 52.3 (1937), p. 191 (cit. on p. 47).
- [156] Nicola Marzari, Arash A Mostofi, Jonathan R Yates, Ivo Souza, and David Vanderbilt. “Maximally Localized Wannier Functions: Theory and Applications”. In: *Reviews of Modern Physics* 84.4 (2012), p. 1419 (cit. on p. 47).
- [157] Jacques Des Cloizeaux. “Orthogonal Orbitals and Generalized Wannier Functions”. In: *Physical Review* 129.2 (1963), p. 554 (cit. on p. 47).
- [158] Nicola Marzari and David Vanderbilt. “Maximally Localized Generalized Wannier Functions for Composite Energy Bands”. In: *Physical Review B* 56.20 (1997), p. 12847 (cit. on p. 48).
- [159] Arash A Mostofi, Jonathan R Yates, Young-Su Lee, et al. “Wannier90: A Tool for Obtaining Maximally-Localised Wannier Functions”. In: *Computer physics communications* 178.9 (2008), pp. 685–699 (cit. on pp. 48, 50).
- [160] D. Go. *ORBITRANS*. [https://iffgit.fz-juelich.de/dgo/wann\\_interpol](https://iffgit.fz-juelich.de/dgo/wann_interpol). To be published (cit. on p. 49).
- [161] Giovanni Pizzi, Valerio Vitale, Ryotaro Arita, et al. “Wannier90 As a Community Code: New Features and Applications”. In: *Journal of Physics: Condensed Matter* 32.16 (2020), p. 165902 (cit. on pp. 49, 52, 71, 85).

- [162] Luká Nádvořník, Oliver Gueckstock, Lukas Braun, et al. “Terahertz Spin-to-Charge Current Conversion in Stacks of Ferromagnets and the Transition-Metal Dichalcogenide NbSe<sub>2</sub>”. In: *Advanced Materials Interfaces* 9.36 (2022), p. 2201675 (cit. on pp. 51, 56, 65).
- [163] Andrea Splendiani, Liang Sun, Yuanbo Zhang, et al. “Emerging Hotoluminescence in Monolayer MoS<sub>2</sub>”. In: *Nano Letters* 10.4 (2010), pp. 1271–1275 (cit. on p. 51).
- [164] Pere Miró, Mahdi Ghorbani-Asl, and Thomas Heine. “Two Dimensional Materials Beyond MoS<sub>2</sub>: Noble-Transition-Metal Dichalcogenides”. In: *Angewandte Chemie International Edition* 53.11 (2014), pp. 3015–3018 (cit. on p. 51).
- [165] Sihan Zhao, Takato Hotta, Takashi Koretsune, et al. “Two-Dimensional Metallic NbS<sub>2</sub>: Growth, Optical Identification and Transport Properties”. In: *2D Materials* 3.2 (2016), p. 025027 (cit. on p. 51).
- [166] I Guillamón, Hermann Suderow, José G Rodrigo, et al. “Chiral Charge Order in the Superconductor 2H-TaS<sub>2</sub>”. In: *New Journal of Physics* 13.10 (2011), p. 103020 (cit. on p. 51).
- [167] Zhiyong Y Zhu, Yingchun C Cheng, and Udo Schwingenschlögl. “Giant Spin-Orbit-Induced Spin Splitting in Two-Dimensional Transition-Metal Dichalcogenide semiconductors”. In: *Physical Review B* 84.15 (2011), p. 153402 (cit. on p. 51).
- [168] Huei-Ru Fuh, Binghai Yan, Shu-Chun Wu, Claudia Felser, and Ching-Ray Chang. “Metal-Insulator Transition and the Anomalous Hall Effect in the Layered Magnetic Materials VS<sub>2</sub> and VSe<sub>2</sub>”. In: *New Journal of Physics* 18.11 (2016), p. 113038 (cit. on p. 51).
- [169] Kin Fai Mak, Kathryn L McGill, Jiwoong Park, and Paul L McEuen. “The Valley Hall Effect in MoS<sub>2</sub> Transistors”. In: *Science* 344.6191 (2014), pp. 1489–1492 (cit. on p. 51).
- [170] Xiaodong Zhou, Run-Wu Zhang, Zeying Zhang, et al. “Sign-Reversible Valley-Dependent Berry Phase Effects in 2D Valley-Half-Semiconductors”. In: *npj Computational Materials* 7.1 (2021), p. 160 (cit. on p. 51).
- [171] Wanxiang Feng, Yugui Yao, Wenguang Zhu, et al. “Intrinsic Spin Hall Effect in Monolayers of Group-VI dichalcogenides: A first-Principles Study”. In: *Physical Review B* 86.16 (2012), p. 165108 (cit. on pp. 51, 54, 69, 77).
- [172] Di Xiao, Gui-Bin Liu, Wanxiang Feng, Xiaodong Xu, and Wang Yao. “Coupled Spin and Valley Physics in Monolayers of MoS<sub>2</sub> and Other Group-VI Dichalcogenides”. In: *Physical Review Letters* 108.19 (2012), p. 196802 (cit. on pp. 51, 53, 69, 77).
- [173] Luis M. Canonico, Tarik P. Cysne, Tatiana G. Rappoport, and R. B. Muniz. “Two-Dimensional Orbital Hall Insulators”. In: *Physical Review B* 101 (7 Feb. 2020), p. 075429 (cit. on pp. 51, 63, 69, 77).
- [174] Aiming Yan, Chin Shen Ong, Diana Y Qiu, et al. “Dynamics of Symmetry-Breaking Stacking Boundaries in Bilayer MoS<sub>2</sub>”. In: *The Journal of Physical Chemistry C* 121.40 (2017), pp. 22559–22566 (cit. on p. 53).



- [175] Ángel Morales García, Elena Del Corro, Martin Kalbac, and Otakar Frank. “Tuning the Electronic Properties of Monolayer and Bilayer Transition Metal Dichalcogenide Compounds Under direct out-of-plane Compression”. In: *Physical Chemistry Chemical Physics* 19.20 (2017), pp. 13333–13340 (cit. on p. 53).
- [176] Humberto Terrones and Mauricio Terrones. “Bilayers of Transition Metal Dichalcogenides: Different Stackings and Heterostructures”. In: *Journal of Materials Research* 29.3 (2014), pp. 373–382 (cit. on p. 59).
- [177] Zui Tao, Bowen Shen, Wenjin Zhao, et al. “Giant Spin Hall Effect in AB-Stacked MoTe<sub>2</sub>/WSe<sub>2</sub> Bilayers”. In: *Nature Nanotechnology* 19.1 (2024), pp. 28–33 (cit. on p. 61).
- [178] Ning Lu, Hongyan Guo, Lei Li, et al. “MoS<sub>2</sub>/MX<sub>2</sub> Heterobilayers: Bandgap Engineering via Tensile Strain or External Electrical Field”. In: *Nanoscale* 6.5 (2014), pp. 2879–2886 (cit. on p. 61).
- [179] M. Chhowalla et al. “The Chemistry of Two-Dimensional Layered Transition Metal Dichalcogenide Nanosheets”. In: *Nature Chemistry* 5 (2013), pp. 263–275 (cit. on p. 63).
- [180] K. K. Liu et al. “Role of the Carrier Gas Flow Rate in Monolayer MoS<sub>2</sub> Growth by Chemical Vapor Deposition”. In: *Physical Review B* 88 (2013), p. 035436 (cit. on p. 64).
- [181] Sohail Ahmad, Sugata Mukherjee, et al. “A Comparative Study of Electronic Properties of Bulk MoS<sub>2</sub> and its Monolayer Using DFT Technique: Application of Mechanical Strain on MoS<sub>2</sub> Monolayer”. In: *Graphene* 3.04 (2014), p. 52 (cit. on p. 64).
- [182] Mahmoud Zeer, Dongwook Go, Johanna P Carbone, et al. “Spin and Orbital Transport in Rare-Earth Dichalcogenides: The Case of EuS<sub>2</sub>”. In: *Physical Review Materials* 6.7 (2022), p. 074004 (cit. on pp. 69, 84, 92).
- [183] Z. Y. Zhu, Y. C. Cheng, and U. Schwingenschlögl. “Giant Spin-Orbit-Induced Spin Splitting in Two-Dimensional Transition-Metal Dichalcogenide Semiconductors”. In: *Physical Review B* 84 (15 Oct. 2011), p. 153402 (cit. on p. 69).
- [184] Huei-Ruh Fuh, Binghai Yan, Shu-chun Wu, Claudia Felser, and ChinRay Chang. “Metal-Insulator Transition and the Anomalous Hall Effect in the Layered Magnetic Materials VS<sub>2</sub> and VSe<sub>2</sub>”. In: *New Journal of Physics* 18 (2016), p. 113038 (cit. on p. 69).
- [185] K. F. Mak, K. L. McGill, J. Park, and P. L. McEuen. “The valley Hall Effect in MoS<sub>2</sub> Transistors”. In: *Science* 344.6191 (June 2014), pp. 1489–1492 (cit. on p. 69).
- [186] Xiaodong Zhou, Run-wu Zhang, Zeying Zhang, et al. “Sign-Reversible Valley-Dependent Berry Phase Effects in 2D Valley-Half-Semiconductors”. In: *npj Computational Materials* 7 (2021), p. 160 (cit. on p. 69).
- [187] Sayantika Bhowal and S. Satpathy. “Intrinsic Orbital and Spin Hall Effects in Monolayer Transition Metal Dichalcogenides”. In: *Physical Review B* 102 (3 July 2020), p. 035409 (cit. on p. 69).

- [188] Yanping Liu, Yuanji Gao, Siyu Zhang, et al. “Valleytronics in Transition Metal Dichalcogenides Materials”. In: *Nano Research* 12 (2019), pp. 2695–2711 (cit. on p. 69).
- [189] H Kontani, T Tanaka, Dai S Hirashima, K Yamada, and J Inoue. “Giant Orbital Hall Effect in Transition Metals: Origin of Large Spin and Anomalous Hall Effects”. In: *Physical Review Letters* 102 (2009), p. 016601 (cit. on pp. 69, 98).
- [190] Daniel Wortmann, Gregor Michalicek, Nadjib Baadji, et al. *FLEUR*. Zenodo. May 2023 (cit. on pp. 71, 104).
- [191] A. B. Shick, A. I. Liechtenstein, and W. E. Pickett. “Implementation of the LDA+U Method Using the Full-Potential Linearized Augmented Plane-Wave Basis”. In: *Physical Review B* 60 (15 Oct. 1999), pp. 10763–10769 (cit. on pp. 71, 85, 105, 119).
- [192] F. Freimuth, Y. Mokrousov, D. Wortmann, S. Heinze, and S. Blügel. “Maximally Localized Wannier Functions Within the FLAPW Formalism”. In: *Physical Review B* 78 (3 July 2008), p. 035120 (cit. on pp. 71, 106).
- [193] Hongyan Guo, Ning Lu, Lu Wang, Xiaojun Wu, and Xiao Cheng Zeng. “Tuning Electronic and Magnetic Properties of Early Transition-Metal Dichalcogenides via Tensile Strain”. In: *The Journal of Physical Chemistry C* 118.13 (2014), pp. 7242–7249 (cit. on p. 74).
- [194] Yalong Jiao, Liujiang Zhou, Fengxian Ma, et al. “Predicting Single-Layer Technetium Dichalcogenides  $\text{TcX}_2$  ( $\text{X}=\text{S}, \text{Se}$ ) with Promising Applications in Photovoltaics and Photocatalysis”. In: *ACS Applied Materials & Interfaces* 8.8 (2016). PMID: 26859697, pp. 5385–5392 (cit. on p. 74).
- [195] Naoto Nagaosa, Jairo Sinova, Shigeki Onoda, A. H. MacDonald, and N. P. Ong. “Anomalous Hall Effect”. In: *Reviews of Modern Physics* 82 (2 May 2010), pp. 1539–1592 (cit. on p. 74).
- [196] Hongbin Zhang, Frank Freimuth, Gustav Bihlmayer, et al. “Engineering Quantum Anomalous Hall Phases with Orbital and Spin Degrees of Freedom”. In: *Physical Review B* 87 (20 May 2013), p. 205132 (cit. on pp. 75, 76).
- [197] H. Kontani, T. Tanaka, D. S. Hirashima, K. Yamada, and J. Inoue. “Giant Intrinsic Spin and Orbital Hall Effects in  $\text{Sr}_2\text{MO}_4$  ( $M = \text{Ru}, \text{Rh}, \text{Mo}$ )”. In: *Physical Review Letters* 100 (9 Mar. 2008), p. 096601 (cit. on p. 77).
- [198] Dongwook Go, Daegeun Jo, Hyun-Woo Lee, Mathias Kläui, and Yuriy Mokrousov. “Orbitronics: Orbital Currents in Solids”. In: *EPL (Europhysics Letters)* 135.3 (Aug. 2021), p. 37001 (cit. on p. 77).
- [199] Dongwook Go, Daegeun Jo, Tenghua Gao, et al. “Orbital Rashba Effect in a Surface-Oxidized Cu film”. In: *Physical Review B* 103.12 (2021), p. L121113 (cit. on p. 78).
- [200] Mahmoud Zeer, Dongwook Go, Peter Schmitz, et al. “Promoting p-Based Hall Effects by p-d-f Hybridization in Gd-Based Dichalcogenides”. In: *Physical Review Research* 6.1 (2024), p. 013095 (cit. on p. 83).



- [201] Ph. Kurz, Gustav Bihlmayer, and Stefan Blügel. “Magnetism and Electronic Structure of hcp Gd and the Gd(0001) Surface”. In: *Journal of Physics: Condensed Matter* 14 (2002), pp. 6353–6371 (cit. on p. 85).
- [202] Johanna P. Carbone, Dongwook Go, Yuriy Mokrousov, Gustav Bihlmayer, and Stefan Blügel. “Engineering Spin-Orbit Effects and Berry Curvature by Deposition of a Monolayer of Eu on WSe<sub>2</sub>”. In: *Physical Review B* 106 (6 Aug. 2022), p. 064401 (cit. on p. 85).
- [203] Xingxing Li, Xiaojun Wu, Zhenyu Li, Jinlong Yang, and JG Hou. “Bipolar Magnetic Semiconductors: A New Class of Spintronics Materials”. In: *Nanoscale* 4.18 (2012), pp. 5680–5685 (cit. on p. 88).
- [204] Haixia Cheng, Jun Zhou, Ming Yang, et al. “Robust Two-Dimensional Bipolar Magnetic Semiconductors by Defect Engineering”. In: *Journal of Materials Chemistry C* 6.31 (2018), pp. 8435–8443 (cit. on p. 88).
- [205] MP Lopez Sancho, JM Lopez Sancho, JM Lopez Sancho, and J Rubio. “Highly Convergent Schemes for the Calculation of Bulk and Surface Green Functions”. In: *Journal of Physics F: Metal Physics* 15.4 (1985), p. 851 (cit. on p. 98).
- [206] QuanSheng Wu, ShengNan Zhang, Hai-Feng Song, Matthias Troyer, and Alexey A Soluyanov. “WannierTools: An Open-Source Software Package for Novel Topological Materials”. In: *Computer Physics Communications* 224 (2018), pp. 405–416 (cit. on p. 98).
- [207] E Myczak, I Aguilera, P Gospodari, et al. “Fe (001) Angle-Resolved Photoemission and Intrinsic Anomalous Hall Conductivity in Fe Seen by Different ab initio Approaches: LDA and GGA Versus GW”. In: *Physical Review B* 105.11 (2022), p. 115135 (cit. on p. 101).
- [208] Ying-Jiun Chen, Jan-Philipp Hanke, Markus Hoffmann, et al. “Spanning Fermi Arcs in a Two-Dimensional Magnet”. In: *Nature Communications* 13.1 (2022), p. 5309 (cit. on p. 101).
- [209] Chengwang Niu, Jan-Philipp Hanke, Patrick M Buhl, et al. “Mixed Topological Semimetals Driven by Orbital Complexity in Two-Dimensional Ferromagnets”. In: *Nature Communications* 10.1 (2019), p. 3179 (cit. on p. 101).
- [210] Jan-Philipp Hanke, Frank Freimuth, Chengwang Niu, Stefan Blügel, and Yuriy Mokrousov. “Mixed Weyl Semimetals and Low-Dissipation Magnetization Control in Insulators by Spin–Orbit Torques”. In: *Nature Communications* 8.1 (2017), p. 1479 (cit. on pp. 101, 114).
- [211] Wenrong Liu, Xinyang Li, Changwen Zhang, and Shishen Yan. “Janus VXY Monolayers with Tunable Large Berry Curvature”. In: *Journal of Semiconductors* 43.4 (2022), p. 042501 (cit. on p. 101).
- [212] Ang-Yu Lu, Hanyu Zhu, Jun Xiao, et al. “Janus Monolayers of Transition Metal Dichalcogenides”. In: *Nature Nanotechnology* 12.8 (2017), pp. 744–749 (cit. on p. 101).

- [213] Miguel Ángel Hernández-Vázquez, Andrés de Luna Bugallo, and Daniel Olgún. “Janus Monolayers of Transition Metal Dichalcogenides: A DFT Study”. In: *Physica Status Solidi (b)* 259.1 (2022), p. 2100248 (cit. on p. 101).
- [214] Hongxin Chen, Ran Liu, Jiajun Lu, et al. “Intrinsic Valley-Polarized Quantum Anomalous Hall Effect and Controllable Topological Phase Transition in Janus Fe<sub>2</sub>SSe”. In: *The Journal of Physical Chemistry Letters* 13.44 (2022), pp. 10297–10304 (cit. on p. 101).
- [215] Mahmoud Zeer, Dongwook Go, Mathias Kläui, et al. “Orbital Torques and Orbital Pumping in Two-Dimensional Rare-Earth Dichalcogenides”. In: *npj Computational Materials* 11.1 (2025), p. 305 (cit. on p. 103).
- [216] Jan Hidding and Marcos HD Guimarães. “Spin-orbit Torques in Transition Metal Dichalcogenide/Ferromagnet Heterostructures”. In: *Frontiers in Materials* 7 (2020), p. 594771 (cit. on p. 103).
- [217] Yan-Dong Guo, Hong-Bo Zhang, Hong-Li Zeng, et al. “A Progressive Metal–Semiconductor Transition in Two-Faced Janus Monolayer Transition-Metal Chalcogenides”. In: *Physical Chemistry Chemical Physics* 20.32 (2018), pp. 21113–21118 (cit. on p. 104).
- [218] Lei Zhang, Zhenjingfeng Yang, Tian Gong, et al. “Recent Advances in Emerging Janus Two-Dimensional Materials: From Fundamental Physics to Device Applications”. In: *Journal of materials chemistry A* 8.18 (2020), pp. 8813–8830 (cit. on p. 104).
- [219] Jing Zhang, Shuai Jia, Iskandar Kholmanov, et al. “Janus Monolayer Transition-Metal Dichalcogenides”. In: *ACS Nano* 11.8 (2017), pp. 8192–8198 (cit. on p. 104).
- [220] Ang-Yu Lu, Hanyu Zhu, Jun Xiao, et al. “Janus Monolayers of Transition Metal Dichalcogenides”. In: *Nature nanotechnology* 12.8 (2017), pp. 744–749 (cit. on p. 104).
- [221] Lei Zhang, Zhenjingfeng Yang, Tian Gong, et al. “Recent Advances in Emerging Janus Two-Dimensional Materials: From Fundamental Physics to Device Applications”. In: *Journal of Materials Chemistry A* 8.18 (2020), pp. 8813–8830 (cit. on p. 104).
- [222] Sheng-Bin Yu, Ma Zhou, Dong Zhang, and Kai Chang. “Spin Hall Effect in the Monolayer Janus Compound MoSSe Enhanced by Rashba Spin-Orbit Coupling”. In: *Physical Review B* 104 (7 Aug. 2021), p. 075435 (cit. on p. 104).
- [223] Nesta Benno Joseph, Saswata Roy, and Awadhesh Narayan. “Tunable Topology and Berry Curvature Dipole in Transition Metal Dichalcogenide Janus Monolayers”. In: *Materials Research Express* 8.12 (2021), p. 124001 (cit. on p. 104).
- [224] San-Dong Guo, Wen-Qi Mu, Xiang-Bo Xiao, and Bang-Gui Liu. “Intrinsic Room-Temperature Piezoelectric Quantum Anomalous Hall Insulator in Janus Monolayer Fe 2 IX (X= Cl and Br)”. In: *Nanoscale* 13.30 (2021), pp. 12956–12965 (cit. on p. 104).



- [225] Vuong Van Thanh, Nguyen Duy Van, Riichiro Saito, Nguyen Tuan Hung, et al. “First-Principles Study of Mechanical, Electronic and Optical Properties of Janus Structure in Transition Metal Dichalcogenides”. In: *Applied Surface Science* 526 (2020), p. 146730 (cit. on p. 104).
- [226] Wenwu Shi and Zhiguo Wang. “Mechanical and Electronic Properties of Janus Monolayer Transition Metal Dichalcogenides”. In: *Journal of Physics: Condensed Matter* 30.21 (2018), p. 215301 (cit. on p. 104).
- [227] Yufeng Luo, Shihao Han, Rui Hu, et al. “The Thermal Stability of Janus Monolayers SnXY (X, Y= O, S, Se): ab-initio Molecular Dynamics and Beyond”. In: *Nanomaterials* 12.1 (2021), p. 101 (cit. on p. 104).
- [228] Xiao Tang and Liangzhi Kou. “2D Janus Transition Metal Dichalcogenides: Properties and Applications”. In: *physica status solidi (b)* 259.4 (2022), p. 2100562 (cit. on p. 104).
- [229] Lei Zhang, Yong Xia, Xudong Li, et al. “Janus Two-Dimensional Transition Metal Dichalcogenides”. In: *Journal of Applied Physics* 131.23 (2022), p. 230902 (cit. on p. 104).
- [230] Libor Vojáček, Joaquín Medina Dueñas, Jing Li, et al. “Field-Free Spin–Orbit Torque Switching in Janus Chromium Dichalcogenides”. In: *Nano Letters* (2024) (cit. on p. 104).
- [231] Yuting Liu and Qiming Shao. “Two-Dimensional Materials for Energy-Efficient Spin–Orbit Torque Devices”. In: *ACS Nano* 14.8 (2020), pp. 9389–9407 (cit. on p. 104).
- [232] Wei Tang, Haoliang Liu, Zhe Li, Anlian Pan, and Yu-Jia Zeng. “Spin-Orbit Torque in Van der Waals-Layered Materials and Heterostructures”. In: *Advanced Science* 8.18 (2021), p. 2100847 (cit. on p. 104).
- [233] Ph Kurz. “Magnetism and Electronic Structure of hcp Gd and the Gd (0001) Surface”. In: *Journal of Physics: Condensed Matter* 14.25 (2002), p. 6353 (cit. on pp. 105, 119).
- [234] Arash A. Mostofi, Jonathan R. Yates, Giovanni Pizzi, et al. “An Updated Version of Wannier90: A Tool for Obtaining Maximally-Localised Wannier Functions”. In: *Computer Physics Communications* 185.8 (2014), pp. 2309–2310 (cit. on p. 106).
- [235] Frank Freimuth, Stefan Blügel, and Yuriy Mokrousov. “Spin-Orbit Torques in Co/Pt (111) and Mn/W (001) Magnetic Bilayers from First Principles”. In: *Physical Review B* 90.17 (2014), p. 174423 (cit. on pp. 114, 117).
- [236] Qidong Xie, Weinan Lin, Baishun Yang, et al. “Giant Enhancements of Perpendicular Magnetic Anisotropy and Spin-Orbit Torque by a MoS<sub>2</sub> Layer”. In: *Advanced Materials* 31.21 (2019), p. 1900776 (cit. on p. 114).
- [237] D MacNeill, GM Stiehl, MHD Guimaraes, et al. “Control of Spin–Orbit Torques Through Crystal Symmetry in WTe<sub>2</sub>/Ferromagnet Bilayers”. In: *Nature Physics* 13.3 (2017), pp. 300–305 (cit. on p. 114).

- [238] Sajid Husain, Rahul Gupta, Ankit Kumar, et al. “Emergence of Spin–Orbit Torques in 2D Transition Metal Dichalcogenides: A Status Update”. In: *Applied Physics Reviews* 7.4 (2020) (cit. on p. 114).
- [239] Matthias Althammer, Mathias Weiler, Hans Huebl, and Sebastian TB Goennenwein. “Spin Pumping”. In: *Spintronics for Next Generation Innovative Devices* (2015), pp. 111–124 (cit. on p. 118).
- [240] Hiroki Hayashi, Dongwook Go, Satoshi Haku, Yuriy Mokrousov, and Kazuya Ando. “Observation of Orbital Pumping”. In: *Nature Electronics* 7.8 (2024), pp. 646–652 (cit. on p. 118).
- [241] Federico Mazzola, Wojciech Brzezicki, Maria Teresa Mercaldo, et al. “Signatures of a Surface Spin–Orbital Chiral Metal”. In: *Nature* 626.8000 (2024), pp. 752–758 (cit. on p. 127).
- [242] Zhihao Gao, Yuehao Yin, Yuwan Wang, et al. “Tunable Electrical Properties and Multiple-Phases of Ferromagnetic GdS<sub>2</sub>, GdSe<sub>2</sub> and Janus GdSSe monolayers”. In: *Physical Chemistry Chemical Physics* 25.34 (2023), pp. 22782–22793 (cit. on p. 131).
- [243] Andrey M Tokmachev, Dmitry V Averyanov, Alexander N Taldenkov, et al. “Lanthanide f 7 Metalloxyenes—A Class of Intrinsic 2D Ferromagnets”. In: *Materials Horizons* 6.7 (2019), pp. 1488–1496 (cit. on p. 131).











Band / Volume 117

**Electrical anisotropy and shear-resistant topology in the quasi one-dimensional van-der-Waals material  $\alpha\text{-Bi}_4\text{Br}_4$**

J.K. Hofmann (2025) xv, 129 pp

ISBN: 978-3-95806-869-8

Band / Volume 118

**Spin-orbital mixing in the topological ladder of the two-dimensional metal  $\text{PtTe}_2$**

M. Qahosh (2025), ix, 170 pp

ISBN: 978-3-95806-872-8

Band / Volume 119

**Functions of SNNs constrained by biology**

A. Korcsak-Gorzo (2025), xvi, 145 pp

ISBN: 978-3-95806-876-6

Band / Volume 120

**On Scalable Integrated Charge State Tuning for Semiconductor Quantum Dot Devices**

F. A. Hader (2026), xx, 145 pp

ISBN: 978-3-95806-884-1

Band / Volume 121

**Efficient Massively Space-Time-Parallel Simulations with Adaptive Spectral Deferred Correction**

T. Saupe (2026), v, 149 pp

ISBN: 978-3-95806-886-5

Band / Volume 122

**Controlled single-molecule manipulation**

P. Kopp (2026), viii, 136 pp

ISBN: 978-3-95806-887-2

Band / Volume 123

**Growth and Excitation Dynamics of Epitaxial 2D Materials and Molecular Layers Studied by Electron and Momentum Microscopy**

M. E. Rath (2026), xvi, 161, XXIX pp

ISBN: 978-3-95806-892-6

Band / Volume 124

**Ferroelectric and Resistive Switching in Epitaxial  $\text{Hf}_{0.5}\text{Zr}_{0.5}\text{O}_2$**

J. R. Knabe (2026), 145 pp

ISBN: 978-3-95806-895-7

Band / Volume 125

**Temperature Influences on Frequency Mixing  
Magnetic Detection for mobile Drinking Water Analysis in Crisis Areas**

M. Jessing (2026), xx, 113 pp

ISBN: 978-3-95806-902-2

Band / Volume 126

**Light-matter interaction from ab-initio Keldysh formalism**

M. D. Merte (2026), xvi, 184, A-27 pp

ISBN: 978-3-95806-906-0

Band / Volume 127

**Towards high-quality decoupled Graphene heterostructures**

M. Hutter (2026), VIII, 149 pp

ISBN: 978-3-95806-912-1 (Print)

ISBN: 978-3-95806-913-8 (E-Book)

Band / Volume 128

**Adaptive-precision interatomic potentials  
for large-scale atomistic simulations**

D. Immel (2026), xxiv, 135 pp

ISBN: 978-3-95806-934-3 (Print)

ISBN: 978-3-95806-935-0 (E-Book)

Band / Volume 129

**Competing spin-orbit coupling and exchange  
interaction in a two-dimensional ferromagnet**

X. L. Tan (2026), ii, 113 pp

ISBN: 978-3-95806-938-1 (Print)

ISBN: 978-3-95806-939-8 (E-Book)

Band / Volume 130

**Analysis of Electrophysiological Data with Invertible Neural Networks**

P. Bouss (2026), xxii, 187 pp

ISBN: 978-3-95806-944-2 (Print)

ISBN: 978-3-95806-945-9 (E-Book)

Band / Volume 131

**Orbitronics with Two-Dimensional Rare-Earth Dichalcogenides  
from First Principles**

M. Zeer (2026), xi, 159 pp

ISBN: 978-3-95806-960-2 (Print)

ISBN: 978-3-95806-961-9 (E-Book)



Information  
Band / Volume 131  
ISBN 978-3-95806-961-9