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Publication Date

2026-05-01

Peer reviewed|Thesis/dissertation

Studies of Magnetic Switching in Intercalated Transition Metal Dichalcogenides

by

Josue Rodriguez

A dissertation submitted in partial satisfaction of the

requirements for the degree of

Doctor of Philosophy

in

Physics

in the

Graduate Division

of the

University of California, Berkeley

Committee in charge:

Professor James G. Analytis, Chair

Professor Joel Moore

Associate Professor Kwabena Bediako

Spring 2026

Studies of Magnetic Switching in Intercalated Transition Metal Dichalcogenides

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by

Josue Rodriguez

Abstract

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Doctor of Philosophy in Physics

University of California, Berkeley

Professor James G. Analytis, Chair

The search for next-generation spintronic devices with ultrafast operation, low power consumption, and fully electrical functionality is a key frontier in condensed-matter physics and materials science, driven by the demand for energy-efficient and high-speed information technologies. Devices of antiferromagnetic (AFM) quantum materials are a promising platform owing to the fact that AFM do not produce stray magnetic fields making them viable to be more tightly packed, they possess ultrafast dynamics, and are robust to external magnetic perturbations.

Intercalated transition-metal dichalcogenides (I-TMDs) provide a tunable platform for exploring electrically driven switching in low-dimensional materials. By inserting magnetic transition-metal ions into the van der Waals gaps of layered dichalcogenides, intercalation stabilizes long-range magnetic order and complex spin textures that are highly sensitive to composition and symmetry. Previous studies in Fe_xNbS_2 , showed that electrical pulses could switch between two distinct resistance states. It was first proposed that electrical pulses would rotate an in-plane component of the Neel vector via spin-orbit transfer torque. Further studies determined that the electrical pulses rotate the principal nematic axis of the magnetic order via magnetoelastic coupling and not the reversal of the Neel vector via a spin-orbit transfer torque [1]. This dissertation focuses on the current-induced switching mechanisms in $\text{Ni}_{1/4}\text{TaSe}_2$ and $\text{Co}_{1/3}\text{TaS}_2$. In $\text{Ni}_{1/4}\text{TaSe}_2$, devices exhibit current-induced magnetization reversal dominated by Joule-heating-assisted dynamics making it an all-electronic read-write scheme for magnetization recording. Electrical pulses in $\text{Co}_{1/3}\text{TaS}_2$, was seen to comb its helical magnetic texture enabling the ability to encode information. Together, these results demonstrate that current-induced switching in intercalated magnets arises from multiple, material-dependent mechanisms controlled by magnetic texture, lattice coupling, and dissipation pathways. This work clarifies when and why electrical switching is possible in frustrated AFM systems and establishes guiding principles for designing scalable, low-power spintronic devices.

To Jordan Elias Rodriguez and Allisen Rodriguez

Dedicated to my family, who taught me dedication, perseverance and the sanctification
that comes from hard work.

Contents

Contents	ii
List of Figures	iv
List of Tables	vi
1 Introduction and Background	1
1.1 Intercalated Transition Metal Dichalcogenides	1
1.2 Magnetism	7
1.3 Spintronics	9
1.4 Switching in Antiferromagnetics Materials	14
1.5 Anomalous Hall Effect in Antiferromagnets	17
1.6 Electronic Transport in Correlated and Disordered Materials	18
2 Synthesis	19
2.1 Introduction	19
2.2 Nickel Intercalated Tantalum Diselenide	22
2.3 Cobalt Intercalated Tantalum Disulfide	22
2.4 Appendix I: Two Zone Furnace Operation “King Mellen”	32
3 Device Fabrication: Focused Ion Beam	38
3.1 Sample Preparation	38
3.2 Microstructuring of crystals	40
4 Nickel Intercalated Tantalum Diselenide	44
4.1 Introduction	44
4.2 Methods	46
4.3 Joule-Heating Assisted Magnetization Reversal	46
5 Cobalt Intercalated Tantalum Disulfide	53
5.1 Introduction	53
5.2 Bulk Transport Measurements	56
5.3 Methods	65

6	Switching in Cobalt Intercalated Tantalum Disulfide	69
6.1	Introduction	69
6.2	Methods	70
6.3	Electric Transport	72
6.4	Switching response in $\text{Co}_{1/3}\text{TaS}_2$	72
7	Conclusion and Outlook	81
	Bibliography	83

List of Figures

1.1	2H and 1T polytypes of TMDs	2
1.2	Unit cell of the intercalated TMDs $T_{1/3}MX_2$ and $T_{1/4}MX_2$	4
1.3	d orbital splitting and spin configuration of the intercalated transition metal.	6
1.4	Magnetization trajectory under LLG equation	11
1.5	Magnetic recording trilemma and HAMR concept	12
1.6	Helimagnetic Ordering and Conical Modulation	16
2.1	Typical solid state reaction temperature profile	20
2.2	Chemical vapor transport scheme	21
2.3	Electron dispersive x-ray map and histogram	28
2.4	MTI two-zone furnace	30
2.5	Two-zone furnace temperature ramp schemes	31
2.6	“King” MELLEN front control panel	32
3.1	Exfoliated crystals	39
3.2	Single fiber wooden stick	40
3.3	SEM micrograph of post-FIB devices	41
3.4	Typical process flow for FIB microstructuring of devices.	42
3.5	Lamella microstructuring process flow.	43
4.1	Current-induced magnetization reversal	45
4.2	Direct imaging of current-induced magnetization reversal using MOKE	47
4.3	Direct measurement of Joule heating	48
4.4	Measured Hall effect under a perpendicular magnetic field with an applied positive and negative DC current. Anomalous Hall resistivity measured (at 2 K) with various applied DC currents.	49
4.5	Pulse current polarity dependence on $Ni_{1/4}TaSe_2$ devices	50
4.6	Pulse width dependence on $Ni_{1/4}TaSe_2$ devices	52
5.1	Magnetic Order of Co_xTaS_2	54
5.2	Scaling analysis of the low-temperature resistivity upturn in Co-intercalated TaS_2	55
5.3	Intercalation dependence of the magnetization, resistivity, and hall resistivity versus temperature in Co_xTaS_2	57

5.4	Intercalation dependence of the ρ , and Hall effect in Co_xTaS_2	59
5.5	Intercalation dependence of the magnetoresistance in Co_xTaS_2	61
5.6	Intercalation dependence of magnetization versus Field in Co_xTaS_2	63
5.7	Thermally-modulated magneto-optical effects	64
5.8	Energy-dispersive X-ray (EDX) spectra for samples with various intercalation levels x.	68
6.1	Switching response in a $\text{Co}_{1/3}\text{TaS}_2$ FIB device	71
6.2	Switching response in a $\text{Co}_{1/3}\text{TaS}_2$ device	73
6.3	Pulse parameters and temperature dependence of $\text{Co}_{1/3}\text{TaS}_2$	74
6.4	Hysteresis dependence on pulse currents in a $\text{Co}_{1/3}\text{TaS}_2$ device	75
6.5	Switching response in $\text{Co}_{1/3}\text{TaS}_2$ to magnetic field training and field dependence	77
6.6	Current-field activation map of the transverse resistance response.	78
6.7	Pulse direction dependence on switching response.	80

List of Tables

2.1	Equilibrium constants for species in $\text{Co}_{1/3}\text{TaS}_2$ CVT reaction.	23
2.2	Calculated partial pressures with an initial amount of 100 mg of I_2 at 1273 K . .	24
2.3	Typical program for a CVT reaction with a target temperature gradient of 950 °C-850 °C to be used in a MTI-Two Zone furnace. Note the last time value of -121 at set point 10 instructs the furnace to switch off power to the heating elements, thereby initiating a natural cool down.	29
2.4	Schematic outline of typical program for a CVT reaction with a target temperature gradient of 950 °C-850 °C to be used in the “King” Mellen Two Zone furnace. A detailed standard operating procedure of the “King” Mellen is given in appendix 1.	29
5.1	Transition temperatures, T^* , obtained from resistivity, hall effect and magnetization for different intercalations.	58
5.2	EDX-derived Co intercalation ratios and uncertainties.	67

Acknowledgments

Above all, I thank our Lord Jesus Christ, the Logos and source of all truth, wisdom and knowledge. It is by His grace that I was able to complete this dissertation, to whom be glory to ages of ages. Amen.

I would like to thank my advisor, **James**, for your patience, guidance, and for never giving up on me, even at times when I had given up on myself. Thank you for your joyful approach to science and your unyielding belief in others. I am also grateful for your leadership as department chair through those tumultuous years during and after COVID and the ridiculous student union strike. Lord knows many of us students were misled by the union leadership and ideologically possessed by the zeitgeist. May God bless you and your family. Thank you, **Hossein**, for your mentorship as my de facto second advisor. I learned a great deal from your meticulous attention to detail and it was remarkable to watch your ability to generate hypotheses from experimental observations. Your dedication to working long hours was a source of motivation for me to also put in the hours in the lab. Thank you also for your grounded practical approach to science. It was truly a pleasure working with you.

To the previous Analytis lab members whom I overlapped with, **Nikola** aka *King Dodo*, **Vikram**, **Shannon**, **Ella**, **Eran** and **Sylvia**, thank you all for creating a friendly and intellectually stimulating environment in which to learn and grow. To my contemporary graduate student colleagues whom I have had the pleasure of working alongside, **Koh** thank you for being a homie. It has been a pleasure to see you grow from a callow proponent of topological materials into a more refined and judicious experimental physicist. **Yacine**, we had way too many great laughs to recount. I expect you to be the Prime Minister of Morocco someday. **Valeria**, you were one of the strongest women I have had the pleasure of working with and I am truly grateful for the time we shared. I wish you the best wherever life takes you. **Mr. Darian**, you have been a breath of fresh air in the lab, youthful, humorous, and grounded (a rare and valuable combination). It was a pleasure trying to make you laugh and that one time we slightly rearranged everything on Yuanqi's desk, for which you took all the blame. **Yuanqi**, I will greatly miss you, my dear brother. We shared many great conversations covering a wide range of topics. Thank you for diligently answering every question I asked, technical or pedestrian. And thank you for reviving the office coffee machine and keeping the flow of coffee and milk flowing for the rest of us. Thank you, **Luke**, for always listening to my jokes, no matter how terrible some were, and for always bringing such a joyful disposition. I hope to visit castles and drink beer with you in the English countryside. I pray that you still have a country to come back to when you return to England. **Ryan**, thank you for being a wonderful example of how a responsible adult should conduct himself. Thank you as well for your friendship and for the many pizzas and beers shared at Fieldwork. And to the next generation of Analytis graduate students **Ichiro**, **Henry**, and **Ry** I wish you all tremendous success. And to the many wonderful post-docs

Lu, Augusto, Grace and **Pranav**, thank you. **Lior**, I truly enjoyed our conversations about powerlifting, bodybuilding, and, of course, the injuries that came with them. I wish you the very best.

Every great lab has in its arsenal many great undergraduate student assistants, and although I attempted many times to dissuade ours from going to graduate school, it was no doubt the glamorous lifestyle that we graduate students live that caused my advice to fall on deaf ears. **Sanjit** and **Rainer**, thank you for your enthusiasm for physics and free food. It has been a pleasure watching you boys grow over the years. **Sanaz**, I wish you tremendous success in all your future endeavors. We shared many meaningful conversations, and do not be afraid to search for the truth, no matter where it may lead you. **Catherine**, thank you for all your hard work and dedication to our research endeavors. For the many beautifully wired devices, the many exquisite crystals you synthesized, the many long hours at LBL, and that one time we missed the last LBL shuttle and got caught in the rain walking down the hill. And thank you for bestowing on me a deeper appreciation for cats.

Science is an enterprise that depends on many people who are not themselves scientists, yet without whom scientific progress would not be possible. Our physics department is no exception. Thank you, **Jon Deasy**, for all the work you do to keep our group afloat. We would go nowhere without you. Thank you **Carlos Bustamante** and **Espe** for keeping the department running as safely and efficiently as possible we are all indebted to you. Thank you, **AJ**, for maintaining the SCIOS.

A special shout out the homies **Bic, Tino, Oscar, Mark, Shrekarardo** and **Luna** who have shared this PhD journey with me in real time via the *shat* through the highs, the lows, and the occasional scientifically questionable life decisions. I am grateful to my master's advisor, **Dr. Claudia Ojeda-Aristizabal**, for giving me my first opportunity in physics research. I would like to acknowledge the American Physical Society Bridge Program, of which I am an alumnus. Without their support at California State University Long Beach, I would not have been able to pursue a graduate education in physics.

I would like to thank the Men's group at Newman Hall Holy Spirit Catholic Center, particularly **Nigel, Pranav, Austin, Michael, Jeff, Jimmy, Alex, Vicente** and from the Women's group **Noella, Abby** and **Immaculada**. "*As the world continues in death, we will carry the cross of life for we clearly see the hell around us, but chose to be crucified personally, mystically; for crucifixion is the only way to resurrection*". A warm thank you to **Fr. John** and **Fr. Alberto** and **Elizabeth** and all the faithful of Saint Joseph the Worker Catholic Church.

Thank you, **Robert Kealhofer**, for your mentorship and friendship during my first year at Cal. I greatly treasure our conversations over Friday pizza and beers at La Val's, and I am grateful for your patience in entertaining all of my questions, no matter how stupid they

may have been. I am indebted to my friend **Aditya N. Prasad**, without whose support through countless problem sets I would not have survived the demanding first year. I carry that conversation we had in our GSI office close to my heart. I am truly grateful for your kindness. May the Lord Jesus bless you wherever you may be.

It was a great disappointment when I first discovered that Cal has one of the most atrocious gyms I have ever stepped foot in. There is a nonzero probability that I would have declined Cal's offer had I toured the gym beforehand. The Lord works in mysterious ways. A special shout out to **City Sports Gym** on San Pablo for providing a sanctuary to lift heavy weights, and to all the strong men and women I have met there over the years. You have all been a tremendous source of motivation. Stay hungry!

To **Ronald, Rudy, Jose, and Jerry**, our friendship began many years ago at Riverside City College, and we have all gone through our respective PhD programs. Alas, I am the last among us to finally cross the finish line! To my friends back home, **Ivan** and **Roger**, thank you for your patience through these years of selfishness and for always welcoming me with gracious hospitality and copious beers during my visits home. Thank you to **my parents** and **family**, for your love and support through all these years away from home. And to my beloved **Deanna**, whose support through the twilight of my PhD has been filled with great memories and countless laughs. I look forward to the next chapter of our lives together and all that the Great Lord has in store for us.

Pater de caelis, Deus, miserere nobis
Fili, Redemptor mundi, Deus, miserere nobis
Spiritus Sancte Deus, miserere nobis
Sancta Trinitas, unus Deus, miserere nobis
Cor Iesu, tabernaculum Altissimi, miserere nobis

Chapter 1

Introduction and Background

“Looking to science and nature, we can only see the truths of physics revealed in the universe...two things are realized: (1) the universe is upheld by an unseen principle of truth. (2) Man, who is part of the universe, is totally subject to these unseen realities of truth.”

— John Marler
Youth of the Apocalypse

1.1 Intercalated Transition Metal Dichalcogenides

Transition Metal Dichalcogenides

Transition metal dichalcogenides (TMDC) with the general formula MX_2 (where M is a transition metal such as Mo, W, Ta, Nb, or Ti and X is chalcogen such as S, Se, or Te.), constitute a broad family of layered van der Waals materials that exhibit a remarkable diversity of novel phenomena ranging from superconductivity, charge density waves (CDW), exciton dynamics light-matter interactions and magnetism. These materials generally crystallize in two common polytypes: the octahedral 1T- MX_2 and the trigonal prismatic 2H- MX_2 structures. In the 1T phase, the transition-metal atoms are coordinated by chalcogen atoms in an octahedral geometry, resulting in metallic or CDW behavior as exemplified by TaS₂ [2, 3] and TiSe₂ [4]. In contrast, the 2H phase features trigonal prismatic coordination, which often stabilizes semiconducting or superconducting states, as in MoS₂ [5], NbS₂ [6] and NbSe₂ [7].

In the 2H polytype, the transition-metal atom M is trigonal-prismatically coordinated by the chalcogenide atom X , and the crystal structure consists of two $X - M - X$ layers stacked in an ABAB sequence within the hexagonal space group P6₃/mmc. By contrast, in the 1T polytype the transition-metal atom M is octahedrally coordinated by the chalcogen

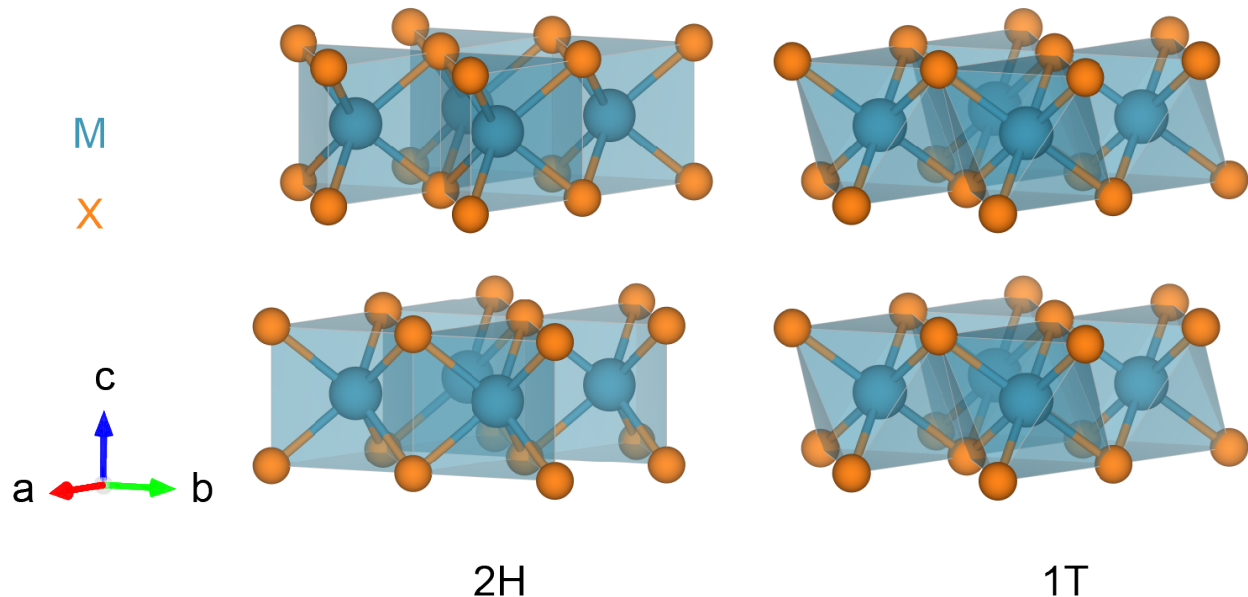


Figure 1.1: **2H and 1T polytypes of TMDs** The two most common polytypes of the family of TMDs.

atoms X , and the structure consists of a single $X - M - X$ layer per unit cell stacked in an AA sequence, resulting in a trigonal lattice with space group $P\bar{3}m1$.

These layered van der Waals materials can be viewed as stacks of hexagonal chalcogen layers with transition-metal atoms occupying the interstitial sites between them, as illustrated in Figure 1.1. The separation between adjacent chalcogen layers defines the van der Waals gap. Variations in the stacking sequence and the relative coordination of the chalcogen hexagons give rise to the different polytypes described above. As a result, these materials exhibit strong in-plane electronic bonding and weak van der Waals bonding along the out-of-plane direction [8]. It is this weak out-of-plane bonding that gives rise to the material's propensity for mechanical exfoliation.

Intercalated Transition Metal Dichalcogenides

Intercalation is the process by which atoms, ions, or molecules are inserted into the van der Waals gaps between the layers of a layered material without destroying the host crystal lattice. In layered van der Waals materials, intercalations involves the insertion of these alien species into the chalcogen-chalcogen gap, minimal disruption of the strong in-plane electronic bonding, and preservation of the overall layered framework of the host material.

The intercalation of layered transition-metal dichalcogenides has been studied since the 1960's, beginning with early work demonstrating that foreign atoms can be reversibly inserted into the van der Waals gaps to systematically tune the electronic, magnetic, and structural properties of these materials [8, 9]. This thesis focuses on the intercalation of magnetic 3d transition-metal ions into layered transition-metal dichalcogenides. In intercalated TMD systems of the form T_xMX_2 , the intercalants can form long-range positional order at specific commensurate concentrations, most notably at $x = 1/3$ and $x = 1/4$, where ordered intercalant superstructures are stabilized. At these compositions the intercalants form commensurate two-dimensional superlattices within the hexagonal van der Waals gaps, specifically, a $\sqrt{3} \times \sqrt{3}$ triangular lattice at $x = 1/3$ and 2×2 superstructure at $x = 1/4$. Figure 1.2 compares the two crystal phases. In the bottom panel, the primitive 2H- MX_2 unit cell is highlighted in red, while the commensurate intercalant superlattice unit cell is indicated in black. In $T_{1/3}MX_2$, the unit cell parameters are $a = b = \sqrt{3}a_o$ and $c \approx c_o$ where a_o and c_o are the lattice parameters of the 2H- MX_2 system. In $T_{1/4}MX_2$, the unit cell parameters are $a = b = 2a_o$ and $c \approx c_o$ [10].

The centrosymmetric space group $P6_3/mmc$ of the parent 2H- MX_2 system is broken upon intercalation at the commensurate level $x = 1/3$. At this composition, the $T_{1/3}MX_2$ system adopts the noncentrosymmetric space group $P6_322$. This reduction in symmetry arises because the intercalant ions can occupy one of two symmetry-inequivalent Wyckoff positions (2c or 2d) within the van der Waals gap, each of which lacks an inversion center [10, 9]. The broken inversion symmetry allows additional spin-orbit-coupled interactions, including Dzyaloshinskii-Moriya exchange and Rashba-type spin splitting, which can strongly influence the magnetic and transport properties of these materials. In contrast, for $x = 1/4$, the $T_{1/4}MX_2$ system retains the original centrosymmetric space group $P6_3/mmc$ of the parent transition-metal dichalcogenide.

Spin State

The intercalated transition-metal ions generally occur in 2+ or 3+ oxidation states and experience a crystal field produced by the neighboring chalcogen atoms (S or Se). This crystal field lifts the degeneracy of the 3d orbitals ($L = 2$) and strongly influences the resulting spin state and magnetic anisotropy. The electronic Hamiltonian that describes the intercalated atom is given by

$$H = \sum_i \left[\frac{p_i^2}{2m} - \frac{Ze^2}{r_{i\mu}} + \sum_j \left[\frac{e^2}{r_{ij}} + \xi_{ij} \mathbf{L}_i \cdot \mathbf{S}_j + \gamma_{ij} \mathbf{J}_i \cdot \mathbf{I}_\mu \right] \right] + V_{cf} \quad (1.1)$$

where m is the mass, Z is the core electron number, i and j are the electron indices, μ is the nuclear index and $\mathbf{I}$ is the nuclear spin. The spin-orbit interaction (with a typical energy scale of ~ 10 meV) and the hyperfine interaction (which is approximately 10^{-5} times weaker than the dominant electronic interactions) can be neglected at leading order. As a result,

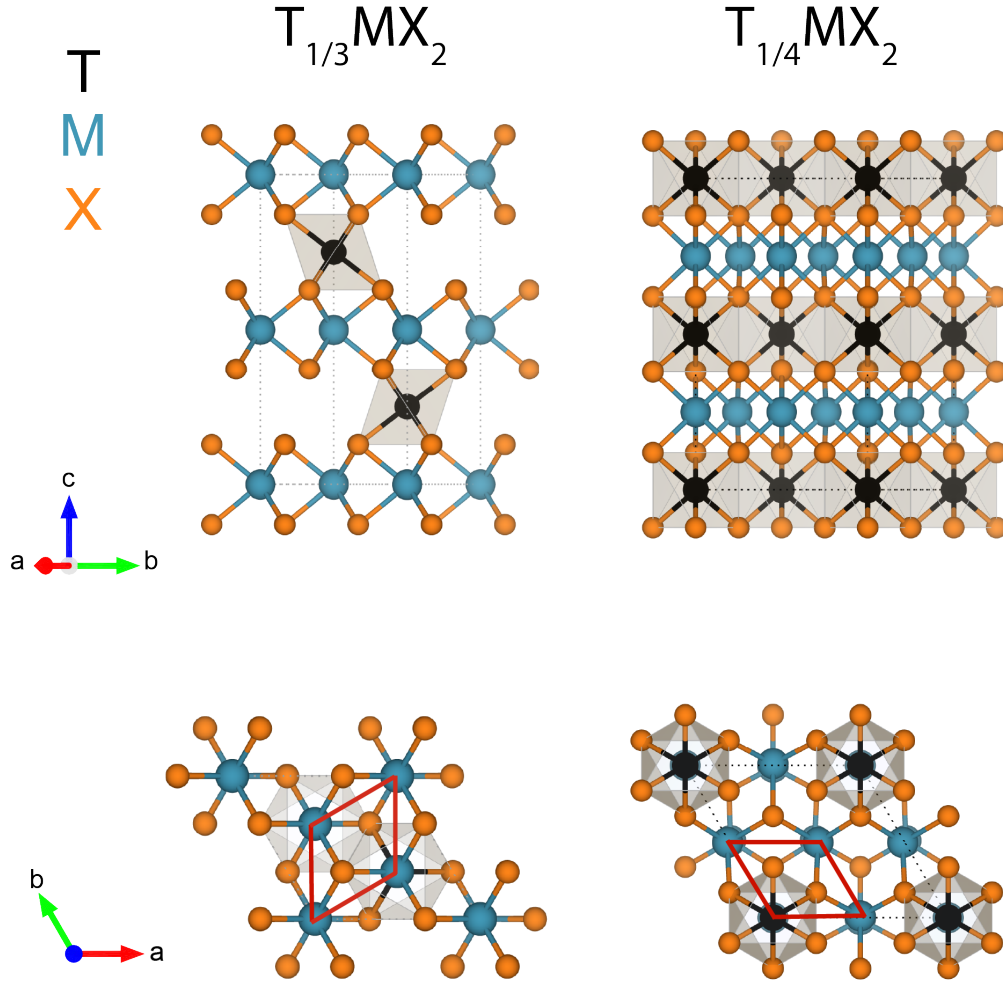


Figure 1.2: **Unit cell of the intercalated TMDs $T_{1/3}MX_2$ (left) and $T_{1/4}MX_2$ (right)** Bottom panel is the unit cells along the c axis with unit cell and 2H-MX₂ highlighted in red.

the crystal-field potential V_{cf} is taken as the primary perturbation to the standard electronic Hamiltonian. The energy eigensates of the unperturbed system are given by the spherical harmonics $Y_m^l(\theta, \phi)$.

The 3d electrons of the intercalated transition-metal ion have orbital angular momentum $L = 2$, corresponding to magnetic quantum numbers $m_l = -2, -1, 0, 1, 2$ and are therefore fivefold degenerate in the absence of a crystal field. Because the intercalated ion is octahedrally coordinated by the surrounding chalcogen atoms of the parent TMD, the crystal-field potential lifts this degeneracy, splitting the 3d levels into a twofold-degenerate e_g manifold

and a threefold-degenerate t_{2g} manifold. In addition, the octahedral environment is subject to a trigonal (threefold) distortion along the crystallographic c axis, which further lifts the degeneracy of the t_{2g} states, splitting them into a nondegenerate a_{1g} level and a twofold-degenerate e'_g level (see Figure 1.3 (a)). The crystal-field splitting of the 3d orbitals and their further symmetry lowering under trigonal distortion are well described within ligand-field theory [11, 12].

Once the 3d band splitting is known, the spin states can be determined. The competition between the crystal field energy and the Coulomb energy cost of pairing two electrons will determine the order in which the orbitals states are filled [13]. For example, Co^{+2} has an electron configuration of $[\text{Ar}]3d^7$ and there is two possible ways to fill the spin states which are shown in Figure 1.3 (b).

According to Hund's rules, the five d orbitals are first singly occupied by electrons with parallel spin (m_s values) before any orbital becomes doubly occupied. Additional electrons then occupy the lower-energy orbitals with opposite spin. In this case, three electrons remain unpaired, resulting in a total spin $S = 3/2$ (left panel of Figure 1.3 (b)). This configuration corresponds to the high-spin state, which is favored when the crystal-field splitting energy is smaller than the intra-orbital pairing energy (the weak-field limit).

In contrast, when the crystal-field splitting energy exceeds the pairing energy (the strong-field limit), electrons preferentially doubly occupy the lower-energy orbitals. This leads to a configuration with only one unpaired electron in the highest-energy level, giving a total spin $S = 1/2$ (right panel of Figure 1.3 (b)). This configuration is known as the low-spin state.

Spin Hamiltonian

The intercalated TMD of the type T_xMX_2 can be described by the following spin Hamiltonian

$$H = - \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_i A (\mathbf{S}_i \cdot \mathbf{n}_i)^2 + \sum_{\langle ij \rangle} \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) - g\mu_B \sum_i \mu_o \mathbf{H} \cdot \mathbf{S}_i \quad (1.2)$$

The first term in Equation 1.2 corresponds to the Heisenberg exchange interaction between neighboring spins, as described in section 1.1. The second term represents the single-ion magnetocrystalline anisotropy, which arises from the interplay between the crystal field and spin-orbit coupling through the partially unquenched orbital degrees of freedom. Depending on the sign and magnitude of the anisotropy parameter, this term can give rise to isotropic, easy-axis, easy-plane, or easy-cone anisotropies. The third term is the Dzyaloshinskii–Moriya interaction (DMI), which originates from spin-orbit coupling in the absence of inversion symmetry and $\mathbf{D}_{ij}$ is a bond-dependent vector whose direction is fixed by the lattice symmetry; it energetically favors canting and can stabilize chiral/noncollinear textures. The last term is the Zeeman term coupling to an external magnetic field. At $x = 1/3$, the noncentrosym-

metric structure ($P6_322$) permits nonzero $\mathbf{D}_{ij}$, whereas in centrosymmetric phases ($x = 1/4$, $P6_3/mmc$) the symmetry can forbid or strongly constrain DMI on certain bonds.

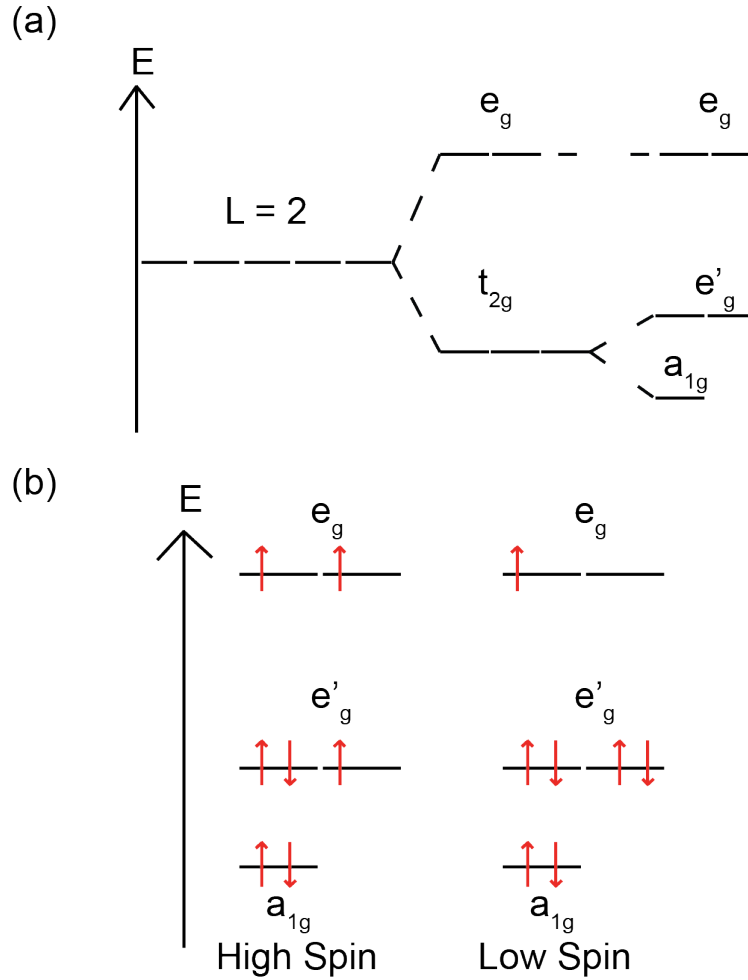


Figure 1.3: **d orbital splitting and spin configuration of the intercalated transition metal.** (a) In the absence of a crystal field, the five 3d orbitals ($L = 2$) are degenerate. Octahedral coordination by the surrounding chalcogen atoms splits the 3d manifold into a twofold-degenerate e_g level and a threefold-degenerate t_{2g} level. A subsequent trigonal distortion along the crystallographic c axis further lifts the degeneracy of the t_{2g} states, resulting in a nondegenerate a_{1g} level and a twofold-degenerate e'_g level. (b) The spin configuration of Co^{+2} in the resulting crystal field (D_{3d})

1.2 Magnetism

In a ferromagnetic material, unpaired electron spins have the proclivity to align parallel to those of their neighbors, whereas in an antiferromagnet the spins preferentially align antiparallel. Both behaviors can be described by the Heisenberg exchange interaction, whose Hamiltonian is given by

$$H = - \sum_{i,j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j \quad (1.3)$$

where J_{ij} is the exchange constant that quantifies the strength of the interaction between spins $\mathbf{S}_i$ and $\mathbf{S}_j$. The sign of J_{ij} determines the nature of the magnetic coupling: a positive exchange constant ($J_{ij} > 0$) favors ferromagnetic alignment, while a negative exchange constant ($J_{ij} < 0$) favors antiferromagnetic alignment.

The total wavefunction of a two-electron system can be written as

$$\Psi = \frac{1}{\sqrt{2}} [\psi_a(\mathbf{x}_1)\psi_b(\mathbf{x}_2) \pm \psi_a(\mathbf{x}_2)\psi_b(\mathbf{x}_1)] \chi \quad (1.4)$$

where ψ_a and ψ_b are the spatial wavefunctions of the two electrons, $\mathbf{x}_1$ and $\mathbf{x}_2$ denote their coordinates, and χ is the spin wavefunction of the two-electron system. The total wavefunction Ψ must be antisymmetric under particle exchange in order to satisfy the Pauli exclusion principle, which governs the behavior of indistinguishable fermions. When the plus sign in Equation 1.4 is chosen, the spatial part of the wavefunction is symmetric under particle exchange. Consequently, the spin wavefunction χ must be antisymmetric, corresponding to a spin-singlet state in which the two electron spins are antiparallel. Conversely, when the minus sign is chosen, the spatial wavefunction is antisymmetric and the spin wavefunction must be symmetric. In this case, the electrons occupy a spin-triplet state, and their spins preferentially align parallel to one another.

The antisymmetric and symmetric spin wavefunctions corresponding to Equation 1.4 can be written explicitly in terms of the single-electron spin states $\uparrow$ and $\downarrow$. The antisymmetric spin-singlet state is

$$|\chi_s\rangle = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle), \quad (1.5)$$

which has total spin $S = 0$. The symmetric spin-triplet states are given by

$$\begin{aligned} |\chi_{t,+1}\rangle &= |\uparrow\uparrow\rangle \\ |\chi_{t,0}\rangle &= \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle) \\ |\chi_{t,-1}\rangle &= |\downarrow\downarrow\rangle \end{aligned} \quad (1.6)$$

The energetic preference between the singlet and triplet configurations originates from the Coulomb interaction combined with the symmetry of the spatial wavefunction. When

the spatial wavefunction is symmetric (spin singlet), the probability amplitude for the two electrons to occupy the same region of space is enhanced, increasing their Coulomb repulsion. Conversely, an antisymmetric spatial wavefunction (spin triplet) suppresses this overlap, reducing the Coulomb energy. This difference in Coulomb energy gives rise to the exchange energy, which favors either parallel or antiparallel spin alignment depending on the microscopic details of the electronic orbitals and their overlap. corresponding to total spin $S = 1$.

The Heisenberg exchange interaction can arise from either direct or indirect exchange mechanisms. Direct exchange originates from the Coulomb interaction between neighboring electrons, where the antisymmetry of the electronic wavefunction allows electrons with parallel spins to reduce their Coulomb energy through exchange [14]. In contrast, indirect exchange mechanisms—such as superexchange [15, 16], double exchange [17], and the Ruderman–Kittel–Kasuya–Yosida (RKKY) [18, 19, 20] interaction—are mediated by intermediate states or particles. In the RKKY interaction, for example, itinerant conduction electrons mediate an effective exchange coupling between localized magnetic moments, leading to an interaction that oscillates in sign and magnitude as a function of the distance between spins.

To illustrate this consider the response of an electron gas to a spatial varying magnetic field which will provide a perturbation to the Hamiltonian of the form

$$\delta H = \pm \frac{g\mu_o\mu_B}{2} H_{\mathbf{q}} \cos(\mathbf{q} \cdot \mathbf{r}) \quad (1.7)$$

where $\pm$ refers to the spin of the electron. The plane waves

$$\psi_{\mathbf{k}\pm}(\mathbf{r}) = \frac{1}{\sqrt{V}} \exp[i\mathbf{k} \cdot \mathbf{r}] |\pm\rangle \quad (1.8)$$

experience the perturbation given by Equation 1.7 and the amplitude of the perturbed state and magnetization can be calculated by first order perturbation theory to give

$$M(\mathbf{r}) = \frac{g^2\mu_o\mu_B^2 m_e H_{\mathbf{q}} \cos(\mathbf{q} \cdot \mathbf{r})}{\hbar^2 V} \left[\frac{1}{(\mathbf{k}^2 + \mathbf{q})^2 - k^2} + \frac{1}{(\mathbf{k}^2 - \mathbf{q})^2 - k^2} \right] \quad (1.9)$$

Integrating equation 1.9 over k -space weighted by the density of states gives the total magnetization

$$M_{\mathbf{q}} = \int_{k < k_f} g(\mathbf{k}) d^3 \mathbf{k} M(\mathbf{r})$$

$$M_{\mathbf{q}} = \frac{k_f m_e g^2 \mu_o \mu_B H_q}{\pi^2 \hbar^2} \left[1 + \frac{4k_f^2 - q^2}{4k_f q} \log \left| \frac{q + 2k}{q - 2k_f} \right| \right] \quad (1.10)$$

A local magnetic moment can be considered as a delta-function perturbation to the magnetic field, $\mathbf{H}(\mathbf{r}) = \delta(\mathbf{r})\mathbf{H}$, and performing a Fourier transform to obtain the real-space dependence of the spin susceptibility χ ,

$$\chi(r) = \frac{1}{(2\pi)^3} \int d^3 \mathbf{q} e^{i\mathbf{q} \cdot \mathbf{r}} \chi_q$$

$$\chi(r) = \frac{1}{(2\pi)^3} \int d^3 \mathbf{q} e^{i\mathbf{q} \cdot \mathbf{r}} \frac{k_f m_e g^2 \mu_o \mu_B}{\pi^2 \hbar^2} \left[1 + \frac{4k_f^2 - q^2}{4k_f q} \log \left| \frac{q + 2k}{q - 2k_f} \right| \right] \quad (1.11)$$

$$\chi(r) \propto k_f^2 \frac{-2k_f r \cos(2k_f r) + \sin(2k_f r)}{(2k_f r)^4}$$

These calculations can be found in detail in Reference [13]. The indirect exchange interaction J_{RKKY} can be shown to be proportional to the static susceptibility either by second order perturbation theory or linear-response analysis, such that $J_{RKKY} \propto \chi(r)$ [19, 21]. One therefore arrives at the characteristic RKKY form,

$$J_{RKKY} \propto \frac{\cos(2k_f r)}{r^3} \quad (1.12)$$

where k_f is the Fermi wavevector, and r is the separation between localized magnetic moments. The RKKY interaction plays a significant role in the family of intercalated transition metal dichalcogenides where it constitutes a dominant magnetic exchange mechanism in several well-studied systems. Notable examples include $M_{1/3}\text{NbS}_2$ ($M = \text{V, Cr}$) and $M_{1/3}\text{TaS}_2$ ($M = \text{V, Cr, Mn, Fe, Co, Ni}$) [22], Fe_xNbS_2 [23, 24], $\text{Fe}_{1/4}\text{TaS}_2$ [25], $\text{Mn}_{1/4}\text{NbS}_2$ [26].

1.3 Spintronics

The field of spin transport electronics, or spintronics, studies how both the charge and spin (and its associated magnetic moments) degrees of freedom of the electron can be exploited

and integrated into solid-state electrical devices. Leveraging the spin degrees of freedom allows the efficiency and data storage capabilities of a traditional charged-based electrical device to increase dramatically. Giant magnetoresistance (GMR) was first observed in Fe/Cr multilayers, where the electrical resistance of the system could be strongly modified by the alignment and realignment of adjacent ferromagnetic layers under an applied external magnetic field. The discovery of GMR is considered the start of spin-based electronics [27]. Since the discovery of giant magnetoresistance, spintronics has expanded from a basic scientific phenomenon into a vibrant research area and a rapidly advancing technological field. New fundamental mechanisms such as spin-transfer torque (STT) [28, 29], spin-orbit torque (SOT) [30, 31], and voltage-controlled magnetic anisotropy (VCMA) [32] have enabled new generations of memory technologies (e.g., MRAM), highly sensitive sensors, and prototype devices for future computing paradigms. At the same time, the range of material platforms has broadened beyond ferromagnets to include ferroelectrics, antiferromagnets, and topological insulators, greatly enriching both the physics and applications of spintronics.

Spin Transfer Torque

A magnetic configuration that is out of equilibrium and experiences dissipation, the magnetization ($\mathbf{M}$), precesses around the local effective field $\mathbf{H}_{\text{eff}}$. The equations of motion of such a system is governed by the Landau-Lifshitz-Gilbert (LLG) equation:

$$\dot{\mathbf{M}} = -\gamma_0 \mathbf{M} \times \mathbf{H}_{\text{eff}} + \frac{\alpha}{M_s} \mathbf{M} \times \dot{\mathbf{M}} \quad (1.13)$$

where γ_0 is the gyromagnetic ratio, α is the damping parameter. The first term in Equation 1.13 describes the precession and the second describes the damping. In the absence of damping the magnetization will simply precess around the magnetic field direction, Figure 1.4 (a). If the system exhibits dissipation, the magnetization precession will eventually decay and the magnetization will be aligned with the magnetic field, Figure 1.4 (b). When a spin polarized current, which carries angular momentum, passes through a magnetic material it will impart spin angular momentum to the local magnetization. This addition of spin angular momentum can induce a torque on the magnetic layer and is given by the Slonczewski spin-transfer torque term:

$$\boldsymbol{\tau}_{STT} \sim \mathbf{M} \times \mathbf{p} + \mathbf{M} \times (\mathbf{M} \times \mathbf{p}) \quad (1.14)$$

where the first term is a field like torque that acts like an additional magnetic field in the direction of polarization ($\mathbf{p}$) and the second is a damping like torque that pulls the magnetization toward (or away from) the polarization direction $\mathbf{p}$ of the injected current. The exchange interaction between the spins of the conduction electron can exert a torque on the localized spins. This allows for current-driven manipulation of the magnetization of the localized spins [33, 34]. Spin-transfer torque (STT) devices have several limitations.

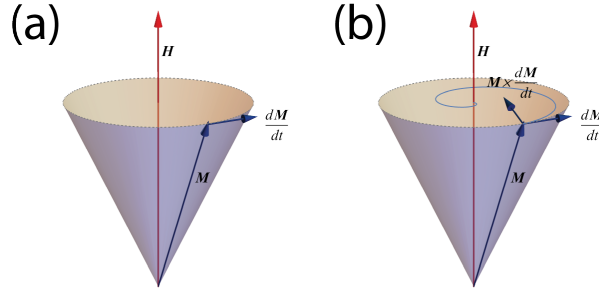


Figure 1.4: **Magnetization trajectory under LLG equation** (a) For a magnetic moment, $\mathbf{M}$, in a magnetic field, $\mathbf{H}$ with no damping will precess around the applied field. (b) Magnetization Trajectory given by the LLG. The magnetization vector will precess around the applied field and will spiral inward until it is aligned with the applied field.

They require two magnetic layers—a fixed and a free layer—and the electric current must pass directly through the magnetic materials. As a result, STT typically relies on vertical heterostructure designs, which can lead to higher power consumption, reduced endurance, and fabrication challenges [35, 36].

Spin Orbit Torque

Spin–Orbit Torque (SOT) is a mechanism by which an electric current in a material with strong spin–orbit coupling (SOC) can exert a torque on an adjacent ferromagnetic or anti-ferromagnetic layer, thereby manipulating its magnetization without an external magnetic field. When an applied charge current is passed through a system with broken inversion symmetry, it will generate a non-equilibrium spin polarized current. This spin polarized current will transfer spin angular momentum to localized moments. There are two microscopic mechanisms that can drive SOT, either by the spin Hall effect (SHE) or by Rashba–Edelstein Effect (REE).

$$\boldsymbol{\tau} = \tau_{\text{FL}} \mathbf{M} \times \boldsymbol{\sigma} + \tau_{\text{DL}} \mathbf{M} \times (\mathbf{M} \times \boldsymbol{\sigma}) \quad (1.15)$$

where $\mathbf{M}$ is the local magnetization and $\boldsymbol{\sigma}$ is the spin polarization induced by the applied current. The first term in Equation 1.15 comes from the interfacial REE spin accumulation and the second comes from the spin current generated by (SHE) [37]. Both types of torques have been used to electrically switch the magnetization in novel devices. Miron et al. [38] demonstrated that an in-plane current can switch the magnetization of a single ferromagnetic layer in a Pt/Co/AlO_x structure through a Rashba-type spin–orbit torque, arising from interfacial spin–orbit coupling and producing a field-like torque. In contrast, Liu et al. [39]

showed that a current in a Ta/CoFeB/MgO bilayer generates a damping-like torque via the spin Hall effect in tantalum, efficiently switching the adjacent ferromagnet.

Both STT and SOT form an alternative hypothesis for the switching results found in chapter 4 and chapter 6 but are ultimately ruled out by the experimental observations.

Heat Assisted Magnetic RAM

The ever-increasing demand for fast, reliable, and energy-efficient information storage continues to push magnetic memory technologies toward their fundamental limits. Conventional

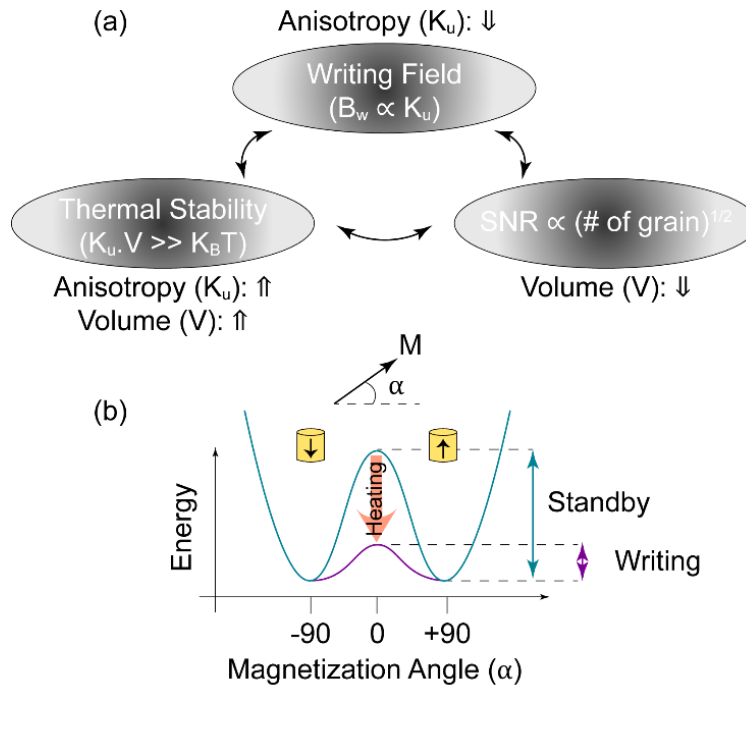


Figure 1.5: **Magnetic recording trilemma and HAMR concept** (a) The trilemma of magnetic recording: increasing areal density by shrinking bit size imposes contradictory requirements for simultaneously maintaining reasonable SNR (depends on volume, V), thermal stability (depends on anisotropy, K_u , and V), and writing field (B_w , depends on K_u). Optimizing one metric necessarily compromises another. (b) HAMR circumvents this limitation: transiently heating the sample above its Curie temperature reduces the anisotropy barrier, enabling bit flipping with modest fields during the write cycle. Upon cooling, the magnetic anisotropy is restored to a large value, ensuring thermal stability for long-term data retention.

scaling strategies, which rely on reducing bit size, inevitably run into the “magnetic recording trilemma,” where signal-to-noise ratio, thermal stability, and writability cannot all be optimized simultaneously. Heat-assisted magnetic recording (HAMR) has emerged as the leading solution, enabling high-density storage by transiently heating the medium during the write cycle. However, the reliance on laser optics and plasmonic transducers restricts HAMR primarily to hard-disk drives, limiting its integration with on-chip or embedded architectures.

The exponential growth of digital data underpins nearly every aspect of modern life, from cloud computing and artificial intelligence to scientific discovery and personal electronics. Meeting this demand requires storage technologies that are not only scalable in density but also energy-efficient and reliable over long timescales. Magnetic and spintronic memories stand out in this regard as the intrinsic non-volatility of magnetization offers low-energy data retention, while their compatibility with fast electronic readout makes them attractive for high-performance applications [40, 41]. Yet, realizing their full potential depends critically on discovering and engineering new materials that can balance thermal stability, writability, and integration with emerging device architectures. In this context, quantum materials and particularly emerging two-dimensional (2D) magnets with tunable anisotropy and Curie temperatures provide unique opportunities to be used as the recording media. The structural versatility and tunable magnetic interactions in 2D magnets expand the design space well beyond conventional alloys, enabling exploration of recording schemes that combine fundamental novelty with technological advancements.

At the center of every magnetic storage technology lies its areal density, which determines how much information can be encoded within a given surface area and ultimately sets the limit for storage capacity. To increase the areal density of the recording media, we need to reduce the size of magnetic bits. This seemingly obvious strategy, however, faces a trilemma that we schematically illustrate in Figure 1.5 (a). The trilemma stems from simultaneously satisfying three major performance measures: signal-to-noise-ratio (SNR), thermal stability, and the magnetic field used for writing (B_w , a measure of writability). In reality, each data bit is a granular area composed of multiple grains, making the boundary between adjacent bits rather rough (not a straight line). As such, to maintain high SNR while shrinking the bit size, the number of grains per bit should be kept constant (~ 10), or the edge roughness reduces the SNR [42, 43]. Thus, reducing the bit size requires reducing the grain volume (V). However, reducing the volume increases the risk of data corruption as the magnetization becomes susceptible to thermal instability (i.e., random bit flipping) due to the superparamagnetic effect [44]. To avoid this issue, the magnetic anisotropy (K_u) of grains should be increased so that the energy barrier for bit flipping (proportional to $K_u \times V$) is sufficiently above the thermal energy (i.e., $K_u V > K_B T$) [45]. Increasing anisotropy, however, directly increases the writing field (B_w) required for the magnetization reversal because the coercivity of the grains scale with the magnitude of the anisotropy, that is $B_w \propto K_u$ [46]. As such, satisfying all three performance measures, while shrinking the bit size, becomes virtually impossible,

hence the trilemma.

To break out of the trilemma, the concept of energy-assisted magnetic recording (EAMR) is introduced and different variants of it have been developed [47, 48, 49]. In EAMR, the idea is to maintain high SNR and robust thermal stability via designing magnetic materials with large K_u and small V , but to alleviate the large B_w penalty, an external source of energy is supplied during the data-writing cycle to assist with the magnetization reversal (Figure 1(b)), hence satisfying the third performance measure. The extra energy is often provided in the form of either microwave [49] or heat, [48] the latter of which has gained widespread adoption and lead to the heat-assisted magnetization recording (HAMR). In the HAMR approach, a laser pulse is used to heat the magnetic media to above its Curie temperature, which temporarily reduces the magnetic anisotropy, hence allowing writing (i.e., reversing) the magnetization direction using a reasonably small writing field, B_w . Following the removal of the laser pulse, the medium restores its large anisotropy for stable data retention. To go beyond the diffraction-limitation of the light, the laser is focused on a plasmonic transducer which enables confining the heat to a smaller spot size, roughly the lateral size of a single bit of data [50]. Although the read-head of hard disk drive can accommodate such bulky additions (lasers, plasmonic transducers, etc.), a similar setup is not appealing for integrated solutions and embedded memory architectures. Chapter 4 explores an electronic variant of HAMR, in which magnetization reversal is assisted not by laser heating but by Joule heating from current pulses with densities as low as $10 \times 10^4 \text{ A cm}^{-2}$. Combined with electronic readout via the anomalous Hall effect, this approach enables an all-electronic read-write scheme for magnetization recording.

1.4 Switching in Antiferromagnetics Materials

Železný *et al*, predicted that that under certain symmetry considerations an in-plane applied current could generate a Neel-order spin-orbit torque (NSOT) fields that can switch the system between two distinct resistance states. The NSOT are analogous to the SOT described above. By considering an AFM where the two spin sub-lattices form space-inversion partners that individually have broken inversion symmetry. Each sublattice gives rise to an inverse spin galvanic effect with opposite sign, resulting in the NSOT field [51]. The anit-damping term (second term in Equation 1.15) becomes

$$\boldsymbol{\tau}^{A/B} \sim \mathbf{M}^{A/B} \times (\hat{\mathbf{z}} \times \mathbf{J}) \quad (1.16)$$

where $\mathbf{M}^{A/B}$ is the magnetization of A and B sublattice and the term $\hat{\mathbf{z}} \times \mathbf{J}$ is a staggered effective field due to the opposite magnetization on the two sublattices. It is this staggered field that allows 90° reorientation of the AFM by the applying an in-plane current

between two orthogonal directions. This was first predicted in Mn_2Au [51] and subsequently experimentally observed in CuMnAs [52] and Mn_2Au [53].

Helical AFM

In a helical AFM, the magnetization rotates in the plane perpendicular to the $\mathbf{q}$ -vector. The $\mathbf{q}$ -vector defines the direction and periodicity of the long-range spiral arrangement of the magnetic spins. Helimagnets are generally favored in systems with non-centrosymmetric symmetry and with an appreciable Dzyaloshinskii-Moriya (DM) interaction. The DM interaction is a first-order correction to the exchange interaction when spin-orbit coupling is treated as a perturbation and the magnitude of the DM interaction is proportional to the SOC interaction strength and the symmetric exchange constant, $D \sim \lambda J$ [54].

In the chiral antiferromagnet Mn_3Sn it was recently shown that the anomalous Hall effect (AHE) could be electrically switched in the presence of a bias field [55]. Similarly, the helically ordered intercalated transition-metal dichalcogenide $\text{Ni}_{1/3}\text{NbS}_2$ was shown to respond to applied currents [56]. In both cases the current was applied perpendicular to the helical axis. It is worth noting that most studies in this area use currents in the range of $\sim 10^7 - 10^9 \text{ A/cm}^2$ which is similar to many of the original works on AFM switching known to cause significant heating. In thin films of FeGe , a different type of helical switching was observed where the helices are “combed” along the axis of the applied current [57]. When an applied current is large enough to overcome the pinning strength in a helical AFM, the dynamics of the magnetization is governed by the following LLGS equation

$$\dot{\mathbf{M}} = -\gamma \mathbf{M} \times \mathbf{B}_{eff} + \alpha \mathbf{M} \times \dot{\mathbf{M}} + \frac{P\mu_{\mathbf{B}}}{eM_s(1 + \beta^2)} \left[(\mathbf{j} \cdot \nabla) \mathbf{M} - \beta \mathbf{M} \times (\mathbf{j} \cdot \nabla) \mathbf{M} \right] \quad (1.17)$$

where γ is the gyromagnetic ratio, $\mathbf{B}_{eff}$ is the effective magnetic field, P is the spin polarization, $\mathbf{j}$ is the current density, and β is the nonadiabatic damping parameter. Here a current will cant the spins by an angle ϕ , creating a cone pointing along the helix. Above a certain critical current, this will align the helical order with $\mathbf{j} \parallel \mathbf{q}$. Such systems will also exhibit a kind of Walker breakdown [58, 57], whereby $\phi \rightarrow 0$ when the current leading to a precession of the domains' spins and energy dissipation that prohibits their motion.

Magnetization dynamics are inherently stochastic at finite temperature due to thermal fluctuations arising from coupling to the lattice. Following Brown's formalism, these fluctuations can be incorporated into Equation 1.17 by introducing an effective time-dependent random thermal field, $\mathbf{h}(t)$, such that

$$\dot{\mathbf{M}} = -\gamma \mathbf{M} \times \mathbf{h}(t) + \dot{\mathbf{M}}' \quad (1.18)$$

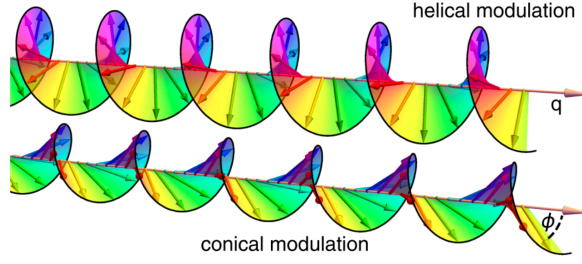


Figure 1.6: **Helimagnetic Ordering and Conical Modulation** Helical phase where the magnetization vector rotates in the plane perpendicular to the helical axis. state out of equilibrium into a conical state where the magnetization vector tilts towards the $\mathbf{q}$ -vector. Image taken from reference [57].

where $\dot{\mathbf{M}}$ denotes the deterministic contribution given by Equation 1.17. The stochastic field $\mathbf{h}(t)$ is modeled as a Gaussian white-noise process with zero mean, delta-correlated fluctuations in time, and isotropic correlations in space [59, 60] :

$$\langle h_i(t) \rangle = 0 \quad (1.19)$$

$$\langle h_i(t) h_j(t') \rangle = \frac{2\alpha k_B T}{\gamma M_s V} \delta_{ij} \delta(t - t'). \quad (1.20)$$

Here, α is the Gilbert damping parameter, k_B is the Boltzmann constant, T is the temperature, M_s is the saturation magnetization, and V is the magnetic volume. The delta function $\delta(t - t')$ indicates that the fluctuations are Markovian, while δ_{ij} reflects the isotropic nature of the random field.

In the context of pulsed-current experiments, transient Joule heating elevates the local temperature during switching events, thereby increasing the amplitude of thermal fluctuations and promoting thermally assisted magnetization reversal or domain wall motion. This effect is particularly relevant in Chapter 6, where the observed switching phase space is governed by the interplay between current-induced torques and thermally activated processes, making it essential to distinguish purely electronic switching mechanisms from those assisted by Joule heating.

1.5 Anomalous Hall Effect in Antiferromagnets

In general, the transverse resistivity ρ_{xy} is given by

$$\rho_{xy} = \rho_o B + R_s \mu_o M + \rho_H^{\text{AF}} \quad (1.21)$$

where the first term in Equation 1.21 is the ordinary hall effect (OHE) which is linear in field. The second term is the anomalous hall effect (AHE) due to any non-zero net magnetization typical of ferromagnetic metals. The last term will give rise to a nonzero hall effect not described by either the OHE or the AHE. In AFM the anomalous hall effect can arise from the Berry curvature.

The intrinsic anomalous hall conductivity (AHC), where $\sigma_{\alpha\beta} = \rho_{\alpha\beta}^{-1}$, is given by

$$\sigma_{\alpha\beta} = -\frac{e^2}{\hbar} \int \frac{d^3k}{(2\pi)^3} \sum_n f(E_n(\mathbf{k}) - \mu) \Omega_{n,\alpha\beta}(\mathbf{k}) \quad (1.22)$$

where n is the band index, $\alpha, \beta = x, y, z$ with $\alpha \neq \beta$, $f(E_n(\mathbf{k}) - \mu)$ is the Fermi-Dirac distribution, and $\Omega_{n,\alpha\beta}(\mathbf{k})$ is the Berry curvature. The Kubo formalism defines the Berry curvature in the AHC by

$$\Omega_{n,\alpha\beta}(\mathbf{k}) = -2\text{Im} \left[\sum_{m \neq n} \frac{v_{nm,\alpha}(\mathbf{k}) v_{nm,\beta}(\mathbf{k})}{[E_m(\mathbf{k}) - E_n(\mathbf{k})]^2} \right] \quad (1.23)$$

where $E_n(\mathbf{k})$ is the eigen value and $v_{nm,\alpha}(\mathbf{k})$ are the off-diagonal velocity matrix elements defined as

$$v_{nm,\alpha}(\mathbf{k}) = \frac{1}{\hbar} \langle u_n(\mathbf{k}) | \frac{\partial H(\mathbf{k})}{\partial k_\alpha} | u_m(\mathbf{k}) \rangle \quad (1.24)$$

where $H(\mathbf{k}) = e^{-i\mathbf{k}\cdot\mathbf{r}} H e^{i\mathbf{k}\cdot\mathbf{r}}$ and $u_n(\mathbf{k})$ is the periodic-cell part of the Bloch states. The symmetry of the Berry curvature will dictate if the AHE is non-zero.

In ferromagnetic systems, spin-orbit coupling links the breaking of time-reversal symmetry by a net magnetization to transverse charge transport, giving rise to the anomalous Hall effect (AHE). In contrast, antiferromagnetic systems can host an AHE through two distinct mechanisms. First, in noncoplanar antiferromagnets, time-reversal symmetry breaking alone can generate a Hall response even in the absence of spin-orbit coupling. In this case, a finite scalar spin chirality, $\mathbf{S}_i \cdot (\mathbf{S}_j \times \mathbf{S}_k)$, produces an emergent real-space magnetic field that acts as a geometrical (topological) Berry curvature, giving rise to the topological Hall effect [61].

Second, in coplanar antiferromagnets, such as the 120° Néel state on a triangular lattice, time-reversal symmetry $\mathcal{T}$ is broken, but the system remains invariant under a combined symmetry operation consisting of time reversal followed by a 180° spin rotation about the

axis normal to the spin plane ($\mathcal{TS}$). Because the spin-rotation operator $\mathcal{S}$ acts only on the spin degrees of freedom and leaves the electronic velocity operators unchanged, this combined $\mathcal{TS}$ symmetry enforces a cancellation of Berry curvature and suppresses the anomalous Hall effect, analogous to the role of $\mathcal{T}$ symmetry in nonmagnetic systems [62]. However, the presence of spin-orbit coupling breaks the $\mathcal{TS}$ symmetry by coupling spin and orbital degrees of freedom, thereby lifting the Berry-curvature cancellation and allowing a finite anomalous Hall response to emerge even in coplanar antiferromagnets [63].

1.6 Electronic Transport in Correlated and Disordered Materials

Electron-electron interactions play a central role in determining the low-temperature behavior of metallic systems and arise fundamentally from the Coulomb repulsion between charged particles. Although these interactions are often screened in metals, they remain important in the low-temperature regime, particularly in disordered systems where electrons propagate diffusively. In such cases, interactions lead to a suppression of the density of states near the Fermi level and give rise to quantum corrections to transport. A hallmark signature of electron-electron interactions in disordered conductors is a $\sqrt{T}$ dependence of the resistivity in three dimensions [64, 65].

In conventional metals, electron correlations can be effectively captured within the framework of Fermi-liquid theory, which maps the many-body problem onto a system of weakly interacting quasiparticles [66, 67, 68]. These quasiparticles retain the quantum numbers of bare electrons, but their properties, such as the effective mass, are renormalized, and they possess finite lifetimes. As a result, electronic transport at low temperatures is governed by quasiparticle scattering processes, most notably electron-electron scattering, which leads to a characteristic resistivity scaling of $\rho \sim T^2$ [67, 68].

The presence of localized magnetic moments introduces a qualitatively different type of interaction known as the Kondo effect. The Kondo effect emerges when conduction electrons interact with localized magnetic impurities, leading to spin-flip scattering processes that become increasingly significant at low temperatures. As the temperature decreases toward the characteristic Kondo temperature T_K , conduction electrons collectively screen the impurity spin, forming a many-body singlet state [69]. This results in an anomalous increase in resistivity, often exhibiting a logarithmic temperature dependence, $\rho \sim \log T$ [70]. At temperatures well below T_K , the system recovers a Fermi-liquid ground state with strongly renormalized quasiparticles [67, 69].

Chapter 2

Synthesis

“Through Him all things were made; without Him nothing was made that has been made.”

— John 1:3

2.1 Introduction

The synthesis of intercalated transition-metal dichalcogenides (I-TMDs) typically involves a two-step process: first, a solid-state reaction is used to prepare polycrystalline precursor compounds, followed by chemical vapor transport (CVT) growth to obtain high-quality single crystals.

Solid State Reaction

The solid state reaction typically follows an additive reaction given by Equation 2.1



where A and B maybe elements or compounds. The kinetics of solid-state reactions are primarily determined by the spatial and temporal sequence in which intermediate and final phases form, the characteristics of the phase boundaries, and the nucleation and growth behavior of the reaction products. A more detailed discussion of the reaction kinetics can be found in Reference [71]. The first step is to prepare the polycrystalline precursor compounds. Elemental powders are prepared in the desired stoichiometric ratios. As an example, $\text{Co}_{1/3}\text{TaS}_2$ was synthesized using elemental cobalt, tantalum, and sulfur in a stoichiometric ratio of (0.333:1.0:2.0), corresponding to a total precursor mass of 1 g. The powders were mixed and ground together with a pestle and mortar. Care must be taken to ensure that the pestle and mortar are thoroughly cleaned and free of any residual elemental contaminants prior to mixing the precursors. The pestle and mortar can be cleaned using a gentle acid

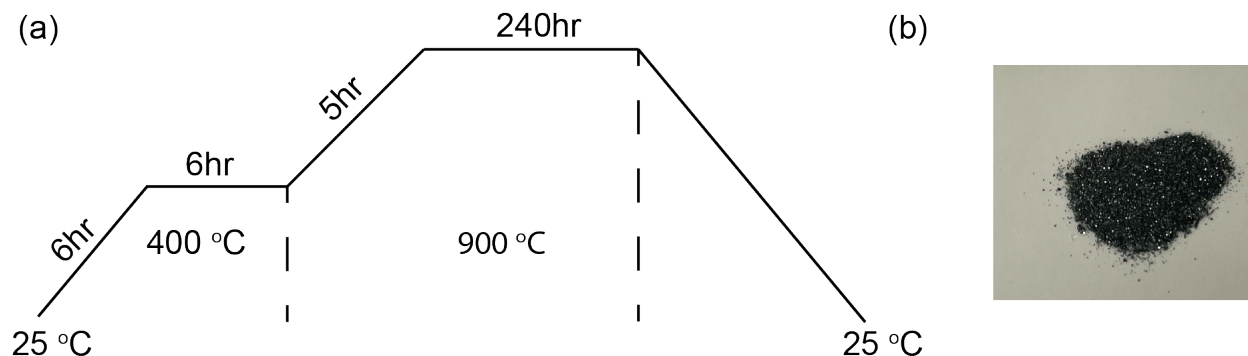


Figure 2.1: **Typical solid state reaction ramp rates schema (a)** Ramp rate and temperature profile for a typical solid state reaction. **(b)** Polycrystalline precursor powders obtained after solid state reaction.

bath. A small amount of deionized water is added to the mortar, followed by a few drops of nitric acid (HNO_3), and the mixture is left to sit for approximately three hours before thorough rinsing and drying.

The powder is then loaded into an alumina crucible and sealed in quartz tubes (the crucible sits on top of quartz wool) under partial pressure (200 Torr) of argon gas. The argon gas and quartz wool reduces the possibility of the quartz tube breaking during the growth process. Figure 2.1 (a) shows a typical solid state reaction growth process. The tube is heated to 400 °C and kept there for 6 hours and then raised to the final reaction temperature and held there for 240 hours (10 days). The intermediate step allows the sulfur to react with the other constituents before boiling. The furnace was then shut off and allowed to cool naturally. The reaction should yield a shiny free-flowing powder of poly-crystals as seen in Figure 2.1 (b). The presence of clumped or loosely flowing black powder in the final product suggests an incomplete reaction or insufficient diffusion during the solid-state synthesis stage. Inspect the resulting powder under a microscope to check for the presence of well-formed crystallites, which indicates a more complete solid-state reaction. If no well-formed crystallites are observed, the powder should be reground to break up any remaining clumps, resealed in a quartz ampoule, and subjected to a repeated solid-state reaction cycle. It is also advisable to verify the calibration and functionality of the furnace thermocouples and heating elements to ensure proper temperature control during synthesis.

Chemical Vapor Transport

Chemical vapor transport (CVT) refers to a class of reactions in which a condensed phase (a solid) is partially volatilized in the presence of a gaseous reactant (transport agent), and subsequently re-deposits at a different location in the form of single crystals. For example,

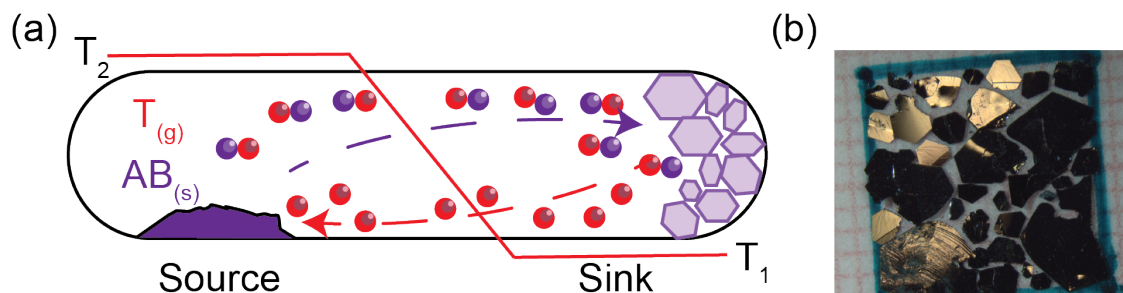


Figure 2.2: **Chemical vapor transport scheme** (a) Chemical vapor transport scheme for an endothermic reaction. The precursor powder is placed in the source end (purple) and reacts with the transport agent (red) to form gaseous products (purple-red spheres). The products diffuse to the sink end via the temperature gradient and deposit single crystals (purple). (b) Single crystals of various size obtained from CVT process.

for the CVT for a solid AB with a transport agent T is given by the following chemical reaction



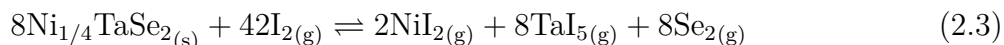
where s and g represent the solid and gas phases, respectively. The forward reaction transforms $A_{(s)}$ into the gas phase called dissolution. At another temperature, the back reaction will deposit $AT_{(g)}$ and $B_{(g)}$ from the gas phase into a solid, $AB_{(s)}$. The place of volatilization is called the source zone and the place of deposition is called the sink zone. The transport agent, typically a halogen and a temperature gradient is used to drive the chemical reaction. A horizontal two-zone furnace is used to maintain the temperature gradient. The source and deposition zones, defined by temperatures T_2 and T_1 , must be high enough for the transport agent and its reaction products with the precursor to remain in the gaseous state, enabling reversible vapor-phase transport. Figure 2.2 (a) illustrates a typical CVT process.

In the case of an endothermic reaction, the temperature at the source zone (T_2) is maintained higher than that at the deposition zone (T_1). The volatile reaction products formed at T_2 migrate through the temperature gradient toward the cooler end, where the reduced temperature favors the reverse reaction. As a result, the gaseous intermediates decompose, releasing the transport agent and depositing the desired compound in single-crystal form. The transport agent will then migrate back to the source zone and repeat the cycle again.

The selection of a suitable transport agent and temperature gradient will depend on the

type of reaction and target compound one is seeking to obtain. The transport agent must be able to volatilize and its reaction products with the precursor also volatilize at reasonable furnace temperatures. The CVT reaction must also be reservable at both the sink and source ends of the reaction vessel.

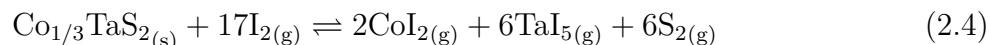
2.2 Nickel Intercalated Tantalum Diselenide



Recipe

High-quality single crystals of $\text{Ni}_{1/4}\text{TaSe}_2$ were grown by a two-step procedure. First, a precursor was prepared. The elements were combined in a ratio Ni:Ta:Se ($x : 1 : 2$), where $x = 0.25$ with a 5% Se excess. The powders were loaded into an alumina crucible and sealed in a quartz tub under a partial pressure of argon gas (200 Torr). The tube was heated to 900 °C and kept there for 5 days. The furnace was then shut off and allowed to cool naturally. This reaction yields a free-flowing shinny powder of crystallites. Second, 1 g of such precursor was loaded with 207 mg of iodine in a 21 cm tube and placed in a horizontal two-zone furnace. The precursor and iodine were placed in one zone (zone1), while the growth occurred at the other end of the tube (zone2). Both zones were first heated to 850 °C for 6 hours to encourage nucleation. Then, while zone 1 was maintained at 850 °C, the temperature in zone 2 was reduced to 750 °C. This condition was maintained for 5 days. The furnace was then shut off and allowed to cool naturally.

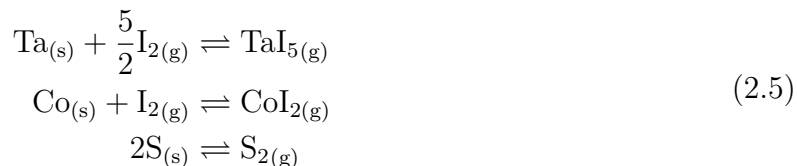
2.3 Cobalt Intercalated Tantalum Disulfide



The balanced chemical reaction for the chemical vapor transport of $\text{Co}_{1/3}\text{TaS}_2$ is given by Equation 2.4. At the source end, the chemical equilibrium is shifted to the right volatilizing the elements into the gas phase. The temperature gradient then drives the chemical equilibrium to the sink end, driving the reverse reaction. At the sink end $\text{Co}_{1/3}\text{TaS}_2$ deposits into single crystals. Equation 2.4 represents the dominant reaction, other iodine forming products such as lower iodides (TaI_4 and TaI_3) and cluster iodides such as Ta_6I_{12} could be obtained during the reaction, further complicating the chemical reaction [72].

For the primary reaction Equation 2.4, the volatile iodine reactions can be described by

the following set of reactions



each reaction has its corresponding Gibbs free energies ($\Delta G^\circ(T)$) at the source temperature of 950 °C. Typically, Equations 2.5 can be used to determine the dominant reaction that will drive the transport reaction by determining which volatilized species has the highest partial pressure. This can be determined by first calculating the equilibrium constant K_p for each reaction and then the partial pressure for $\text{TaI}_{5(g)}$, $\text{CoI}_{2(g)}$, and $\text{S}_{2(g)}$.

$$K_p = \exp\left(\frac{-\Delta G^\circ}{RT}\right)\tag{2.6}$$

The equilibrium constant can be calculated by Equation 2.6 with knowledge about the Gibbs free energy (ΔG°) at the reaction temperature. These can be obtained from online resources such as the JANAF Thermochemical Tables [73], NIST Chemistry WebBook.

Species	$\Delta G^\circ(1273 \text{ K})$	K_p
TaI ₅	−180 kJ mol ^{−1}	2.03×10^7
CoI ₂	−95 kJ mol ^{−1}	3.36×10^4
S ₂	220 kJ mol ^{−1}	9.4×10^{-10}

Table 2.1: Equilibrium constants for species in $\text{Co}_{1/3}\text{TaS}_2$ CVT reaction.

Now we can compute the partial pressures by defining the reaction extents, let ξ_1 and ξ_2 be the fraction of I_2 that forms TaI_5 and CoI_2 , respectively.

$$P_{\text{TaI}_5} = \xi_1 P_o\tag{2.7}$$

$$P_{\text{CoI}_2} = \xi_2 P_o\tag{2.8}$$

$$P_{\text{I}_2} = P_o(1 - 2.5\xi_1 - \xi_2)\tag{2.9}$$

Note that, according to Equation 2.5, each TaI_5 molecule consumes 2.5 mol of I_2 . The term P_o represents the initial partial pressure of I_2 before any reaction occurs. This value can be estimated by assuming an initial iodine mass of 100 mg, a reaction temperature of 1273 K, and an ampoule with a length of 25.4 cm and an inner diameter of 1.4 cm, corresponding to a total volume of 129.24 cm³. Using the ideal gas law, the initial iodine vapor pressure is calculated to be $P_o = 0.32$ mbar. The equilibrium constants for the reactions can then be expressed in terms of the partial pressures as follows:

$$K_{\text{TaI}_5} = \frac{P_{\text{TaI}_5}}{P_{\text{I}_2}^{2.5}} = \frac{\xi_1 P_o}{(P_o(1 - 2.5\xi_1 - \xi_2))^{2.5}}$$

$$K_{\text{CoI}_2} = \frac{P_{\text{CoI}_2}}{P_{\text{I}_2}} = \frac{\xi_2 P_o}{P_o(1 - 2.5\xi_1 - \xi_2)} \quad (2.10)$$

$$K_{\text{S}_2} = P_{\text{S}_2}$$

By simultaneously solving the two non-linear equilibrium equations for the reaction extents, we obtain $\xi_1 = 7.97 \times 10^{-4}$ and $\xi_2 = 0.99788$, indicating that nearly all of the I_2 reacts to form CoI_2 , while only a negligible fraction is converted to TaI_5 . The resulting total equilibrium pressure is $P_{\text{Total}} \approx 0.3223$ bar, which lies within the optimal transport window of $0.1 \text{ bar} < P_{\text{total}} < 0.5 \text{ bar}$. The corresponding equilibrium partial pressures of each gaseous species are summarized in Table 2.2.

Species	P_{Species} (bar)
CoI_2	0.32196
TaI_5	2.57×10^{-4}
I_2	4.07×10^{-5}
S_2	9.4×10^{-10}
T (K)	1273 K

Table 2.2: Calculated partial pressures with an initial amount of 100 mg of I_2 at 1273 K

From the calculated partial pressures, several trends can be identified. The partial pressure of CoI_2 is the dominant component in the vapor phase, indicating that its formation is strongly favored and that most of the available iodine reacts to form CoI_2 . In contrast, the formation of TaI_5 requires 2.5 moles of I_2 per mole of product, so only a small fraction of iodine is consumed to form TaI_5 . The partial pressure of S_2 is negligible under these conditions, suggesting that sulfur contributes minimally to the gas phase composition unless present in large excess; therefore, there is no risk of overpressure from sulfur.

These trends are consistent with the thermodynamic parameters summarized in Table 2.1, where the negative Gibbs free energies of formation (ΔG°) for TaI_5 and CoI_2 at 1273 K correspond to large equilibrium constants (K_p), confirming that halide formation is strongly favorable. Among these, CoI_2 exhibits the largest effective vapor-phase stability due to its high K_p and lower stoichiometric iodine requirement. Conversely, the positive ΔG° for S_2

formation and its correspondingly small K_p value explain its negligible equilibrium partial pressure. Overall, the thermodynamic equilibrium strongly favors the formation and transport of CoI_2 over TaI_5 , with sulfur playing only a minor role under the present reaction conditions.

These calculations serve as a first approximation for assessing the extent and feasibility of the transport reaction, and should be interpreted accordingly. For example, the very large equilibrium constant for CoI_2 (2.03×10^7) naturally raises the question: What if the equilibrium constant is sufficiently large that essentially all of the I is converted into CoI_2 ? A more physically accurate assessment would require an improved thermodynamic model for evaluating the equilibrium constants and recomputing the resulting partial pressures of all species involved. This is necessary because the full Gibbs free energy functions—and therefore the equilibrium constants—are strongly temperature dependent.

$$\Delta G(T) = \Delta H^\circ + T\Delta S^\circ + \int dT(\Delta C_p/T) \quad (2.11)$$

The following section outlines the procedure for determining the standard Gibbs free energy, ΔG° , of a given gaseous species, which is required for calculating equilibrium constants and partial pressures in chemical transport reactions.

1. Obtain polynomial coefficients:

$$c_p = a_0 + a_1T + a_2T^2 + a_3T^3 + a_4T^{-2} \quad (2.12)$$

and corresponding fitted values for $H^\circ(T)$ and $S^\circ(T)$.

2. Integrate from a reference (298.15 K) up to T

$$H^\circ(T) - H^\circ(298.15 \text{ K}) = \int_{298.15}^T dT' c_p^\circ(T') \quad (2.13)$$

$$S^\circ(T) - S^\circ(298.15 \text{ K}) = \int_{298.15}^T dT' [c_p^\circ(T')/T'] \quad (2.14)$$

3. Compute $\Delta G^\circ(T)$ at T :

$$\Delta G^\circ(T) = \Delta H_f^\circ(298.15) + [H^\circ(T) - H^\circ(298.15)] - TS^\circ(T) \quad (2.15)$$

4. For the reaction, compute:

$$\Delta G_{rxn}^\circ(T) = \sum_i \nu_i \Delta G_i^\circ(T) \quad (2.16)$$

where ν_i are the stoichiometric coefficients.

5. Finally compute equilibrium constant

$$K_p(T) = \exp\left(-\frac{\Delta G_{rxn}^\circ(T)}{RT}\right) \quad (2.17)$$

A temperature gradient must be chosen such that the transport rate is large enough to drive crystal growth rate at a reasonable rate and that the iodine is freed by decomposition at the cold end, consistent with its catalytic role. The transport rate can be modeled as a one-dimensional steady-state diffusion as rate-determining step. This model first suggested by Schaefer under the assumptions [74]:

- $\Delta T < T$
- No reaction takes place within the diffusion path.
- Partial pressure gradient is linear.
- Steady-state is established quickly.
- The mean free path-length of the molecules is small compared to the dimensions of the transport ampoule.

Consider a simple heterogeneous reaction where solid A reacts with gas B to form a gaseous species C it can be described by the following chemical reaction,



For a simple heterogeneous reaction, the transport rate $\dot{n}$ (mol/sec), can be described by stoichiometric flow and diffusion given by

$$\dot{n}_A = \frac{n_A}{t} = \frac{a}{b} \frac{n_B}{t} = -\frac{a}{b} Dq \frac{dC_B}{ds} - \frac{a}{b} q C_B W \quad (2.19)$$

$$\dot{n}_A = \frac{n_A}{t} = \frac{a}{c} \frac{n_C}{t} = \frac{a}{c} Dq \frac{dC_C}{ds} + \frac{a}{c} q C_C W \quad (2.20)$$

where a , b and c are the stoichiometric coefficients, C_i are the concentrations, D is the diffusion coefficient, q the cross section of the diffusion path, s the length of the diffusion path, t is time and W is the flow of the gas phase as a whole. Solving for W and noting that $\frac{dC_B}{ds} = -\frac{dC_C}{ds}$ we can recast them in terms of the concentrations of the species,

$$\dot{n}_A = -\frac{a}{c}Dq \frac{dC_B}{ds} \left[\frac{b(C_B + C_C)}{cC_B + bC_C} \right] \quad (2.21)$$

$$\dot{n}_A = -\frac{a}{b}Dq \frac{dC_C}{ds} \left[\frac{c(C_B + C_C)}{cC_B + bC_C} \right] \quad (2.22)$$

If we assume the conditions for steady-state and for a small ΔT , the concentrations can be replaced by pressure via the ideal gas law ($PV = nRT$, $P = CRT$). The temperature can be replaced by the average temperature along the diffusion path (which can be approximated as the average temperature between the source and sink $\bar{T} = \frac{T_1+T_2}{2}$). The derivative of the partial pressure with respect to the position along the diffusion path can be replaced by the difference of partial pressures of the gas species per length of the diffusion path (i.e $\frac{dP}{ds} \rightarrow \frac{\Delta P}{s}$). $P_i \rightarrow \bar{P}_i = \frac{P_i(T_1)+P_i(T_2)}{2}$ to give

$$\dot{n}_A = \frac{a}{c} \frac{Dq}{RT} \frac{\Delta P_C}{s} \left[\frac{c(\bar{P}_B + \bar{P}_C)}{c\bar{P}_B + b\bar{P}_C} \right] \quad (2.23)$$

The term in the parenthesis is called the individual flow factor, F , which is determined by the stoichiometric coefficients and the other terms are determined by the experimental conditions. Consider the second equation in Equation 2.5, for this reaction, the flow factor $F = 1$ and the transport rate for CoI_2 becomes

$$\dot{n}_{\text{CoI}_2} = Dq \frac{\Delta P_{\text{CoI}_2}}{RT_s} \quad (2.24)$$

As a first approximation, the transport performance of each species can be evaluated from the difference in its partial pressure between the two temperatures, generally a larger ΔP drives the crystal growth.

Recipe

High quality single crystals of $\text{Co}_{1/3}\text{TaS}_2$ were grown by a two-step procedure. First, a precursor was prepared. The elements were combined in a ratio Co:Ta:S (0.3333:1.0:2.0). The powders were loaded in alumina crucibles and sealed in quartz tubes under a partial pressure (200 Torr) of argon gas. The tube was heated to 900 °C and kept there for 10 days (Figure 2.1 (a)). The furnace was then shut off and allowed to cool naturally. This reaction yields a free-flowing shiny powder of crystallites (Figure 2.1 (b)) that was ground with a mortar and pestle separately for each batch.

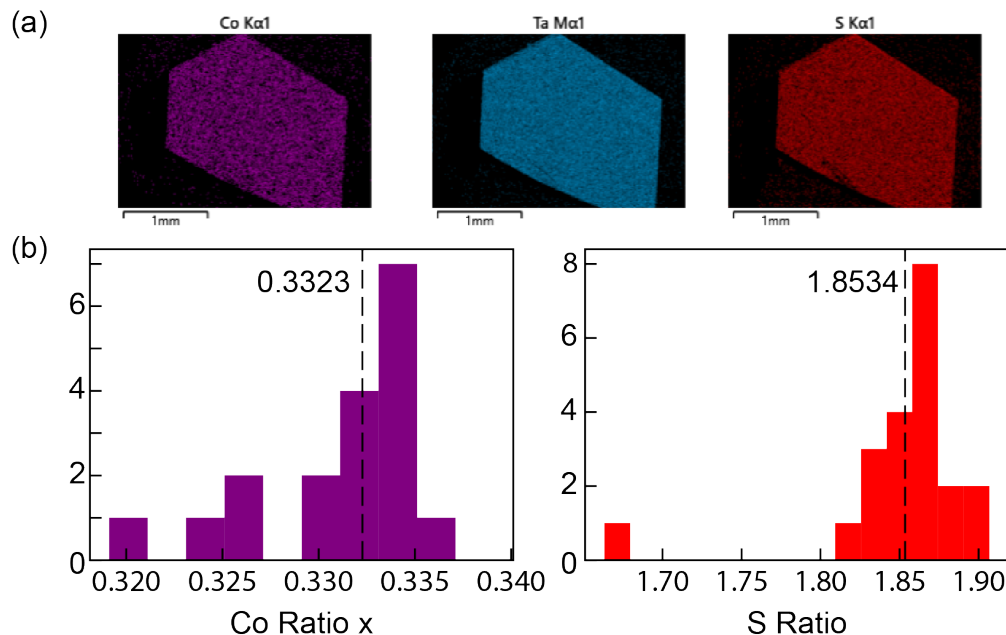


Figure 2.3: **Electron dispersive x-ray map and histogram** (a) EDX mapping analysis of a $\text{Co}_{1/3}\text{TaS}_2$ single crystal acquired using an Oxford Instruments EDX detector integrated into a ThermoFisher SCIOS-2 dual-beam SEM. An accelerating voltage of 30 kV and a beam current of 3.2 nA were used for data collection. (b) Histograms obtained from EDX measurements for the Co/Ta and S/Ta ratios, using bin sizes of 0.002 and 0.0121, respectively. The standard deviations determined from ten measurement points are 0.0051 for Co/Ta and 0.0484 for S/Ta. The dashed line in each graph indicates the average value of the corresponding ratio.

Second, 1 g of precursor (powder of crystallites) was loaded with 10 mg cm^{-3} of iodine in a 10" (21 cm for the "King" Mellen two-zone furnace) long quartz tube, evacuated, and placed in a horizontal two-zone furnace. The precursor and iodine were placed in the source zone (hot side) and the other end of the tube is the sink zone (cold side). Both zones were heated to 850°C for 6 hours to encourage nucleation. Then, the source zone was raised to 950°C while the sink zone was kept at 850°C . This condition was maintained for 10 days. The furnace was then shut off and allowed to cool naturally. Shiny hexagonal crystals up to several mm in lateral length were collected from the sink zone.

Energy-dispersive X-ray spectroscopy (EDX) measurements were performed to determine the cobalt intercalation ratio x (Figure 2.3). Spectra were acquired at ten distinct locations within a $1 \text{ mm} \times 1 \text{ mm}$ area on a single crystal to assess compositional uniformity. In addition, EDX spatial mapping was conducted to evaluate the elemental homogeneity across the

crystal surface. Figure 2.3 (a) shows uniform spatial distributions of all three constituent elements—Co, Ta, and S—indicating good lateral compositional homogeneity. The histogram of the extracted cobalt intercalation ratio x , along with the average value obtained from all measured points, is shown in Figure 2.3 (b). The calculated mean intercalation ratio is $x = 0.3323$ with a standard deviation of 0.002. For sulfur, the average stoichiometric ratio is 1.8634 with a standard deviation of 0.0121, suggesting that the crystal surface may be slightly sulfur deficient. A more accurate determination of the bulk stoichiometry can be obtained using inductively coupled plasma optical emission spectroscopy (ICP-OES).

Set Points	1	2	3	4	5	6	7	8	9	10
Hot (°C)	300	300	600	600	850	850	950	950	950	950
Cold (°C)	300	300	600	600	850	850	850	850	850	850
Time (min)	60	10	60	10	60	360	120	9999	4401	-121
Total Time (min)	15080									
Total Time (Days)	10.47									

Table 2.3: Typical program for a CVT reaction with a target temperature gradient of 950 °C-850 °C to be used in a MTI-Two Zone furnace. Note the last time value of -121 at set point **10** instructs the furnace to switch off power to the heating elements, thereby initiating a natural cool down.

Set Points	1	2
Hot (°C)	0	950
Cold (°C)	0	850
Time (hour)	0	240
Total Time (hour)	240	
Total Time (Days)	10	

Table 2.4: Schematic outline of typical program for a CVT reaction with a target temperature gradient of 950 °C-850 °C to be used in the “King” Mellen Two Zone furnace. A detailed standard operating procedure of the “King” Mellen is given in appendix 1.

A typical temperature program profile for a CVT reaction in an MTI two-zone furnace is given in Table 2.3. The first four setpoints in program are implemented to allow a slow ramp up to the soaking temperature. This will help to extend the life time of the heating elements of the furnace. It also advised to alternate the hot and cold side between growths to also

extend the life time of the heating elements and thermocouples. It is advised to periodically check the resistance across the heating elements with a multimeter. The MTI two-zone has four independent heating elements the resistances should be similar and on the order of $10\ \Omega$. See Figure 2.4 (a) for the measurement access points. The thermocouples should also be periodically inspected for any potential damaging. The resistance of each thermocouple should also be measured periodically and should be on the order of $0.1\ \Omega$. Figure 2.4 (b) shows the measurement access points.



Figure 2.4: **MTI two-zone furnace** Left: Interior of the tube furnace, showing the measurement points used to verify heating elements' resistance with a multimeter (highlighted in red). Right: Access panel to the thermocouples; the regions highlighted in red indicate the locations where resistance measurements are performed using a multimeter.

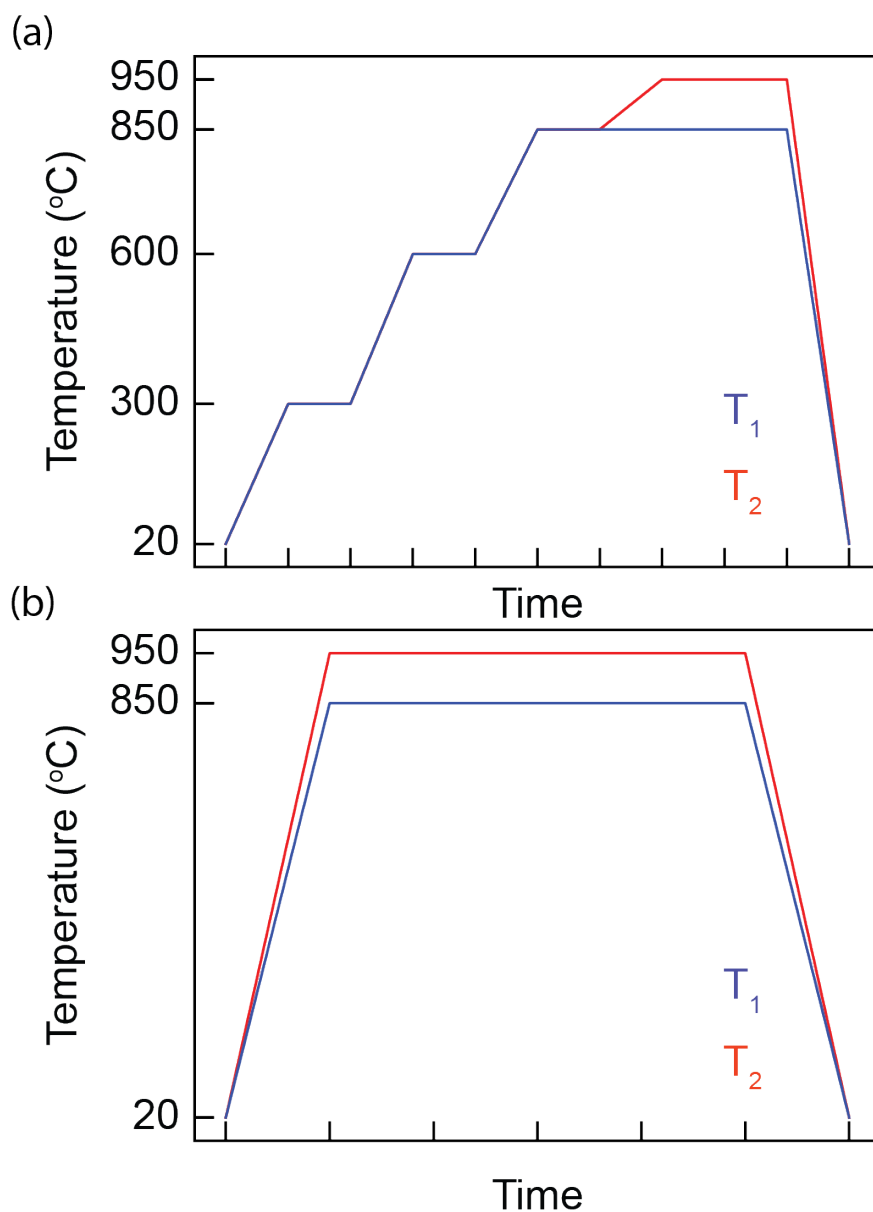


Figure 2.5: **Two-zone furnace temperature ramp schemes** Temperature set point values (K) versus time (arbitrary units). The hot side and cold side is represented by red and blue curve respectively. The time axis has been scaled uniformly to show clarity. **(a)** Temperature ramp scheme for CVT reaction in a MTI horizontal two-zone furnace corresponding to the CVT program outlined in Table 2.3 **(b)** Temperature ramp scheme for CVT reaction in a Mellen Company TS2 Tube 1550 horizontal two-zone furnace corresponding to the CVT program outlined in Table 2.4

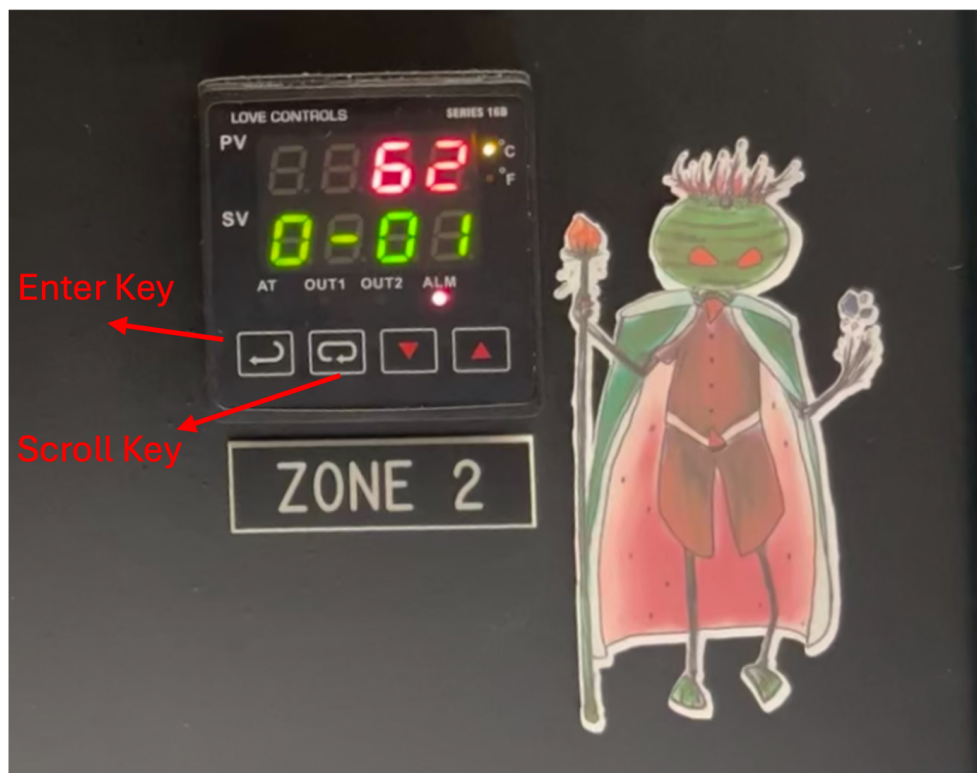


Figure 2.6: **“King” Mellen front control panel** LOVE temperature controller installed on the Mellen two-zone furnace. The Enter and Scroll keys are highlighted in red.

2.4 Appendix I: Two Zone Furnace Operation “King Mellen”

The “King” Mellen furnace is powered by an external power supply and is equipped with a programmable LOVE temperature controller (Model 16B-53). The controller supports four modes of operation: proportional–integral–derivative (PID), on–off, manual, and ramp/soak control. The procedure described below outlines the standard method used in the Analytis Lab to program a custom ramp/soak temperature profile for chemical vapor transport (CVT) growth. For alternative operating modes or advanced programming features, the reader is referred to the manufacturer’s documentation: LOVE Series 4B, 8B, 16B, and 32B Microprocessor-Based Temperature Process Control: Specifications, Installation, and Operating Instructions.

Programming the Controller

Basic guide on how to make or change a program on the Love controller, power on the system and follow the steps listed below.

1. Press and hold the enter key for more than three (3) seconds to enter the Initial Setting mode.
2. Click the scroll key until Ctrl_/PID is shown on the screen (four (4) times).
3. Change the control method to ProG by pressing the up or down arrows. Each option name should be flashing prior to entering that mode.
4. Press the enter key. Now, the ProG name should no longer be flashing.
5. Press the index to key to see the parameter PAtn.
 - a) This is the pattern you can program, numbered 0-7
6. There are eight (8) segments per pattern, numbered 0-7. Each step has values SP00 (through SP07), followed by the time of that step in the pattern ti00 (through ti07).
 - a) The values for SP are the target setpoints of the ramp or soak.
 - b) The values for Ti are the time for each segment in hours.minutes (hh.mm).
 - c) IMPORTANT: the first segment must be a soak. You can soak for a small amount of time (1 minute) at room temperature and have the ramp start at the next segment.
7. After the first required soak, program the segments as desired, with ramps or soaks. Note, the controller will determine the ramp rates based on the temperature and time inputs.
8. Press the enter key to return to the operation mode.
9. To start the program, enter the operation mode by pressing the enter key, then press the scroll key until r-S is displayed
 - a) Using the arrow key, select rUn. The program will now begin.
 - b) To stop the program, select StoP.

Programming the Controller

The following guide provides an example of how to program the LOVE 16B-53 temperature controller. The procedure demonstrates the implementation of a CVT ramp/soak profile corresponding to the temperature program shown in Figure 2.5 (b). The program consists of a temperature gradient of 950 °C–850 °C between the hot and cold zones, respectively, with a total soak time of 10 days.

Creating the Program

The “King” MELLEN furnace is a split two-zone system and therefore requires independent programming of two separate temperature controllers to define and ramp the hot and cold zone temperatures. The programming procedure is identical for both controllers. Throughout the following discussion, the controller associated with the hot zone is denoted in red, while the controller associated with the cold zone is denoted in blue for clarity.

1. Power on the system
2. Press and hold the enter key for more than three seconds to enter the Initial Setting mode.
3. This should display the thermocouple (input) type in green.
 - a) MELLEN recommends confirming your configured thermocouple type matches the actual thermocouple that is being used. (Currently, the furnace is operating with an r-type thermocouple which has a temperature range 0 °C-1700 °C)
 - b) Failure to match the thermocouple types could lead to catastrophic system failure.
4. Press the scroll key until Ctrl is displayed on the screen.
 - a) To set the program, press up or down arrow until ProG is displayed in green.
5. Press the enter key again to save this setting. The screen flashes until the setting is saved.
6. Press the scroll key until PAtn is displayed on the screen.
 - a) PAtn is the pattern, or the program. There are eight programs available in this controller (PAtn 0-8).
7. Select the pattern number 0 using the up or down arrows, then press the scroll key to start the programming.
8. SP00 is the setpoint temperature of the first segment (segment 00)

- a) This will constitute the mandatory initial soak which will be set to 0°C (0°C).
 - b) Use the up and down arrows to select 0, press the enter key to save this temperature setpoint.
9. Press the scroll key to set the time for the first segment, ti00.
 - a) The time display is hh.mm (hours.minutes).
 - b) For the initial soak, the time is set to 00.01.
 - c) Press the enter key to save this time setting.
10. Press the scroll key to set the next segment SP01
 - a) Set this value to 950 (850) to make the second segment ramp up to 950°C (850°C).
 - b) Press the enter key to save this temperature setpoint.
11. Press the scroll key to view the time for the second segment, ti01.
 - a) Use the arrow keys to set this to 6.00 (6 hours).
 - b) 950°C (850°C) over 6 hours gives a ramp rate of $\sim 2.638^{\circ}\text{C min}^{-1}$ ($\sim 2.361^{\circ}\text{C min}^{-1}$)
 - c) Press the enter to key to save this time setting.
12. Press the scroll key to program the next segment, SP02.
 - a) Set this value to 950 (850) to make the second segment 950°C (850°C).
 - b) Press the enter to key to save this setting.
13. Press the scroll key to and set ti02 to 15.00 to soak for 15 hours.
 - a) Use the up and down arrows to set 15.00.
 - b) Press enter to save this time setting.
14. Press the scroll key to program SP03.
 - a) Set this value to 0.
 - b) Press the enter key to save this setting.
15. Press the scroll key to set ti03 to 0.
 - a) Set this value to 00.00.
 - b) Press the enter key to save this setting.
16. For setpoints P04-SP07 and ti04-ti07 set them to 0 and 00.00, respectively and save those settings.

17. Press the scroll key to set PSY0 parameter.
 - a) This parameter determines the last step for pattern 0 that will be performed in pattern 0. Since the temperature values for ti04-ti07 were set to 0, the controller will skip these steps.
 - b) Use the up and down arrows to set PSY0 to 7.
 - c) Press the enter key to save this setting.
18. Press the scroll key to set CYC0 parameter.
 - a) This parameter sets the number of times that the current pattern will be repeated.
 - b) Use the up and down arrows to set CYC0 to 0.
 - c) Press enter to save this setting.
19. Press the scroll key to set LIN0 parameter.
 - a) This parameter sets the next pattern that will be performed after the current pattern. When set to off, the program will end and maintain last set point.
 - b) Use the up and down arrows to set LIN0 to 1.
 - c) Press the enter key to save this setting.
20. Press the scroll key to program the next PAtn
 - a) Use the up and down arrows to set PAtn to 1.
 - b) Press enter to save this setting.
21. Press the scroll key to set SP10.
 - a) Set this value to 950 (850).
 - b) Press enter to save this setting.
22. Press scroll key to set ti10 to 15.
 - a) Set this value to 15.00
 - b) Press enter to save this this setting.
23. For setpoints SP11-SP17 and ti11-ti17 set them to 0 and 00.00, respectively and save those settings.
24. Press the scroll key to set PSY1 parameter.
 - a) Use the up and down arrows to set PSY1 to 7.
 - b) Press the enter key to save this setting.

25. Press the scroll key to set CYC1 parameter.
 - a) Use the up and down arrows to set CYC0 to 14.
 - b) Press enter to save this setting.
26. Press the scroll key to set LIN1 parameter.
 - a) Use the up and down arrows to set LIN0 to oFF.
 - b) Press the enter key to save this setting.
27. Press the enter key once to enter the Operation Menu.
28. Press the scroll key once in the Operation Menu to access the run settings r-S. If the green keys read rUn, the program will run.
 - a) Press the up and down arrows to set the program to rUn, or StoP.
 - i. rUn begins the user defined patterns
 - ii. StoP deactivates the outputs and stops the user defined pattern.
 - b) Set the value for s-R to rUn
 - c) Press enter. The program should now start.

Chapter 3

Device Fabrication: Focused Ion Beam

*“Shall we indeed accept good from God, and shall we not accept adversity?
In all this Job did not sin with his lips.”*

— Job 2:10

3.1 Sample Preparation

Exfoliating of crystals

If one is blessed with having to work van der Waals materials that can be exfoliated, the preparation process is relatively straightforward. Intercalated transition metal dichalcogenides (TMDs) can be most effectively exfoliated using low-residue tape, such as Nitto’s PCV film coated with an acrylic-based adhesive (“blue tape”). To exfoliate, place the crystal of interest onto the adhesive side of the tape, fold the tape over onto itself, and then peel it apart repeatedly until a good distribution of flakes is obtained on the tape. The thickness of the resulting flakes is a critical factor, as it determines the duration of focused ion beam (FIB) processing: the thicker the crystal, the longer the milling time. A practical rule of thumb is as follows: if the flake is thin enough (approximately 2 μm or less), focusing the microscope on the tape will also bring the crystal edge into focus. If the crystal edge goes out of focus, the flake is too thick. Figures 3.1(a)–(c) illustrate examples of flake thickness, shown in decreasing order. Figure 3.1(c) represents the optimal thickness to ensure reasonable FIB operation times. If, however, one is not endowed with such grace one must endure the path of lamella structuring, which is outlined below.

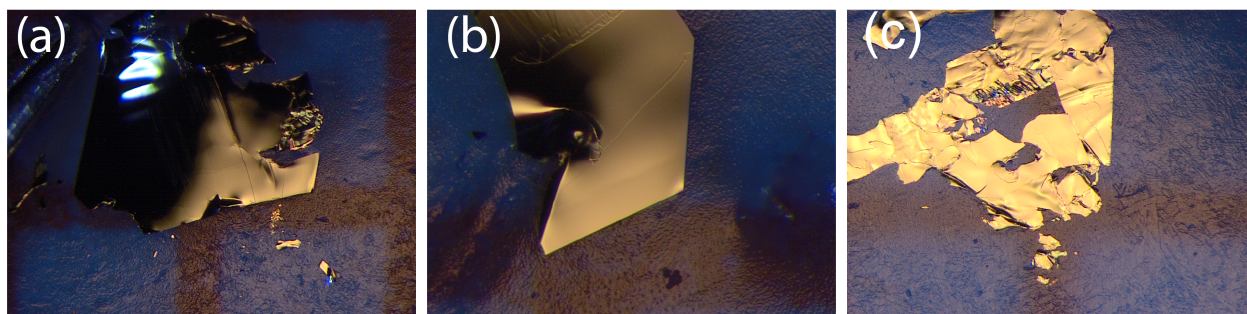


Figure 3.1: **Microscope images of exfoliated crystals with decreasing thickness** (a) The depth of the crystal is evident at highest magnification of the microscope. This flake is too thick for FIB. (b) Much thinner and viable for FIB. (c) Optimal thickness for FIB.

Mounting crystals

Once viable flakes have been identified, they must be transferred to a suitable substrate. For intercalated TMDs, there are two possible transfer techniques. The first method employs a splintered wooden stick (Figure 3.2). The splinter is gently rolled across the flake so that it lifts from the tape. The splinter is then statically charged by rubbing it against a latex glove, allowing the flake to adhere to the splinter. The flake can be mounted by pressing it down onto the substrate, and the splinter may also be used to ensure that the flake lies flush with the substrate surface.

The second method involves transferring a viable flake directly from the tape to the substrate. The crystal-bearing side of the tape is pressed onto the substrate surface, and gentle pressure is applied with tweezers. When the tape is peeled away, flakes remain attached to the substrate. These methods rely on van der Waals forces, which allow the flakes to adhere; they are therefore only effective for van der Waals crystals. For other materials, flakes can be mounted using an adhesive such as TorrSeal. A small amount of TorrSeal is deposited on the substrate, and the crystal flake is positioned on top of the adhesive with a wooden splinter, ensuring that the crystal is flush with the surface but not submerged. The TorrSeal is then cured under the appropriate conditions. Further details on this technique can be found in reference [1].

Once viable flakes are mounted on preferred substrates, gold deposition can be done. First, a shadow mask made of Kapton tape is applied to cover the edges of the substrate, leaving a window of about 1 mm exposed. This exposed region is then sputter-coated with a thin gold layer of approximately 100 nm.

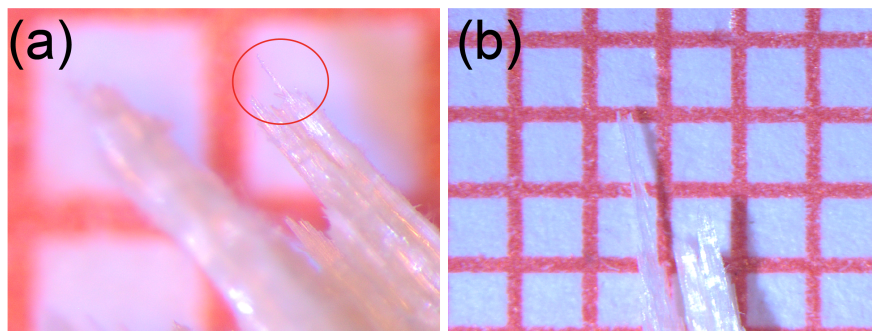


Figure 3.2: **Broken stick tip used to pick up small flakes via static electricity.** (a) Red circle highlights the optimal splinter for picking up small crystals. (b) Slightly larger size splinter.

3.2 Microstructuring of crystals

Patterning Devices

All focused ion beam (FIB) work presented in this thesis was carried out using a Thermo Scientific Scios-2 FIB–SEM microscope. This system is equipped with a Ga^+ ion beam and a gas injection system (GIS) for in situ platinum deposition.

The GIS needle is carefully positioned in close proximity to the surface of the region of interest; care must be taken to avoid accidental contact between the needle and the sample. Prior to inserting the needle, the sample must be aligned at the eucentric height, the electron and ion beams set to the coincidence point, and the stage verified to be free of obstructions. Once correctly positioned, the needle releases a platinum precursor gas that adsorbs onto the sample surface. A commonly used precursor is methylcyclopentadienyl-trimethylplatinum ($((\text{CH}_3)_3(\text{CH}_3\text{C}_5\text{H}_4)\text{Pt})$). A selected region of the adsorbed gas is then irradiated by the Ga^+ ion beam. The energetic ions dissociate the organometallic precursor: the volatile organic fragments are pumped away, while platinum atoms remain bonded to the surface.

The deposited metal is not pure platinum. Tao et al. reported that Ga–FIB platinum depositions typically contain approximately 46% Pt, 24% C, 28% Ga, and 2% O [75]. Because of this composition, the two-point contact resistance would increase significantly if platinum alone were used as the contact material. To mitigate this issue, a localized platinum contact is deposited to bridge the gold layer on the substrate with the gold layer on the flake surface.

This is accomplished through a two-step deposition process. In the first step, platinum is deposited from the base of the substrate until the contact reaches the level of the flake. In the second step, an additional deposition is performed to bridge the gap and establish electrical

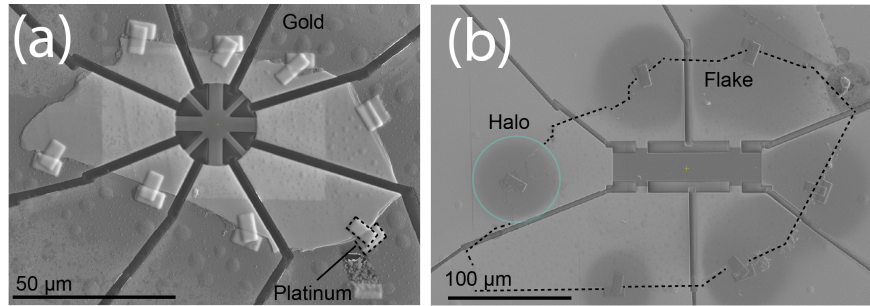


Figure 3.3: **SEM micrograph of post-FIB devices.** (a) Dashed box highlights the platinum welds used to ensure electrical connect the gold on the flake to the gold on the substrate. (b) In circle highlights the thin halo of deposited material from the GIS deposition.

continuity. Figure 3.3 (a) is typical FIB device. Platinum contacts can be seen in the dashed box. There is a non-zero probability that Ga^+ ions collided with a gas molecule above the surface and will react with the surface. This will result in a thin halo of deposited material. These halos are mostly carbon but given their chemically active nature it is possible that they may create conductive channels [76]. Therefore, it is imperative that the platinum welds be completed before any milling is performed. In this way, it is ensured that any conductive overspray is severed during the milling process. See Figure 3.3 (b). A dose test is first performed to determine the optimal ion beam current for metal deposition. Typically, a test contact is deposited away from the flake using an initial current of 1 nA and an accelerating voltage of 30 kV. If milling of the substrate is observed after only a few minutes (approximately 2 min), the current is too high and should be decreased. Conversely, if no deposition is observed after several minutes (2 min), the current is too low and should be increased.

After all of the platinum contacts have been deposited, the gold in the intended active region must be milled away. At this stage, the milling process must be monitored closely; if one is not vigilant, the ion beam can mill through the entire flake, rendering it irreparable. Typically, an ion beam current of 1.6 nA and an accelerating voltage of 30 kV is used for this step. The main device region can be patterned using a higher ion beam current (typically 3 nA). In Figure 6.1 (a)–(b), the device region consists of triangular and rectangular geometries, respectively. To ensure proper contact definition, cuts are extended slightly beyond the edges of the flake. At this stage, the ion beam current may be further increased to 5 nA to mill completely through the crystal. Finally, using the lowest magnification and an ion beam current of 5 nA, long cuts are made out to the edge of the gold layer to define the electrical contacts. These contacts will later be connected to silver wires with either a two part silver paint adhesive (EPO-TEK H20E) or silver compositions (DuPont 4929N). A typical FIB microstructuring process flow is shown in Figure 3.4 (a)–(d).

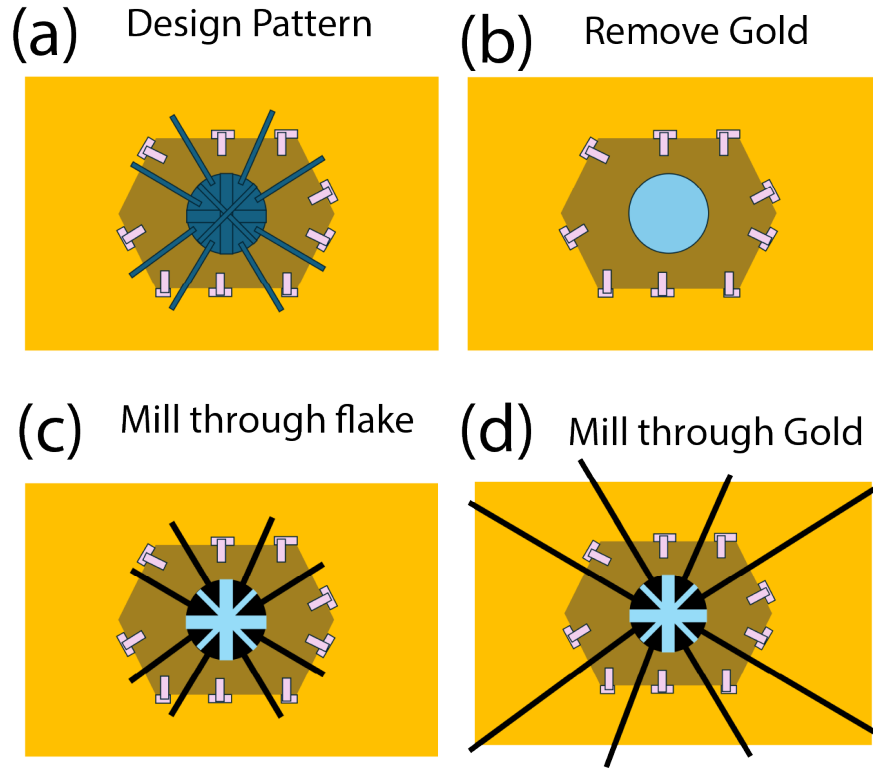


Figure 3.4: **Typical process flow for FIB microstructuring of devices.** (a) First step is to design the FIB pattern to be milled (in blue) according to user's dimensions. (b) Next is to remove the gold from the active region where current will be flowing (exposed flake surface in cyan). (c) The patterned designed in (a) is removed. (d) Remove gold to create separate current and voltage contacts for the device.

Lamella Structuring

A thorough explanation of all possible details of lamella structuring can be found in references [76, 77]. Here we will briefly outline the general process of lamella microstructuring. The bulk crystal of interest is first mounted onto an SEM stub using silver adhesive. The adhesive both secures the crystal mechanically to the stub surface and provides an electrical pathway to the SEM ground. This ensures that the crystal is well bonded to the stage of the SEM and prevents charge buildup during imaging and processing.

First, a protective layer of platinum is deposited on the surface with a width of $+2\mu\text{m}$ of the target thickness of the resulting lamella, a length of $100\mu\text{m}$ and a thickness of $1\mu\text{m}$. This metal layer will protect the cross-sectional area of the lamella during the resulting steps. Trenches are milled using two rectangular cross-section (RCS) patterns placed on either side

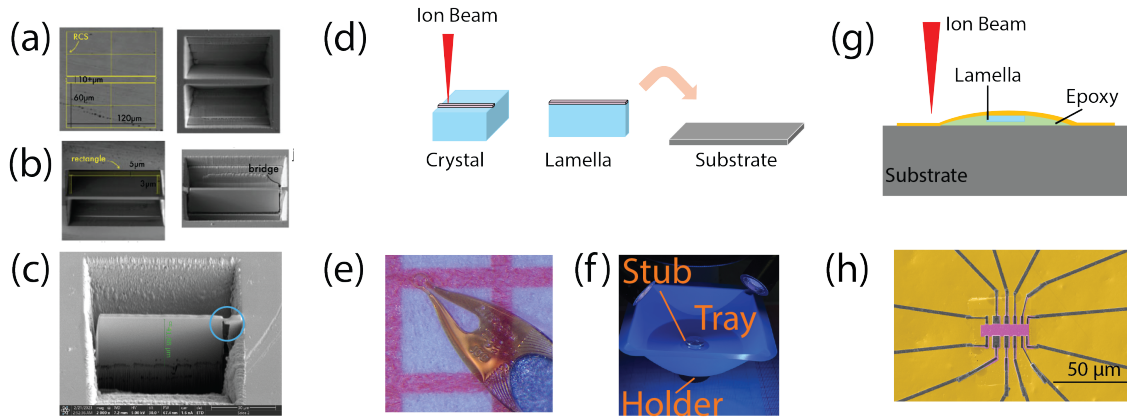


Figure 3.5: **Lamella microstructuring process flow.** (a) Right panel shows RCS used for trench milling. Left pane shows a completed trench. (b) Panels shows the the undercut done to separate out the lamella from crystal. (c) SEM image of cross section of a lamella cut out. (bridge in cyan) (d) General lamella process flow. (e) Kapton loop with 100 μm sample aperture (f) Lamella extraction setup under a microscope. (g) FIB device processing of a lamella cut out. (h) False colored SEM image of lamella transport device. Panels (a) and (b) adapted from Reference [77].

of the platinum deposition, Figure 3.5 (a). Once the desired depth is reached, an undercut is made to sever the lamella from the bulk crystal, Figure 3.5 (b). Three rectangular patterns cuts are placed on the face of the lamella and a bridge is left on one side of the lamella for stability, see Figure 3.5 (c). Thus, the lamella machining process is complete.

Once the lamella has been excavated, it must be transferred to a substrate. To minimize the risk of loss, a small weigh tray is positioned between the SEM stub and the stub holder to catch the lamella in the event that it is launched during extraction (which is highly likely). A Kapton tip loop (M2-L18SP-10) is affixed to the end of a wooden stick and used as the extractor. Owing to electrostatic attraction, the Kapton tip is expected to lift the lamella when it is brought into contact with the sample. The tip is pressed gently against the bridge until the lamella detaches. Before detachment, a small droplet of TorrSeal is prepared on the desired substrate. Using a single-hair brush, the TorrSeal is spread at the center of the substrate to form a thin adhesive layer. The lamella is then placed on top of the TorrSeal, ensuring that it rests flush on the surface without becoming submerged in the epoxy. The TorrSeal is cured for 30 min at 60 °C. Once cured, the FIB patterning steps described above can be repeated to fabricate the desired lamella transport device.

Chapter 4

Nickel Intercalated Tantalum Diselenide

“Einstein and Feynman ushered me into grad school, reality ushered me out.”

— Dr. Bob Henderson

Materials in this chapter were previously published in

- Josue Rodriguez et al., *Electrically Controlled Heat Assisted Magnetic Recording in Intercalated 2D Magnets*, arXiv:2605.06645 (2026).

4.1 Introduction

Here, we demonstrate an electronic variant of HAMR, in which magnetization reversal is assisted not by laser heating but by Joule heating from current pulses. Combined with electronic readout via the anomalous Hall effect, our approach enables an all-electronic read-write scheme for magnetization recording. We combine electronic transport measurements and magneto-optical imaging and provide unambiguous evidence that Joule heating transiently drives the magnetic medium above its Curie temperature, allowing magnetization reversal under a writing field of only $B_w \sim \pm 2$ mT, nearly 100 times smaller than the coercive field. The magnetic medium we employ in this study is Ni-intercalated TaSe₂, a two-dimensional (2D) itinerant metallic ferromagnet with Ni_{1/4}TaSe₂ composition. The magnetization direction in Ni_{1/4}TaSe₂ points along the out-of-plane direction (c-axis). This effective perpendicular magnetic anisotropy (PMA) is highly desirable for high-density magnetic recording.

The use of intercalated 2D magnets presents an attractive pathway for data storage technologies, offering a vast design space through diverse combinations of layered hosts and

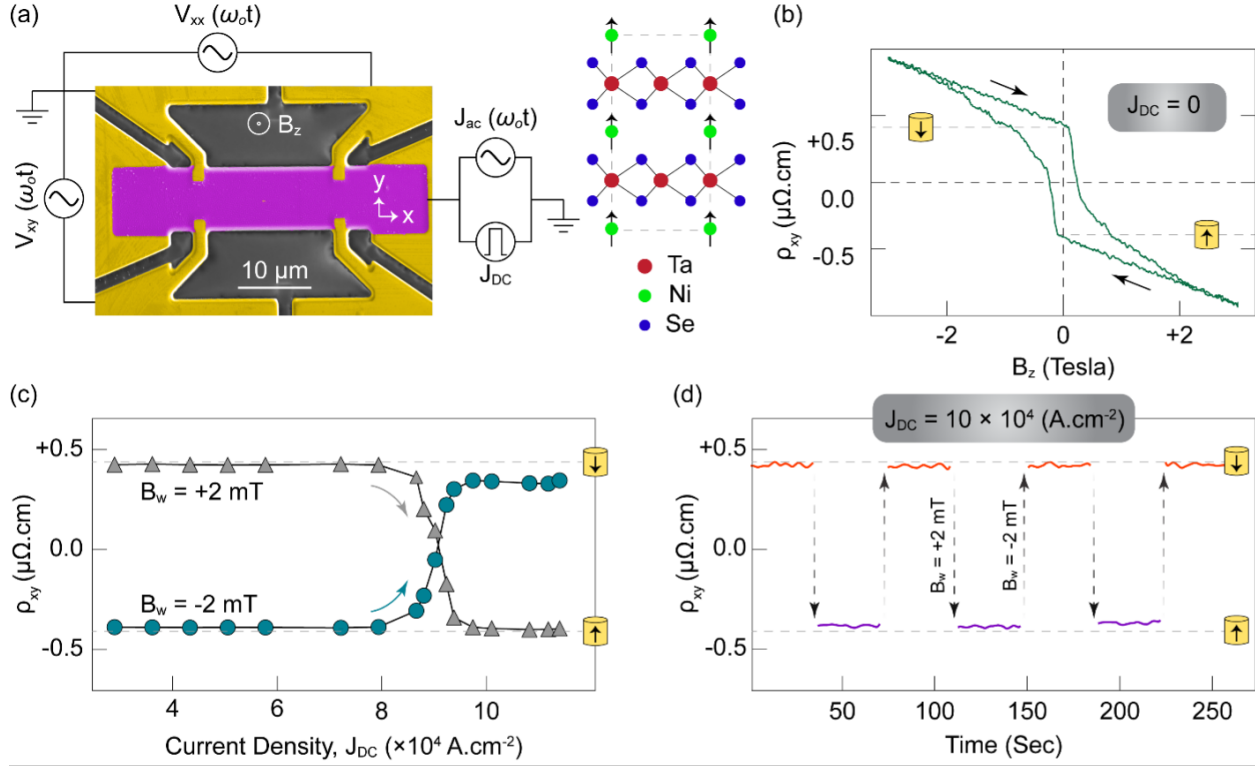


Figure 4.1: **Current-induced magnetization reversal in a $\text{Ni}_{1/4}\text{TaSe}_2$ FIB device.** (a) False colored SEM micrograph of a $\text{Ni}_{1/4}\text{TaSe}_2$ crystal patterned into a Hall bar. The magnetic field is applied along the out-of-plane direction (i.e., z-direction), which is the magnetic easy axis. Two current sources are used, a small ac component for electronic readout, and pulsed DC current for magnetization reversal. (b) The anomalous Hall resistivity, showing distinct low- and high-resistance states, corresponding to the UP and DOWN magnetization states, respectively. (c) Anomalous Hall resistivity measured after applying current pulses of various amplitudes. The device was initially set to a low (high) resistance state with a 3 T (−3 T) external field. A complete magnetization reversal is achieved at current densities above $1 \times 10^5 \text{ A cm}^{-2}$. A writing field of $B_w = \pm 2 \text{ mT}$ was applied while pulsing. (d) Magnetization can be repeatedly reversed by only changing the sign of B_w under a fixed pulse current density of $J = 1 \times 10^5 \text{ A cm}^{-2} > J_{th}$. Current pulses are 1 m sec long. Measurements are done at 2 K temperature.

magnetic intercalants. Such materials not only expand the portfolio of candidate materials for next-generation high-density magnetic memories, but also open opportunities to explore novel strongly correlated phenomena at the intersection of magnetism and low-dimensional physics.

4.2 Methods

We synthesized 2D $\text{Ni}_{1/4}\text{TaSe}_2$ crystals using a standard chemical vapor transport method, as described in Chapter 2. As schematically illustrated in Figure 4.1 (a), the material consists of layered basal planes of 2H-TaSe₂, with nickel intercalants occupying the van der Waals gaps and aligning vertically with the tantalum atomic columns. As we have previously reported, $\text{Ni}_{1/4}\text{TaSe}_2$ is an itinerant metallic ferromagnet with a Curie temperature of 55 K, exhibiting a uniaxial magnetic anisotropy along the crystallographic c-axis, effectively mimicking PMA [78]. For electronic transport measurements, high-quality crystals are patterned into Hall-bar devices using focused-ion beam (FIB) milling. A representative false-color scanning electron microscope image of a device is shown in Figure 4.1 (a), where the yellow regions correspond to gold contacts and the purple highlights the longitudinal transport channel. As schematically shown, the magnetization state of the device is probed by measuring the transverse resistivity (i.e., anomalous Hall effect, ρ_{xy}) and longitudinal resistivity (i.e., ρ_{xx}) in response to a small ac current applied along the longitudinal direction (x-direction). In current-assisted magnetization reversal experiments, a second current source delivers pulses of varying amplitude, while a writing field, B_w , is applied along the z-axis. Unless otherwise noted, all measurements were carried out at 2 K.

4.3 Joule-Heating Assisted Magnetization Reversal

First, we perform transport measurements under an external magnetic field to establish a reference for interpreting the current-assisted magnetization reversal experiments. Figure 4.1 (b) shows the transverse resistivity of the device, which contains contributions from the anomalous Hall effect (i.e., the hysteretic component) and the ordinary Hall effect (i.e., the linear-in-field background) over the field range of -3 T to 3 T . We distinguish two distinct resistivity states at $\rho_{xy} \approx \pm 0.3\text{ }\mu\Omega\text{ cm}$, corresponding to the DOWN and UP saturated magnetic states of $\text{Ni}_{1/4}\text{TaSe}_2$. From these measurements, we extract a coercive field of 2 T , required for complete magnetization reversal.

To demonstrate the current-assisted magnetization reversal, we first initialize the device in the UP state using an external magnetic field of 3 T . Then, we simultaneously apply a writing field of $B_w = -2\text{ mT}$ and 1 m sec current pulses, J_{DC} , of varying amplitudes. After each current pulse, we record the ρ_{xy} value. As shown in Figure 4.1 (c), a sharp transition occurs at a threshold current density of $J_{th} = 9 \times 10^4\text{ A cm}^{-2}$, where ρ_{xy} abruptly switches from $-0.3\text{ }\mu\Omega\text{ cm}$ to $0.3\text{ }\mu\Omega\text{ cm}$, indicating magnetization reversal. The final resistivity value above J_{th} matches that of a fully saturated DOWN state obtained under an external magnetic field in Figure 4.1 (b), confirming complete UP-to-DOWN reversal. An identical threshold current density is observed when the device is initialized in the DOWN state with -3 T and pulsed under $B_w = +2\text{ mT}$, resulting in full reversal to the UP state (triangular symbols in Figure 4.1 (c)). This establishes that reversible, back-and-forth magnetization switching can

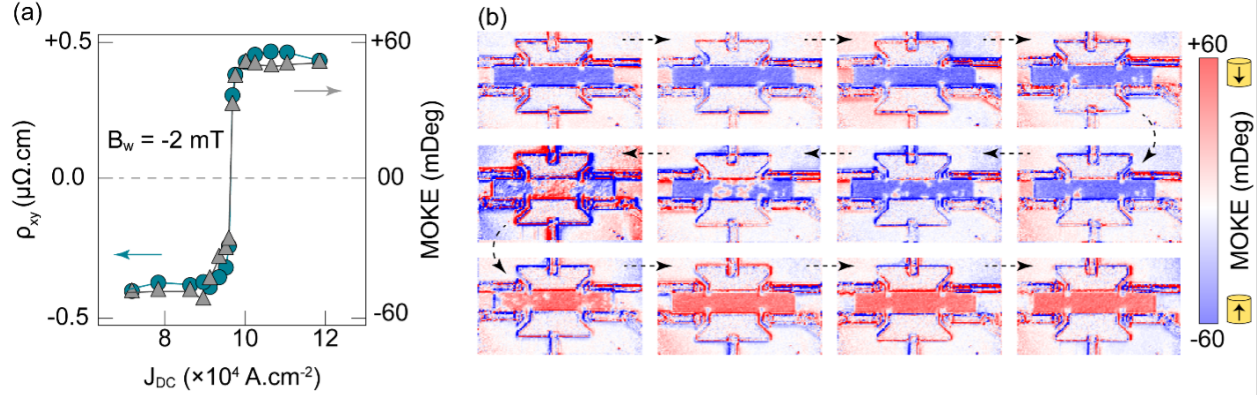


Figure 4.2: **Direct imaging of current-induced magnetization reversal using MOKE** (a) Current-induced reversal probed electrically via anomalous Hall resistivity (ρ_{xy} , left axis) and optically via Kerr rotation (MOKE, right axis). Both readout modalities consistently yield a threshold current density of $J_{th} \sim 9.7 \times 10^4 \text{ A}\cdot\text{cm}^{-2}$ (b) Sequence of Kerr microscopy maps acquired after successive current pulses of increasing amplitude, corresponding to the data points presented in panel (a). The device is initialized to the UP state under a 3 T magnetic field and then pulsed under a writing field of $B_w = -2 \text{ mT}$. Above the critical current density ($J > J_{th}$), the MOKE contrast switches sign, directly confirming magnetization reversal from UP to DOWN.

be reliably achieved by simply reversing the polarity of the writing field while maintaining a fixed pulse amplitude above J_{th} . Such reversible switching is demonstrated in Figure 4.1 (d). Notably, the writing field of $\pm 2 \text{ mT}$ is orders-of-magnitude smaller than the measured coercive field of 2 T , clearly highlighting that the reversal is primarily assisted by the applied current pulses. Furthermore, the switching behavior is insensitive to the polarity of the current pulses (Figure 4.5), providing the first indication of the Joule-heating-assisted reversal mechanism.

We further provide direct evidence of current-induced magnetization reversal by the magneto-optical Kerr effect (MOKE) microscopy, which enables the real-space visualization of the magnetic state of the $\text{Ni}_{1/4}\text{TaSe}_2$ under current pulses. As described in the Methods section, the polarization rotation of linearly polarized light upon reflection from the crystal (i.e., Kerr rotation) directly tracks the magnetization direction. Thus, UP and DOWN magnetization directions produce polarization rotations of opposite signs, allowing MOKE measurements to spatially map the magnetization state across the device. Following the protocol of Figure 4.1 (c), we initialize the device in the UP state using an external field of 3 T, then apply a writing field of $B_w \sim -2 \text{ mT}$ while sending current pulses of varying amplitudes. After each pulse, we simultaneously record the Kerr rotation and the anomalous Hall resistivity, and we plot them together in Figure 4.2 (a). One can immediately see that

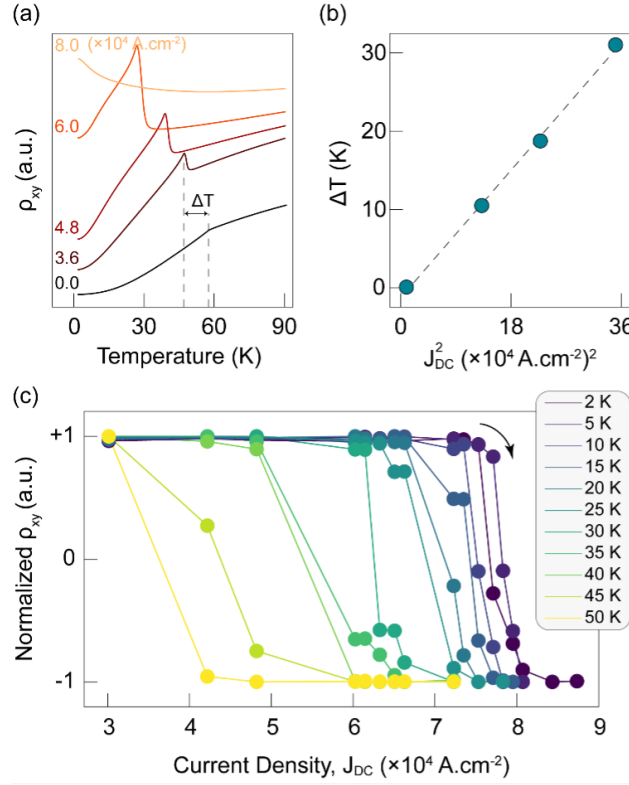


Figure 4.3: **Direct measurement of Joule heating.** (a) The temperature dependence of the longitudinal resistivity ρ_{xx} in response to a small measuring ac current of $J_{AC} = 20 \text{ A cm}^{-2} \times \cos \omega_o t$ superimposed on a large DC current (J_{DC}) of varying amplitudes as indicated on the figure. The total applied current is $J_{\text{total}} = J_{AC} + J_{DC}$, with ρ_{xx} calculated from the measurement of the ac component of V_{xx} at ω_o frequency. The shift of the apparent Curie temperature, T_c ($J_{DC} \neq 0$), relative to the principal Curie temperature, T_c ($J_{DC} = 0$), is defined as $\Delta T = T_c(J_{DC} \neq 0) - T_c(J_{DC} = 0)$. (b) The temperature change, ΔT , plotted as a function of J_{DC}^2 , exhibits a linear dependence, indicating that the observed variation arises from Joule heating during the applied DC current. (c) Temperature dependence of the switching response. The threshold current density (J_{th}) decreases with increasing temperature.

the jump in the Hall resistivity at J_{th} is accompanied by the sharp change in the sign of the Kerr rotation, confirming the magnetization reversal as the origin of the step-like response in both optical and electronic readouts.

Leveraging the Kerr contrast, we spatially map the magnetic configuration of $\text{Ni}_{1/4}\text{TaSe}_2$ across the Hall bar while gradually increasing the current density from below to above J_{th} (Figure 4.2 (b)). Above the threshold current, the device switches into a spatially uniform

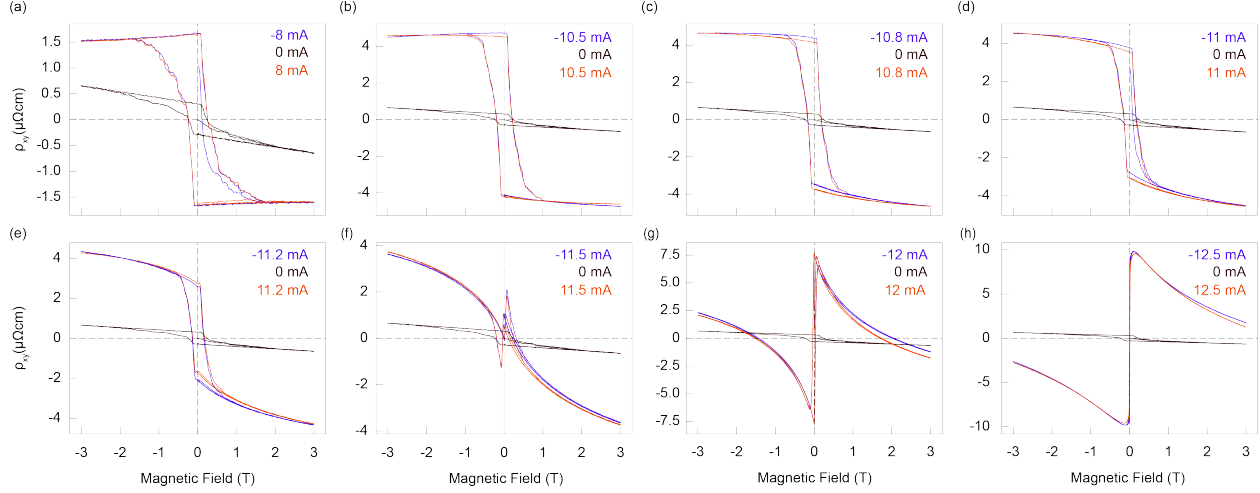


Figure 4.4: Measured Hall effect under a perpendicular magnetic field with an applied positive and negative DC current Anomalous Hall resistivity measured (at 2 K) with various applied DC currents. There is no current-dependent shift in the hysteresis loop at any values of the applied DC current. Ruling out the possibility of SOT-induced switching mechanism.

DOWN state across the entire channel, confirming a complete magnetization reversal. Once saturation is reached, additional current pulses induce no further change (bottom row, Figure 4.2 (b)). Importantly, the Kerr maps around J_{th} reveal the underlying reversal mechanism: magnetization switching begins with the nucleation of small domains of opposite polarity, followed by their growth and coalescence into a fully saturated monodomain state. Such a nucleation – growth mechanism is the characteristic of magnets with macroscopic dimensions, consistent with our Hall bar geometry of $1.77 \times 9.48 \times 46.9 \mu\text{m}^3$.

Having established the current-assisted magnetization reversal and its switching mode, we next examine the role of assisting current in the switching mechanism. We first exclude torque-based contributions [38], primarily because (i) reversing the current polarity does not alter the switching characteristics, and (ii) no asymmetric shift appears in the $\rho_{xy} - B_z$ hysteresis loop (Figure 4.4) in the presence of a DC switching current, a signature of torque-based switching [79, 80]. We believe that the Joule-heating due to the current pulses during the write cycle is the main mechanism at play. To unambiguously probe this effect, we measure the temperature-dependence of the longitudinal resistivity, ρ_{xx} , in the presence of a DC current bias. As shown in Figure 4.3 (a), we drive the device with a total current density

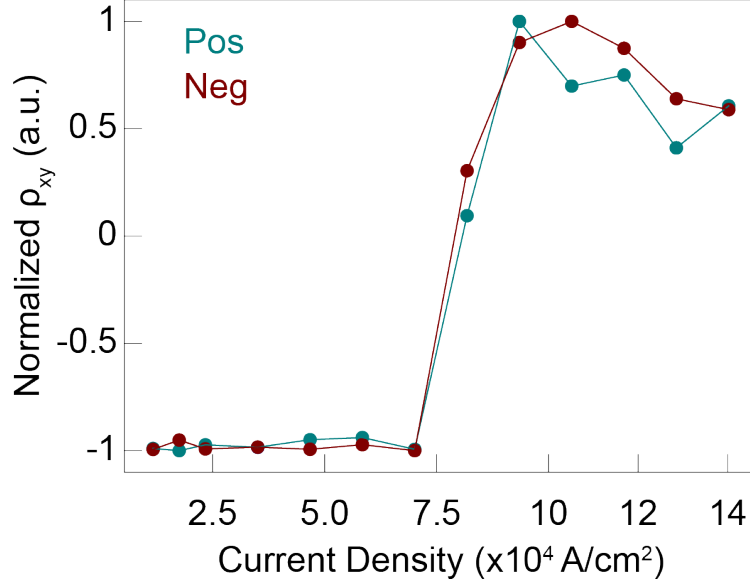


Figure 4.5: **Pulse current polarity dependence on the switching response** Normalized anomalous Hall resistivity measured (at 2 K) after applying current pulses of opposite polarity. The device was initially set to a low resistance state with a 3 T external field. Reversing the polarity has the same effect on the switching response.

$$J_{total} = J_{DC} + J_{AC} \cos \omega_o t \quad (4.1)$$

where the probe current is much less than the applied constant dc current, $J_{AC} \ll J_{DC}$.

Using lock-in detection technique at the ac-drive frequency of $\omega_o = 37.77$ Hz, we measure the V_{xx} , from which we calculate ρ_{xx} . At $J_{DC} = 0$, the temperature-dependence of ρ_{xx} exhibits a kink at 55 K, corresponding to the equilibrium Curie temperature of $\text{Ni}_{1/4}\text{TaSe}_2$ compound. With increasing J_{DC} , the kink shifts progressively to lower values, which we refer to as the “apparent” Curie temperature. Such a downshift indicates that the $\text{Ni}_{1/4}\text{TaSe}_2$ must be cooled below its equilibrium Curie temperature of 55 K for an ordered state to be established in the crystal. At current density approaching $\sim 8 \times 10^4 \text{ A cm}^{-2}$, the apparent Curie temperature is suppressed to zero, implying that long-range ferromagnetic order cannot be sustained at any accessible temperature. Importantly, this critical current density of $8 \times 10^4 \text{ A cm}^{-2}$ coincides with the threshold current density $J_{th} \sim 9 \times 10^4 \text{ A cm}^{-2}$ identified independently from transverse resistivity ρ_{xy} and MOKE measurements shown in Figure 4.1 and Figure 4.2. Thus, these results provide unambiguous evidence that, at J_{th} , the current pulses transiently heat the $\text{Ni}_{1/4}\text{TaSe}_2$ medium above its Curie temperature. During this short-lived paramagnetic window, a small writing field of $B_w = \pm 2 \text{ mT}$, 100 times smaller

than the coercive field, is sufficient to reverse the magnetization. This experiment, therefore, constitutes direct proof of Joule-heating-assisted magnetization reversal.

As shown in Figure 4.3 (b), the shift of the apparent Curie temperature relative to its equilibrium value given by,

$$\Delta T = T_c(J_{DC} \neq 0) - T_c(J_{DC} = 0) \quad (4.2)$$

scales quadratically with the applied current density, that is $\Delta T \propto (J_{DC})^2$. This quadratic dependence stems from the Joule heating. in the context of EAMR, the extra energy supplied by the current pulses follows

$$E \propto \tau_{pulse} \times R \times (J_{DC})^2 \quad (4.3)$$

where R and τ_{pulse} are the device resistance and the pulse width, respectively. At base temperature of $T_o = 2$ K, a current density of $J_{th} \sim 9 \times 10^4$ A cm⁻² increases the temperature of Ni_{1/4}TaSe₂ device by ~ 52 K (i.e., 55 K $- T_o \sim 52$ K). Accordingly, at higher measurement temperatures, smaller temperature increases are required to reach the Curie point, and thus lower current densities are sufficient for complete magnetization reversal. This technologically favorable scaling is experimentally demonstrated in Figure 4.3 (c). Furthermore, the observed current-density dependence is quantitatively consistent with the Equation 4.3 particularly when accounting for the temperature dependence of the resistivity.

We note that the DC current used in measurements of Figure 4.3 were constant (not pulsed), yet the extracted threshold current closely matches the value obtained from pulsed experiments, as discussed above. This agreement indicates that during the 1 msec current pulse the system reaches the thermal equilibrium. However, reducing the pulse width eventually increases the required threshold current density for magnetization reversal (Figure 4.6).

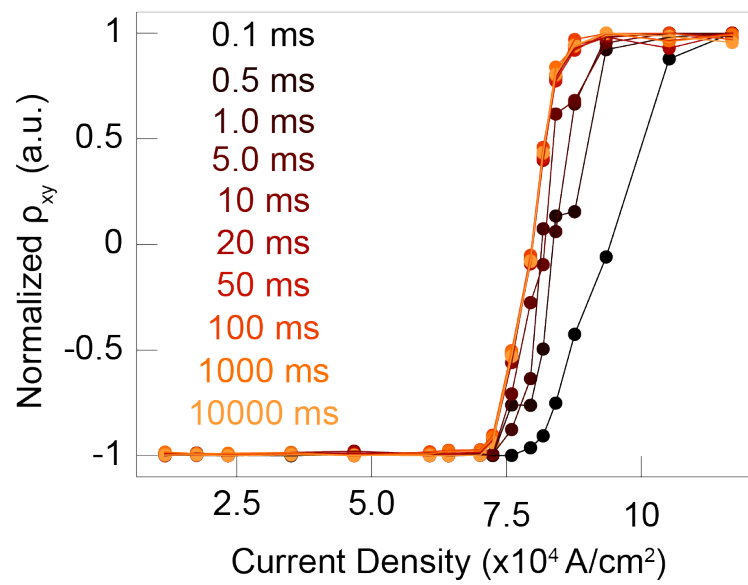


Figure 4.6: **Pulse width dependence** Normalized anomalous Hall resistivity measured (at 2 K) after applying current pulses of various amplitudes and various pulse durations. The device was initially set to a low resistance state with a 3 T external field. Reducing the pulse duration eventually increases the required threshold current density for magnetization reversal.

Chapter 5

Cobalt Intercalated Tantalum Disulfide

“I am going to do a little tax fraud.”

— Dr. Yuanqi Lyu

Materials in this chapter were previously published in

- Jonathan Kruppe, Josue Rodriguez, et al., *Anisotropic multi-Q order in Co_xTaS_2* . npj Quantum Mater. (2026). <https://doi.org/10.1038/s41535-026-00856-w>

5.1 Introduction

The first neutron-diffraction investigation established $\text{Co}_{1/3}\text{TaS}_2$ as a triangular-lattice antiferromagnet with a coplanar 120° spin configuration described by $\mathbf{q}_m = (1/3, 1/3, 0)$ ordering vector and a magnetic supercell of $\sqrt{3} \times \sqrt{3}$ times the crystallographic cell [82]. Interest in this material platform was reignited following the discovery of a large anomalous Hall effect (AHE) in the metallic triangular lattice antiferromagnet Co_xTaS_2 despite an almost zero net magnetization [83, 84]. Temperature dependent magnetization measurements showed that Co_xTaS_2 undergoes two magnetic phase transitions, $T_{N1} \sim 35$ K and $T_{N2} \sim 26.5$ K where the AHE manifest at the lower phase transition. *Ab initio* calculations based on the 120° ordering suggested that a hourglass-shaped Weyl fermions from the non-symmorphic symmetry triggers the AHE and that the AHE could be characterized by a field-tunable toroidal moment [84]. However, in this study, the cobalt intercalation reported was $x = 0.29$.

Subsequent neutron-scattering experiments in samples with intercalation $0.29 \leq x \leq 0.325$ revealed that Co_xTaS_2 undergoes a transition from a collinear Single- $\mathbf{Q}$ ($T_{N2} \leq T \leq T_{N1}$) to a non-coplanar Triple- $\mathbf{Q}$ ($T \leq T_{N2}$). The true ground state below $T_{N2} \approx 26.5$ K

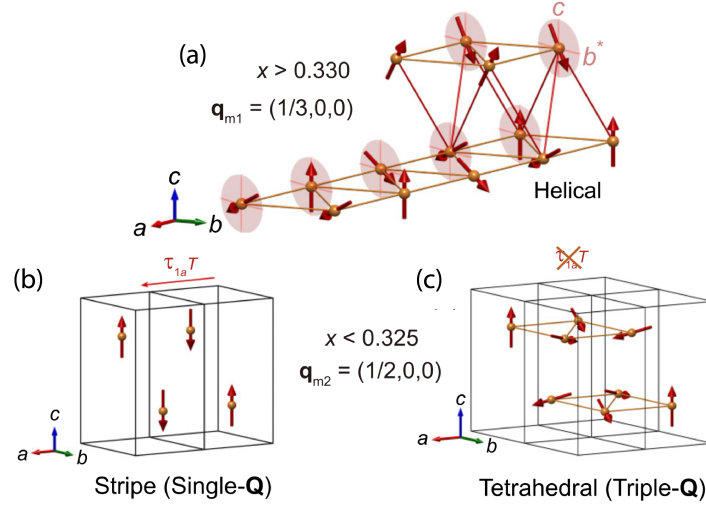


Figure 5.1: **Magnetic Order of Co_xTaS_2** Magnetic ordering was determined by Rietveld refinement of powder neutron diffraction measurements. (a) Coplanar Helical magnetic order described by a Single- $\mathbf{q}$ with magnetic moments lying in b^*-c plane for $x = 0.33 - 0.34$ (b)-(c) Magnetic order at intercalation $x = 0.325$. A collinear magnetic order described by Single- $\mathbf{q}$ at an intermediate temperature $26.5 \text{ K} < T < 35 \text{ K}$. Below 26.5 K the system orders into a non-coplanar magnetic order described by Triple- $\mathbf{Q}$ which manifest the AHE. The figure was taken from reference [81].

is a non-coplanar tetrahedral Triple- $\mathbf{Q}$ magnetic order ($\mathbf{q}_{m2} = (1/2, 0, 0)$ and symmetry-related vectors) [85, 86]. Each four-site magnetic unit forms an all-in/all-out tetrahedron with uniform scalar spin chirality producing a large spontaneous Hall conductivity $\sigma_{xy}(\mathbf{H} = 0) \simeq 70 \text{ } \Omega^{-1}\text{cm}^{-1}$. Thus showing a direct microscopic link between non-coplanar spin topology and Berry-curvature-driven transport in a metallic triangular lattice antiferromagnet (TLAF). Their results demonstrated that the Hall response originates from a non-coplanar antiferromagnetic state carrying finite scalar spin chirality, confirming that real-space Berry curvature can generate Hall conductivity even in materials with vanishing net magnetization. Most recently, Park *et al.* demonstrated that small deviations from the ideal composition ($0.299 \leq x \leq 0.34$) drives a transition between two distinct magnetic phases [81]. For $x \geq 0.330$ Co_xTaS_2 undergoes a phase transition into a coplanar helical antiferromagnet described by a Single- $\mathbf{Q}$ ordering vector ($\mathbf{q}_{m1} = (1/3, 0, 0)$) with $\sigma_{xy}(\mathbf{H} = 0) = M_z(\mathbf{H} = 0) = 0$. The magnetic structure is shown in Figure 5.1 (a). In contrast for $x \leq 0.325$, Co_xTaS_2 retains the non-coplanar Triple- $\mathbf{Q}$ state with spontaneous $\sigma_{xy}(\mathbf{H} = 0) \neq 0$. Figure 5.1 (b) shows the high temperature magnetic structures for $x = 0.325$ that preserves the combined lattice translation (τ_{1a}) and time-reversal symmetry (T). The non-coplanar magnetic structure is illustrated in Figure 5.1 (c), the scalar spin chirality breaks time-reversal

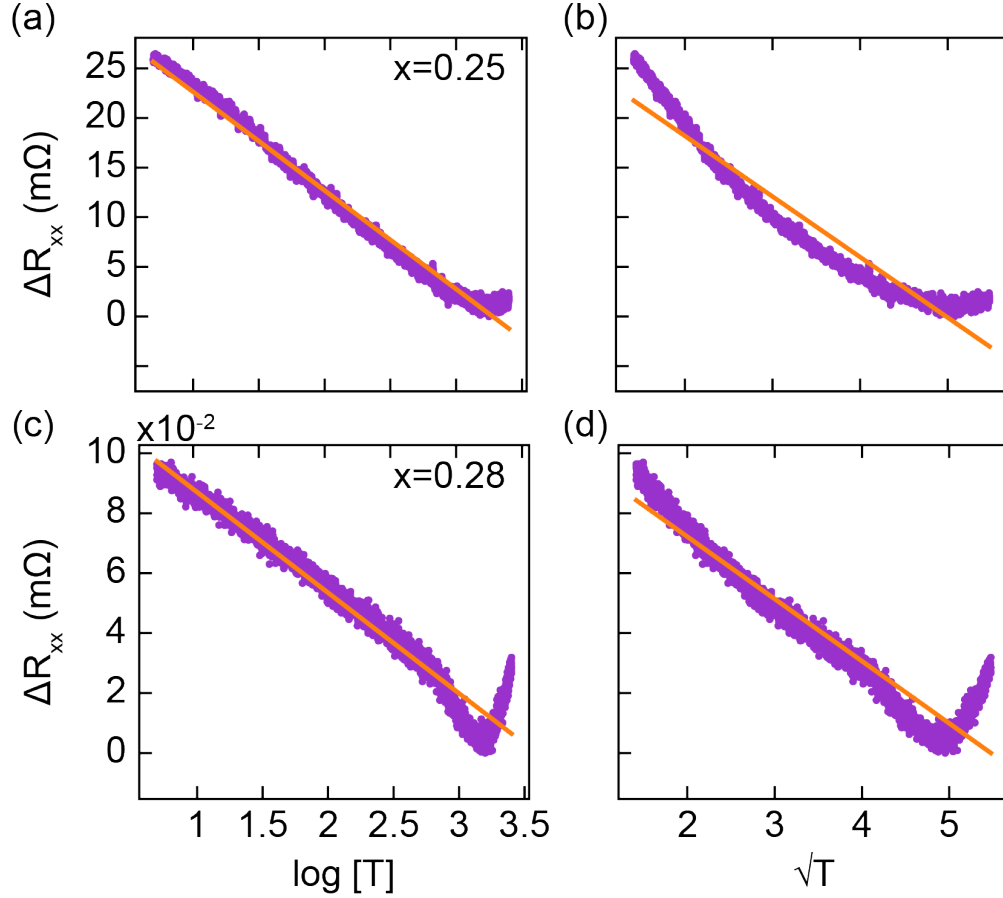


Figure 5.2: **Scaling analysis of the low-temperature resistivity upturn in Co-intercalated TaS₂.** Diagnostic scaling plots of the low-temperature resistance change $\Delta R_{xx} = R_{xx}(T) - R_{xx}(T_{min})$ for $T \leq 30$ K is plotted as a function of (a) and (c) $\log[T]$, (b) and (d) $\sqrt{T}$ for samples with intercalation ratios of $x = 0.25$ and $x = 0.28$, respectively. Linear fits (orange lines) are included as guides to the eye to identify the scaling that most closely describes the low-temperature transport behavior. A logarithmic temperature dependence is expected for Kondo-like scattering from localized magnetic moments, and a $\sqrt{T}$ dependence associated with electron–electron interaction corrections in disordered metals [64, 65].

symmetry and by extension, the combined $\tau_{1a}T$ symmetry is broken.

5.2 Bulk Transport Measurements

To distinguish a Kondo-like logarithmic upturn from disorder-driven interaction corrections, we examined the low-temperature transport below the resistivity minimum using diagnostic plots of R_{xx} versus $\log[T]$, R_{xx} versus $\sqrt{T}$ and R_{xx} versus T^2 . A linear dependence in R_{xx} versus $\log[T]$ is consistent with Kondo-like scattering where conduction electrons scatter off localized magnetic moments, producing a logarithmic enhancement of resistivity as temperature decreases [70, 69]. Linearity in R_{xx} versus $\sqrt{T}$ is more consistent with electron–electron interaction corrections in disordered three-dimensional metals, where diffusive electron motion modifies the density of states near the Fermi level [64, 65]. The T^2 scaling, reflects dominant electron–electron scattering within a coherent quasiparticle band, consistent with Landau Fermi-liquid theory [66, 67].

The change in longitudinal resistance was calculated by $\Delta R_{xx} = R_{xx}(T) - R_{xx}(T_{\min})$ where $R_{xx}(T_{\min})$ corresponds to the minimum resistance occurring at $T_{\min} = 25.7$ K and $T_{\min} = 24.5$ K for intercalation of $x = 0.25$, $x = 0.28$, respectively. Figure 5.2 (a-d) show ΔR_{xx} plotted as a function of $\log[T]$, and $\sqrt{T}$ for samples with intercalations $x = 0.25$ and $x = 0.28$. The $\log[T]$ scaling displays the most linear behavior over the measured temperature range for both compositions, while the other two scalings show larger deviations from linearity. These observations suggest that the resistance upturn is consistent with a logarithmic temperature dependence commonly associated with Kondo-like scattering of conduction electrons from localized magnetic moments. In Co_xTaS_2 , intercalated cobalt ions introduce localized magnetic moments within the van der Waals gaps of the host lattice providing a plausible mechanism for such scattering process. Notably, comparable low-temperature resistivity upturn is not typically observed in other intercalated transition metal dichalcogenides such as Co_xNbS_2 , Fe_xNbS_2 , or Fe_xTaS_2 , suggesting that the observed behavior may arose from scattering mechanism specific to the cobalt intercalated system.

Figure 5.3 (a) shows the normalized resistivity as a function of temperature for different intercalations of Co_xTaS_2 . The curves were normalized by the resistivity value at the base temperature, 2 K for clarity. All samples exhibit a kink as the temperature approaches the transition temperature T^* . Above T^* , all the samples exhibit metallic behavior. However, for $x = 0.25$, 0.28, an anomalous upturn in resistivity is observed.

To see the temperature evolution of the AHE, the longitudinal (ρ_{xx}) component of the resistivity must be removed from the transverse (ρ_{xy}) signal of the resistivity. To this end, the samples were field cooled from 100 K to 2 K with an applied field 1 T (−1 T). Then the hall voltage was measured from 2 K to 100 K with the field removed. The resulting data sets were antisymmetrized to remove the longitudinal component. In Figure 5.3 (b) curves show $\rho_{xy} = 0$ above a critical temperature T^* and below T^* , the hall effect ρ_{xy} takes a nonzero value. For intercalation $x = 0.28$, there is a second transition at ~ 5 K that is not observed in the corresponding ρ_{xx} .

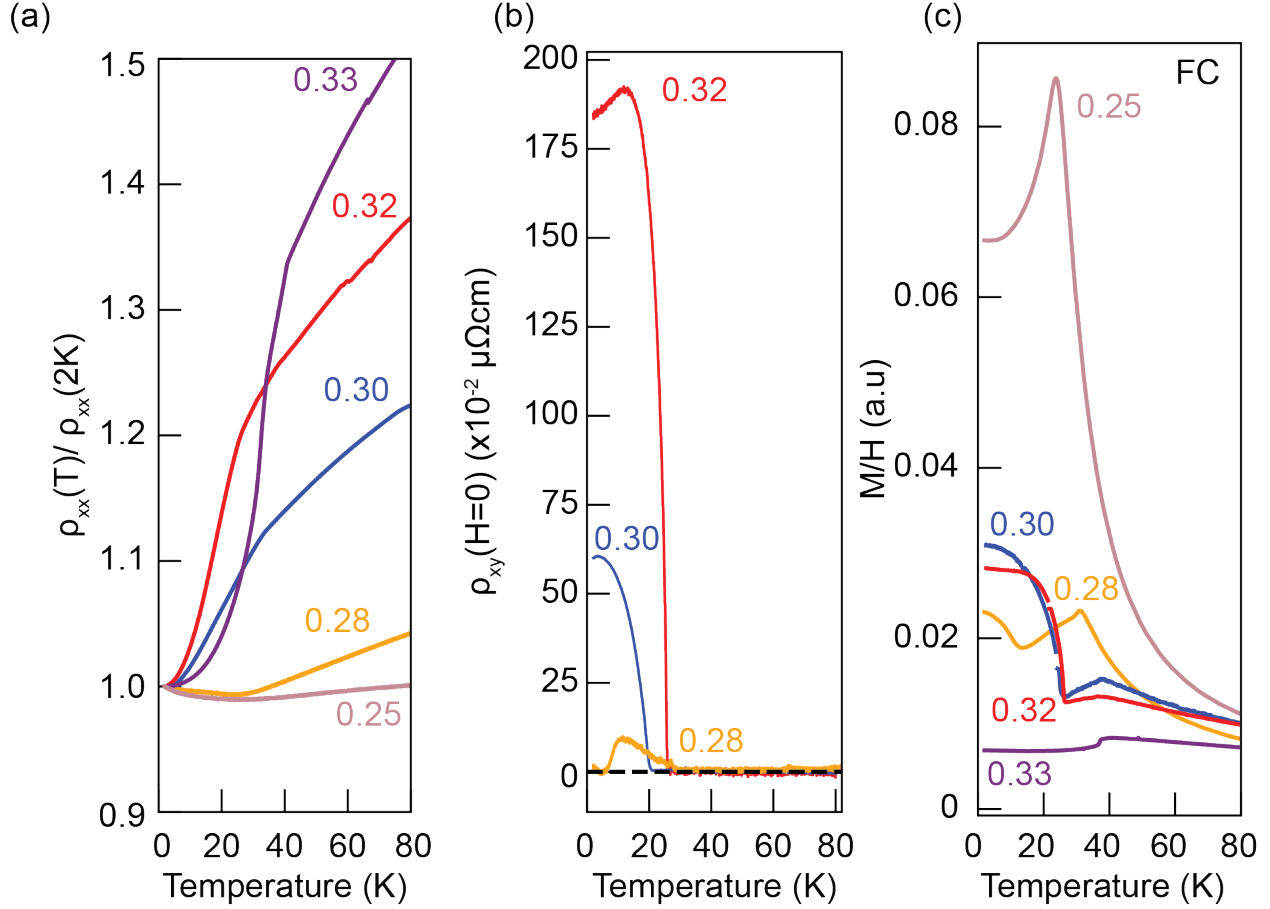


Figure 5.3: **Intercalation dependence of the magnetization, resistivity, and hall resistivity versus temperature in Co_xTaS_2 .** (a) Normalized resistivity of Co_xTaS_2 . For all intercalations metallic behavior is seen down to 2 K except for $x = 0.25, 0.28$ where an upturn in the resistivity at ~ 25 K (b) Hall resistivity of Co_xTaS_2 for three different intercalations measured after a field cooling process. (c) Field cooled magnetization of Co_xTaS_2 for various intercalations. The curves have been normalized by the high temperature magnetization (linear regime) for clarity.

Field-cooled magnetic susceptibility was also measured for various intercalations of the material Co_xTaS_2 . The susceptibility curves were normalized to the high-temperature regime, where susceptibility is in the linear regime. For $x = 0.25$ and $x = 0.33$, the magnetic susceptibility exhibits a single cusp at 23.8 K and 38.9 K, respectively. Such a sharp cusp in $\chi(T)$ is a well-known hallmark of an antiferromagnetic (AFM) transition, reflecting the onset of long-range AFM order. By contrast, the other intercalations show *two* distinct anomalies in the temperature dependence of the susceptibility. The higher-temperature anomaly has the form

x	RvT	$R_{\text{H}}\text{vT}$	MvT
0.25	27 K	-	23.8 K
0.28	25 K	5 K, 29 K	13.8 K, 31.3 K
0.30	32 K	20.2 K	25.9 K, 37.5 K
0.32	26.5 K	26.5 K	26.6 K, 36.8 K
0.33	41 K	-	38.9 K

Table 5.1: Transition temperatures, T^* , obtained from resistivity, hall effect and magnetization for different intercalations.

of an AFM-like cusp, consistent with antiferromagnetic ordering. The lower-temperature anomaly is more ferromagnetic (FM) in character, displaying an upturn/divergence in $\chi(T)$ consistent with the development of a net magnetization component. This two-step behavior indicates that, for those concentrations, the system first enters an antiferromagnetically ordered state and subsequently undergoes an additional low-temperature transition that generates a weak ferromagnetic component.

Anomalous Hall Effect

Field dependent Hall measurements were carried out on various cobalt intercalation levels spanning from $x \approx 0.25 - 0.33$ over a wide temperature range. Figure 5.4 compares the magnetic-field dependence of the anomalous Hall resistance. At low intercalation ($x = 0.25$) the Hall response is weak and has a broad hysteresis ($H_c \sim 6$ T) at low temperatures (2-5 K). By ~ 20 K, the field dependence is relatively flat, with only a small step-like feature near zero field and the hysteresis has collapsed, indicating the suppression of long-range magnetic order or a strong reduction in the net Berry curvature. By 40-60 K, the response is essentially featureless. The origin of the anomalous Hall effect is a mystery. As described in section 1.5, $\text{Co}_{1/4}\text{TaS}_2$ retains centrosymmetric symmetry ($P6_3/\text{mmc}$), the temperature dependent magnetization shows AFM behavior and the field dependent magnetization exhibit hysteretic behavior albeit with small magnetization ($m_z(\mathbf{H} = \mathbf{0}) < 0.1\mu_{\text{B}}$) and there is no known neutron diffraction measurements in the literature to elucidate the type of AFM order. A similar compound $\text{Co}_{1/4}\text{TaSe}_2$ has recently been shown to order as an itinerant A-type (collinear) AFM with no signatures of an AHE [87]. One possible explanation for the observation of the AHE in $\text{Co}_{1/4}\text{TaS}_2$, lies in the possibility that it forms a non-coplanar AFM structure giving rise to scalar spin chirality that forms a real space Berry curvature that then drives the AHE. The other is that it forms a coplanar AFM ordering in the presence of non-negligible spin-orbit interaction which breaks the TS symmetry allowing a finite Berry curvature thus driving an AHE [88].

The $x = 0.28$ sample shows a qualitatively different behavior, characterized by a smooth antisymmetric Hall response that evolves continuously with magnetic field. At low temper-

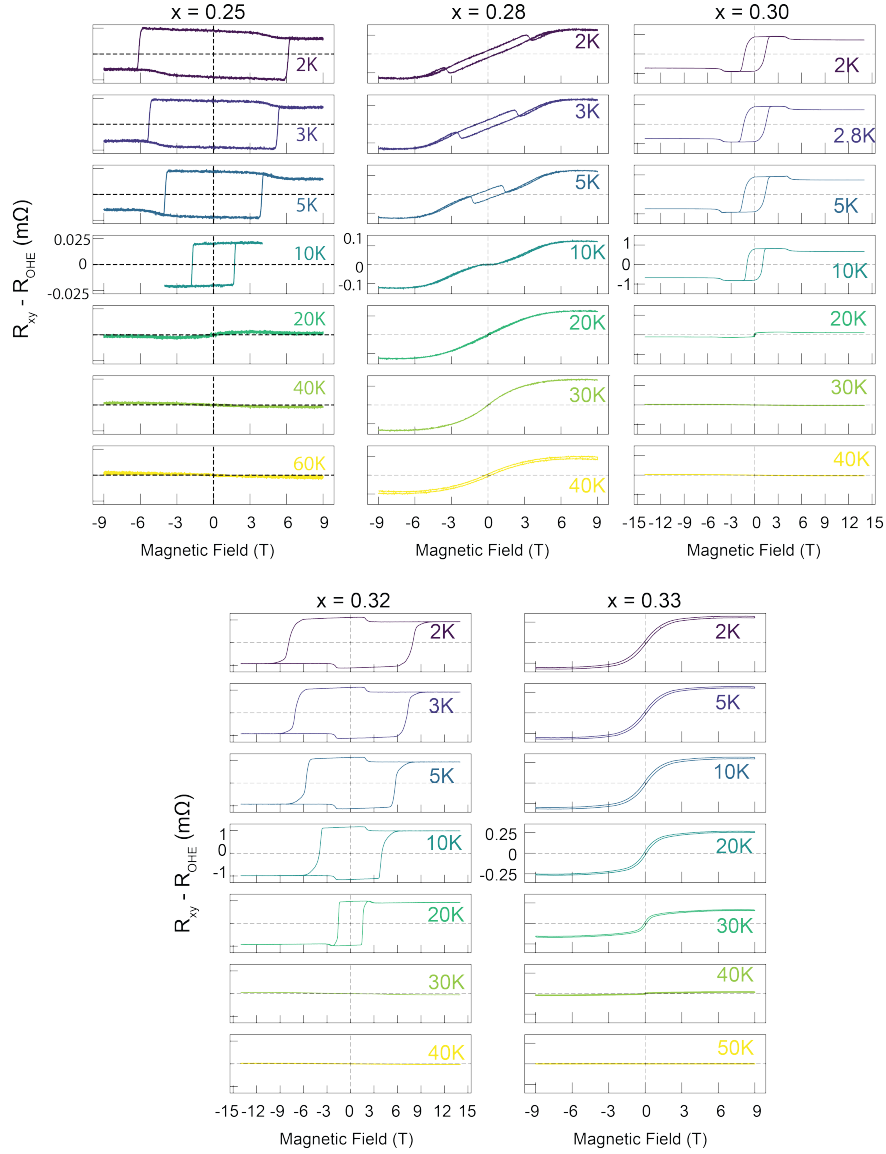


Figure 5.4: **Intercalation dependence of the Hall effect in Co_xTaS_2** The anomalous Hall resistance was obtained from subtracting the ordinary Hall effect from the measured transverse resistance. The data set has been anti-symmetrized.

atures (2-5 K), the Hall response displays a pronounced S-shape with minimal hysteresis, reminiscent of a field-driven reorientation process as opposed to an abrupt switching. The Hall slope decreases gradually with increasing temperature, persisting up to ~ 40 K before becoming weak and nearly linear. Compared to $x = 0.25$, the response is larger in magnitude and more coherent across temperature. For $x = 0.30$, the Hall response becomes sharply hys-

teretic at low temperatures, with clear switching events at well defined coercive fields ($H_c \sim 1.5$ T). At 2-10 K, the loops are rectangular in shape, indicating robust magnetic domains with abrupt reversal. The hysteresis remains visible up to 20 K but is reduced in amplitude and coercivity. Above 30 K, the Hall signal is zero. This behavior marks a transition from field response to domain-dominated switching.

At $x = 0.32$, the Hall response is maximally pronounced and strongly hysteretic and low temperatures. The 2-20 K data show large, square hysteresis loops with sharp switching, indicating strong magnetic anisotropy and well defined domain states. Multiple step-like features are visible at intermediate fields, suggesting complex domain or multi-stage switching. As temperature increases, the hysteresis gradually narrows and collapses by 30 kelvin, above which the Hall signal is zero. The $x = 0.33$ sample displays a smaller but smoother Hall response compared to $x = 0.32$. At the lowest temperatures, the field dependence is nonlinear but with a reduced hysteresis, indicating a more continuous field-driven evolution of the magnetic state. The Hall signal persists to higher temperatures (~ 30 K) the switching is less abrupt, suggesting a more homogeneous or less strongly pinned domain configuration. Taken together, the data reveal a non-monotonic evolution of Hall amplitude and hysteresis with intercalation, with the strongest switching behavior occurring near the commensurate compositions.

Magnetoresistance

Figure 5.5 shows the magnetic field dependence of the magnetoresistance (MR) for intercalations spanning $x \approx 0.25 - 0.33$ measured over a wide range of temperatures. The magnetoresistance was calculated according to Equation 5.3. At the lowest intercalation ($x = 0.25$), the MR exhibits a broad, dome like field dependence with weak structure in a temperature range of 2-5 K, indicative of gradual field-induced supersession of spin disorder. The MR magnitude decreases monotonically with increasing temperature, and above 20 K the response becomes smooth and weakly field dependent, with little evidence of hysteresis. At $x = 0.28$, the MR remains largely symmetric in magnetic field and displays a smooth, nearly parabolic field dependence across all temperatures. The low-temperature MR is enhanced compared to $x = 0.25$, but shows minimal hysteresis, consistent with a continuous field-driven reorientation of the magnetic texture rather than discrete domain switching. The MR gradually weakens with increasing temperature but remains discernible up to 40 K and the hysteresis disappears above 20 K.

In contrast, the $x = 0.32$ sample shows pronounced low-temperature MR anomalies, including sharp features and hysteretic behavior. At 2-10 K, the MR curves exhibit abrupt changes in the butterfly hysteresis that coincide with field scales associated with magnetic switching, reflecting strong coupling between charge transport and magnetic domain re-configuration. These features weaken progressively with increasing temperature and are largely absent above 30-40 K, where the MR reverts to a smooth, weakly field-dependent

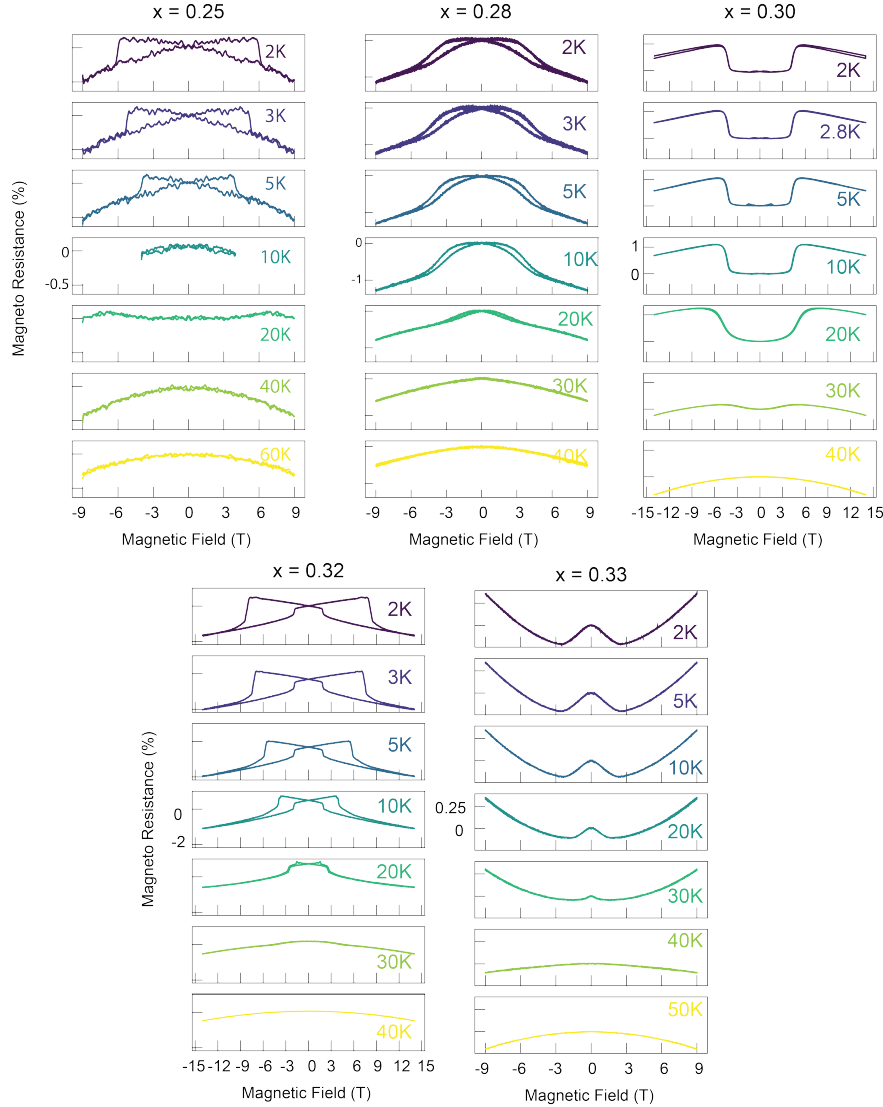


Figure 5.5: **Intercalation dependence of the magnetoresistance in Co_xTaS_2** The magnetoresistance in percentage has been symmetrized.

form or negative magnetoresistance. At the highest intercalation level ($x = 0.33$) and for temperatures below 40 K, the magnetoresistance exhibits a pronounced local maximum at zero field and initially decreases with increasing magnetic field ($d\rho/dH < 0$). Upon further increasing the field beyond a characteristic crossover scale, the MR becomes positive ($d\rho/dH > 0$). At low fields, canting of the localized moments in the helical antiferromagnetic state toward the applied field suppresses spin-disorder and spin-flip scattering, resulting in a reduction of the resistivity. At higher fields, the classical orbital contribution dominates,

where the Lorentz force bends charge-carrier trajectories and produces a positive, approximately parabolic MR. The zero-field MR maximum remains discernible up to 30 K, but its amplitude is progressively reduced with increasing temperature. Above 40 K, this low-field feature is fully suppressed and the MR response is governed by a monotonic positive field dependence.

Magnetization Field Sweeps

Figure 5.6 shows the magnetic-field dependence of the magnetization $M_z(\mathbf{H})$ for samples spanning $x \approx 0.25 - 0.33$ measured over a wide temperature range. At low intercalation ($x = 0.25$), the magnetization exhibits the largest overall magnitude and a pronounced non-linearity at low temperatures, with a visible hysteresis at fields up to 3 T that gradually evolves into a S-shape between 15 K-35 K. The absence of sharp saturation and the smooth field dependence are consistent with antiferromagnetic order with a sizable susceptibility and weak field-induced canting. For $x = 0.28$, the magnetization is reduced in magnitude compared to $x = 0.25$ and displays a largely linear field dependence across all temperatures, with minimal hysteresis below 10 K. This behavior indicates a predominantly antiferromagnetic response characterized by continuous field-driven canting rather than abrupt spin or domain switching, consistent with the smooth Hall and MR responses observed at this composition.

At intermediate intercalation ($x = 0.30$), the magnetization curves remain mostly linear at higher temperatures but develop clear hysteresis below 25 K, which is consistent with the second transition in the FC magnetization at the same intercalation. Between 30 K-10 K a step like field transition appears that remains outside the hysteresis loop and below 10 K the hysteresis loop grows and envelops the step like feature. The apparent jump in magnetization is induced due to meta-magnetic transition. These features reflect field-driven reconfiguration of the magnetic structure or domain populations, occurring at field scales similar to those of the switching features seen in the AHE and MR, indicating strong coupling between magnetization and transport near this composition.

Near commensurate ordering ($x = 0.32$), the magnetization shows the most pronounced low-temperature anomalies, including enhanced curvature (meta-magnetic transition) and clear hysteresis (below 25 K), while remaining unsaturated up to the highest measured fields. In this case, the hysteresis loop envelops the meta-magnetic transition below 15 K. The persistence of linear behavior at high fields together with low-field hysteresis suggests a robust antiferromagnetic background with strongly pinned domains or spin reorientation transitions.

At the highest intercalation level ($x = 0.33$), the magnetization magnitude is strongly suppressed, nearly an order of magnitude smaller than at lower x , and the field dependence is strictly linear across all temperatures with no observable hysteresis. This behavior is consistent with a more homogeneous magnetic state or reduced ordered moment, in agreement

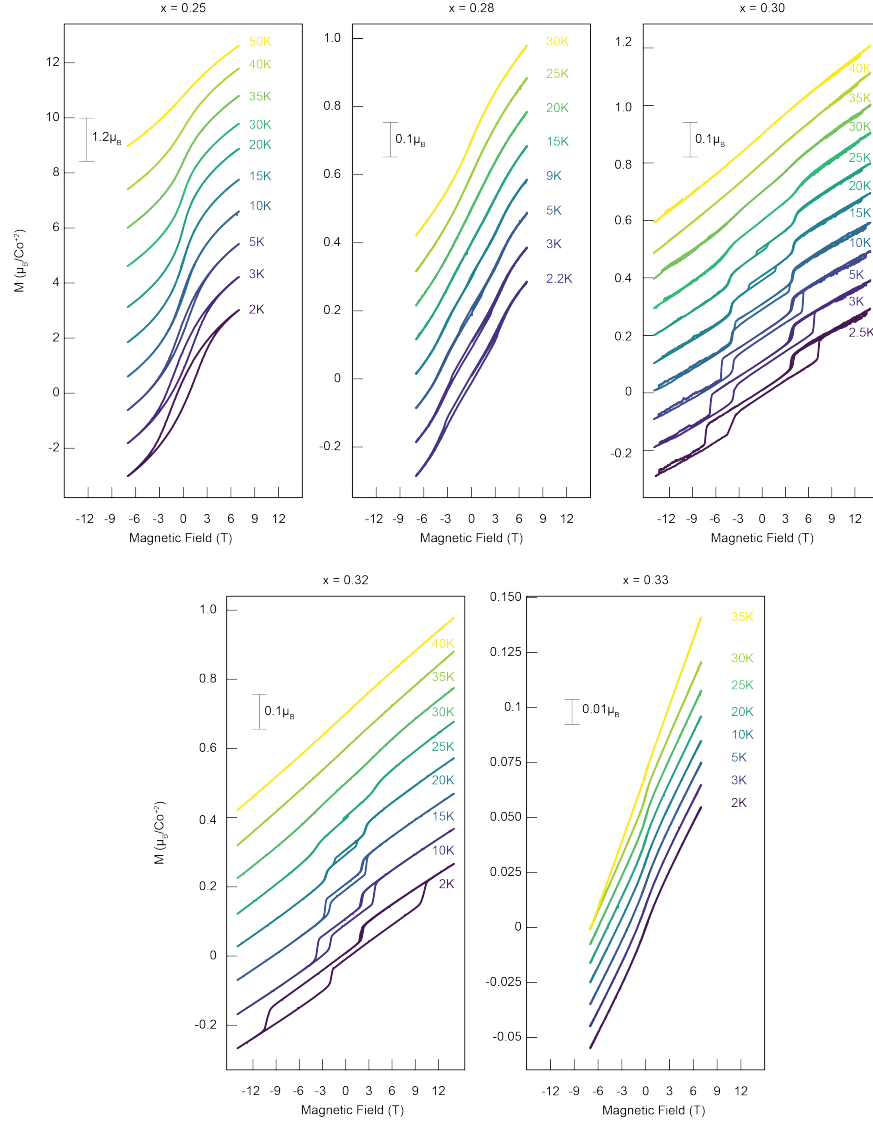


Figure 5.6: **Magnetization versus Field of Co_xTaS_2** For $x = 0.25$, field sweeps look similar to typical FM. No saturation is observed up to 7 T and hysteresis closes below 10 K. For $x = 0.28$, hysteresis closes below 20 K. For all the data sets, the curves were offset for clarity.

with the absence of sharp switching features in the corresponding MR and AHE data.

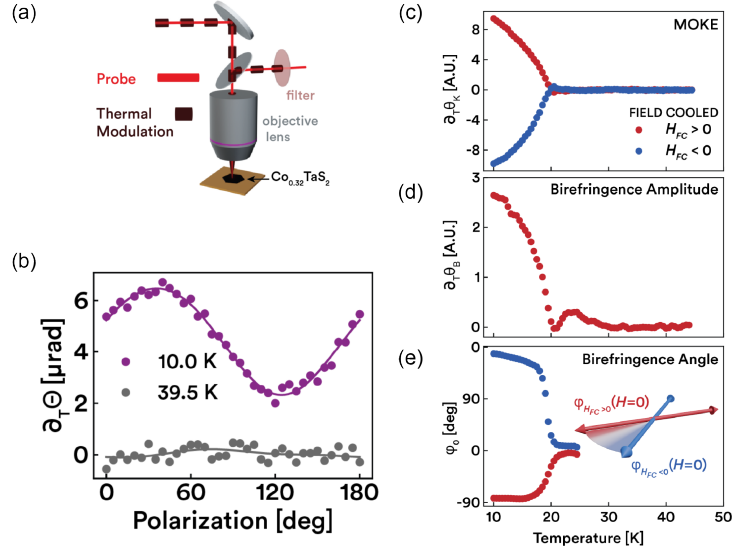


Figure 5.7: **Temperature dependence of thermally-modulated magneto-optical effects** (a) Schematic of optical setup for measuring polarization rotation (Θ) as a function of incident polarization (φ): (b) Thermally-modulated polarization rotation polarization at low temperature (10 K) and in the paramagnetic phase (above 39 K), (c) Temperature dependence of thermally-modulated MOKE (d) Temperature dependence of thermally-modulated birefringence (e) Temperature dependence of thermally-modulated principal axis orientation (φ_o) [89]

Magneto-Optical Measurements in $\text{Co}_{0.325}\text{TaS}_2$

In collaboration with Krupp in the Orenstein group, magneto-optical measurements were carried out on a $\text{Co}_{0.325}\text{TaS}_2$ sample [89]. Symmetry-based analysis identified the AHE-active phase as an anisotropic (2+1)- $\mathbf{Q}$ state, in which magnetic modulation at one wave vector ($\mathbf{Q}$) differs in symmetry from the other two wave vectors. The (2+1)- $\mathbf{Q}$ naturally supports a scalar spin chirality which gives rise to the AHE.

The sample was probed with linearly polarized light, and a change of polarization angle upon reflection as a function of incident light polarization was measured. This measurement is sensitive to time-reversal symmetry breaking and the three-fold rotational symmetry around the z axis (C_{3z}). Broken time-reversal symmetry yields the magneto-optical Kerr effect (MOKE) and broken C_{3z} yields birefringence, optical anisotropy. The measured polarization rotation is given by

$$\Theta(\varphi, T) = \theta_B(T) \sin [2(\varphi + \varphi_o(T))] + \theta_K(T) \quad (5.1)$$

where φ is the incident polarization, φ_o is the orientation of the optical principal axis, θ_B is the amplitude of birefringence, θ_K is the amplitude of the Kerr effect and T is the temperature. Figure 5.7 (b) shows the measured temperature modulated polarization rotation as a function of φ at 10 K (purple) and 39.5 K (gray). The low temperature curve shows broken time-reversal symmetry (offset) and broken C_{3z} symmetry (sinusoidal variation), while the high temperature curve shows neither dependence. Figure 5.7 (c) shows that the MOKE response behaves as expected from a broken-time-reversal symmetry state however, the non-zero birefringence in both temperature regions shows that the low temperature and high temperature phase of $\text{Co}_{0.325}\text{TaS}_2$ break rotational symmetry not know (Figure 5.7 (d)). Moreover, the birefringence angle abruptly becomes temperature-dependent at low temperatures and in zero-field (Figure 5.7 (e)). A symmetry analysis reveals the (2+1)-**Q** ground state can account for these three optical effects observed and allows a finite scalar spin chirality that drives the AHE in $\text{Co}_{0.325}\text{TaS}_2$.

5.3 Methods

Four-probe transport measurements were conducted in a Quantum Design 9 T and 14 T Dynacool PPMS. Standard lock-in ac measurements were done with a Zurich Instruments MFLI lock-in amplifier. An ac excitation current of 5 mA ran through the device's source and drain terminals. Simultaneously, the longitudinal (V_{xx}) and transverse voltage (V_{xy}) were recorded with a Zurich Instruments MFLI lock-in amplifiers. The field-dependent R_{xx} and R_{xy} data were symmetrized (anti-symmetrized) in accordance with Equations 5.2.

$$\begin{aligned}
 R_{xx,symm}^{Forward}(\mu_o H) &= \frac{1}{2} \left[R_{xx,raw}^{Forward}(\mu_o H) + R_{xx,raw}^{Reverse}(-\mu_o H) \right] \\
 R_{xx,symm}^{Reverse}(\mu_o H) &= \frac{1}{2} \left[R_{xx,raw}^{Reverse}(\mu_o H) + R_{xx,raw}^{Forward}(-\mu_o H) \right] \\
 R_{xy,asymm}^{Forward}(\mu_o H) &= \frac{1}{2} \left[R_{xy,raw}^{Forward}(\mu_o H) - R_{xy,raw}^{Reverse}(-\mu_o H) \right] \\
 R_{xy,asymm}^{Reverse}(\mu_o H) &= \frac{1}{2} \left[R_{xy,raw}^{Reverse}(\mu_o H) - R_{xy,raw}^{Forward}(-\mu_o H) \right]
 \end{aligned} \tag{5.2}$$

The field dependent R_{xx} was converted to magnetoresistance by Equation 5.3

$$\text{MR}\% = \frac{R_{xx}(\mu_o H) - R_{xx}(H = 0)}{R_{xx}(H = 0)} \times 100\% \tag{5.3}$$

The field dependent R_{xy} was separated into the ordinary hall effect (OHE) and anomalous Hall effect (AHE) according to the following equation

$$R_{AHE} = R_{xy} - R_{OHE} \quad (5.4)$$

The R_{OHE} term is linear in magnetic field and the term R_{AHE} in ferromagnetic materials is proportional to the magnetization. In systems with zero net magnetization the term may be driven by nonlinear or noncoplaner AFMs as described in section 1.4. The ordinary Hall resistance was determined by fitting a linear function at the high field limit of the measured transverse resistance and subsequently subtracted from the measured transverse resistance to give the anomalous Hall resistance. The measured transverse resistance was anti-symmetrized prior to the removal of the ordinary Hall effect.

Growth

All crystals were grown by chemical vapor transport method described in chapter 2.

Electron dispersive x-ray was acquired using an Oxford Instruments EDX detector integrated into a ThermoFisher SCIOS-2 dual-beam SEM. An accelerating voltage of 30 kV and a beam current of 3.2 nA were used for data collection.

$$\frac{\delta(\text{Co/Ta})}{\text{Co/Ta}} = \sqrt{\left(\frac{\delta\text{Co}}{\text{Co}}\right)^2 + \left(\frac{\delta\text{Ta}}{\text{Ta}}\right)^2} \quad (5.5)$$

The cobalt intercalation ratio x was determined using energy-dispersive X-ray spectroscopy (EDX) measurements performed at multiple locations across the surface of each single crystal. For each sample, EDX spectra were collected at $N=10-21$ distinct points within an area of approximately $1\text{ mm} \times 1\text{ mm}$ to assess both the average composition and the spatial uniformity of the intercalation. Quantitative elemental analysis was carried out by extracting the Co and Ta atomic concentrations and computing the ratio $x=\text{Co/Ta}$ at each measurement location. The reported intercalation value for each sample corresponds to the mean Co/Ta ratio obtained from all measured points. The associated uncertainty is taken as the standard deviation of the Co/Ta ratio, which reflects spatial inhomogeneity in the intercalation across the crystal.

In addition to spatial variations, the EDX technique introduces an intrinsic systematic uncertainty arising from counting statistics, detector calibration, and matrix correction procedures (ZAF or $\Phi(\rho z)$). In principle, the relative uncertainty of the Co/Ta ratio arising from instrumental effects can be estimated using Equation 5.5. In this expression, δCo (δTa)

denotes the intrinsic EDX uncertainty of the Co (Ta) concentration measured at a single location, and Co (Ta) is the corresponding measured concentration. This uncertainty reflects instrumental and systematic contributions and is distinct from spatial variations observed across multiple measurement points.. For transition-metal elements such as Co and Ta, this instrumental uncertainty is typically on the order of 1–3% relative, corresponding to an uncertainty in x of approximately 3% [90, 91]. This systematic contribution is nearly constant across samples and is reported separately from the spatial standard deviation. The mean intercalation values, spatial standard deviations, instrumental uncertainties, and the number of measured points for all samples are summarized in Table 5.2. For intercalated transition-metal dichalcogenides, the spatial variation in the Co/Ta ratio constitutes the dominant source of uncertainty and provides a direct measure of the homogeneity of the intercalation.

Nominal x	Mean x (EDX)	σ	$\delta x \sim 3\%$	N spots
0.25	0.25688	0.0038	0.0077	10
0.28	0.2848	0.0024	0.0085	11
0.30	0.3035	0.0044	0.0091	20
0.32	0.3225	0.0035	0.0095	10
0.33	0.3323	0.0051	0.0100	21

Table 5.2: EDX-derived Co intercalation ratios and uncertainties.

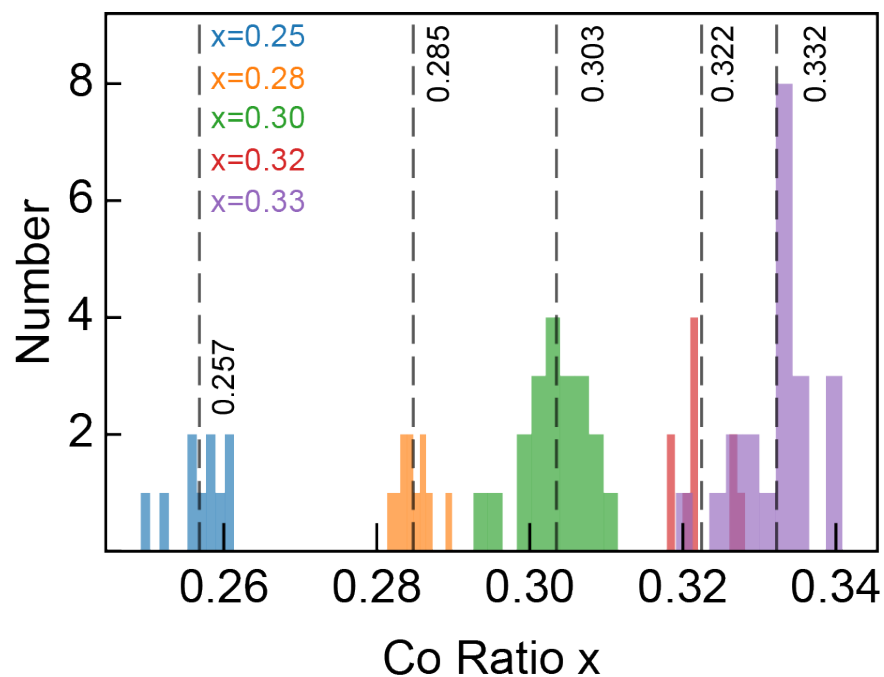


Figure 5.8: **Energy-dispersive X-ray (EDX) spectra for samples with various intercalation levels x .** Histograms of the Co intercalation ratio x determined from EDX analysis with a bin size of 0.002 for samples with nominal compositions $x \sim 0.25$ (light blue), $x \sim 0.28$ (orange), $x \sim 0.30$ (green), $x \sim 0.32$ (red), and $x \sim 0.33$ (violet). The dashed vertical lines indicate the average Co ratio for each sample, with the corresponding mean values.

Chapter 6

Switching in Cobalt Intercalated Tantalum Disulfide

“... it could always be worse.”

— Dr. Hossein Taghinejad

6.1 Introduction

The current-driven dynamics of antiferromagnetic spins provides a compelling example of the intricate interplay among symmetry, topology, and non-equilibrium phenomena [92, 37]. The principle in general is as follows: an applied current drives the system out of equilibrium and couple to a symmetry allowed component of the order parameter or its excitations. If the current is spin-polarized by some mechanism such as the spin-Hall effect or to the magnetization of the metallic medium it can transfer angular momentum to the magnetic order to favor one state or another. Helical and other non-collinear spin textures have recently drawn considerable attention owing to their complex symmetry and emergent functionalities, offering new avenues for the development of energy-efficient spintronic memory and logic devices [93, 94, 95].

Antiferromagnetic helical order, generally emerges due to the presence of competing antiferromagnetic exchange and/or Dzyaloshinskii-Moriya interactions (DMI) [57]. Helical spin configurations allow for higher packing densities, as the associated stray magnetic fields are greatly reduced or completely eliminated. Because of the strong antiferromagnetic exchange coupling, helical domain walls, or the helices themselves, can propagate rapidly and linearly under applied stimuli, presenting a key advantage for racetrack-type memory architectures [57, 96].

In the chiral antiferromagnet Mn_3Sn it was recently shown that the anomalous Hall effect

could be electrically switched in the presence of a bias field [55]. Similarly, the helically ordered intercalated transition-metal dichalcogenide $\text{Ni}_{1/3}\text{NbS}_2$ was shown to respond to applied currents. In both cases the current was applied perpendicular to the helical axis [56]. It is worth noting that most studies in this area use currents in the range of 10^7 A cm^{-2} to 10^9 A cm^{-2} which is similar to many of the original works on AFM switching known to cause significant heating.

In this chapter we study the system $\text{Co}_{1/3}\text{TaS}_2$, focusing specifically on composition $x \geq 0.33$. In the compositional range studied here, this material shows helical order with wave vector $\mathbf{q} = (1/3, 0, 0)$ and co-planar spins along the $b^* - c$ plane [81]. We show that devices created from single crystals show evidence of bi-directional switching when the current is applied parallel and perpendicular to the helical axis, responding to current densities orders of magnitude smaller than comparable studies, never exceeding $\sim 10^5 \text{ A cm}^{-2}$. Beyond a critical current, the system enters the Walker breakdown regime and all switching stops. We show that although the critical current is insensitive to applied magnetic fields, the Walker breakdown can be tuned by the direction of the applied field. This opens a new paradigm in spintronic switching in helical systems that is neither polar nor bi-directional but mono-directional and robust to extreme environments.

6.2 Methods

We synthesized 2D $\text{Co}_{1/3}\text{TaS}_2$ crystals using a standard chemical vapor transport method, as described in Chapter 2. the material consists of layered basal planes of 2H-TaS₂, with cobalt intercalants occupying the van der Waals gaps and aligning vertically with the tantalum atomic columns. As previously reported, $\text{Co}_{1/3}\text{TaS}_2$ is an itinerant metallic antiferromagnet with a Neel temperature of 35 K. The system orders into coplanar helical magnetic order described by Single- $\mathbf{Q}$ vector, $\mathbf{q} = (1/3, 0, 0)$ [81].

For electronic transport measurements, high-quality crystals are patterned into 8-prong switching device using focused-ion beam (FIB) milling. A representative false-colored scanning electron microscope image of a device is shown in Figure 6.2. The yellow regions correspond to the traverse readout contacts ($V_{\perp}(\omega t)$) and the ac current channel (white arrow (I_{ac})). The red and blue bars correspond to the applied pulsed dc currents (I_{pulse}). The longitudinal readout ($V_{\parallel}(\omega t)$) was done across one end of the blue and red bar. The purple regions are milled regions.

The dc pulse currents were single square waves administered with Keithley 6221 Current Sources. Transport measurements were carried out in a Quantum Design 9T Dynacool PPMS. Standard lock-in ac measurements were done with a Zurich Instruments MFLI lock-in amplifier. An ac excitation current ran through the device both during and in between switching events and each measurement in this work had an rms value of 50 μA and a

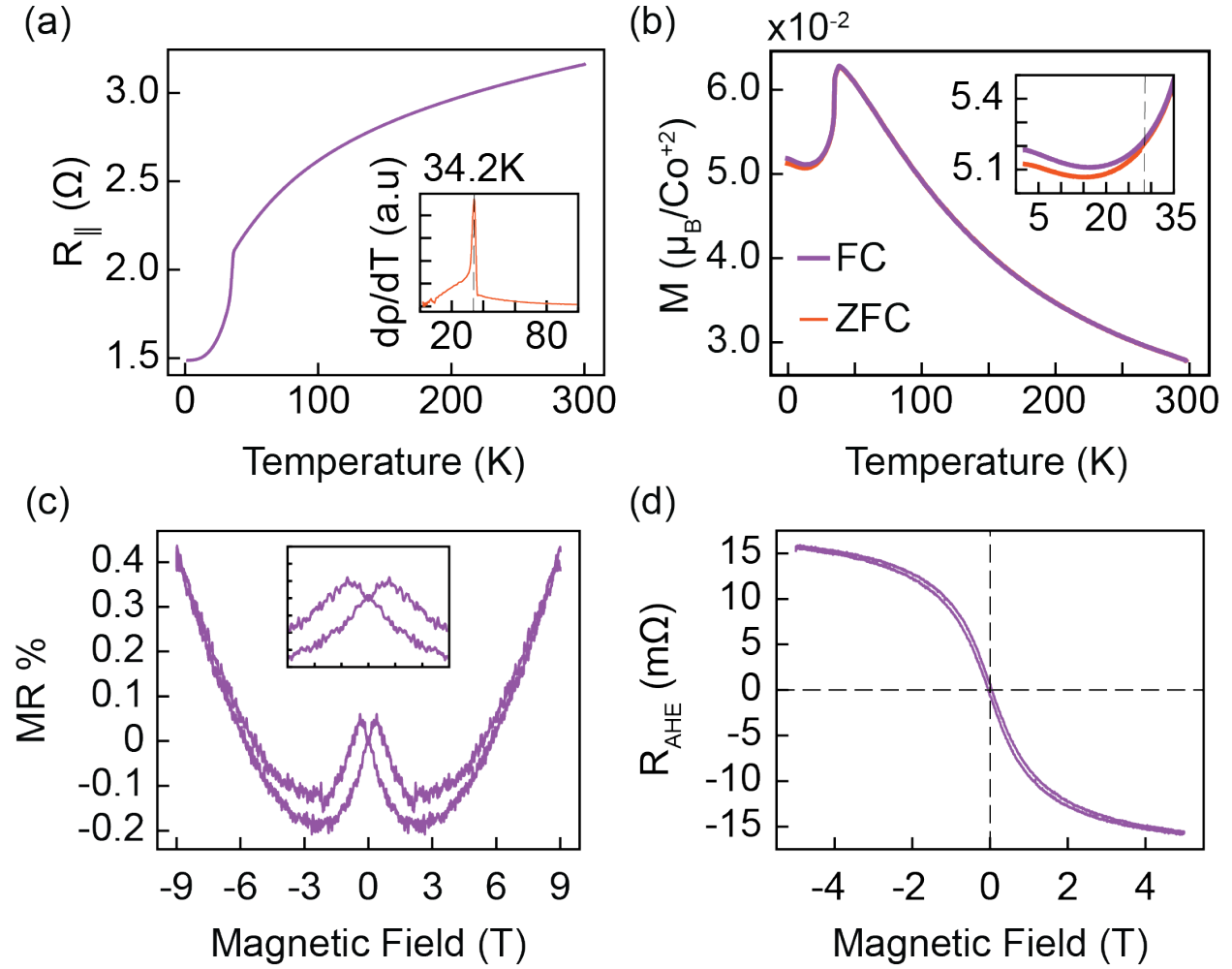


Figure 6.1: (a) Longitudinal resistance versus temperature in a $\text{Co}_{1/3}\text{TaS}_2$ FIB device. Inset shows the derivative of the resistance highlighting a transition at ~ 34 K. (b) Magnetization of a bulk crystals of $\text{Co}_{1/3}\text{TaS}_2$ showing a cusp at ~ 38.9 K. Inset shows splitting in the ZFC and FC curves at ~ 34 K. (c) Magnetoresistance at 2 K in a $\text{Co}_{1/3}\text{TaS}_2$ FIB device. The butterfly-shaped magnetoresistance reflects hysteretic reorientation of the non-collinear spin texture and the formation of a weak-ferromagnetic component in $\text{Co}_{1/3}\text{TaS}_2$. (d) Anomalous hall effect at 2 K in a $\text{Co}_{1/3}\text{TaS}_2$ FIB device.

frequency of 13 Hz.

6.3 Electric Transport

In Figure 6.1 (a), the longitudinal resistance ($R_{\parallel}$) is plotted as a function of temperature. A pronounced decrease in resistance appears approximately 34 K, signaling the onset of a phase transition. The inset displays the temperature derivative of the resistance, where a sharp peak further highlights this transition. In Figure 6.1 (b), the magnetization versus temperature shows a cusps with an onset transition at ~ 40 K. The inset shows the splitting of the ZFC and FC curves at ~ 34 K.

The field-dependent $R_{\parallel}$ and $R_{\perp}$ data were symmetrized (anti-symmetrized) in accordance with equations 5.2. We observe clear evidence for hysteresis in the magnetization of the system, which can be observed in the magnetoresistance (Figure 6.1 (c)), Hall effect (Figure 6.1 (d)) and bulk magnetization (Figure 6.1 (b)). In Figure 6.1 (c), the magnetoresistance exhibits a characteristic butterfly-shaped hysteresis, where the up-sweep and down sweep curves do not retrace and form nested loops. While we do not know the origin of the hysteresis, it does turn on only below the helical order transition temperature, suggesting it is part of the same order parameter or coupled to it. The hysteresis could arise from field-driven reconfiguration of the non-collinear antiferromagnetic state and the emergence of a weak ferromagnetic component at low temperature. Similar butterfly-like MR signatures have been observed in systems with spin-flop transitions, weak ferromagnetism, and topological spin textures. In Figure 6.1 (d) shows the anomalous hall effect at 2 K given by Equation 5.4 with a small coercive field of ~ 50 mT and a hall coefficient of $-1.39 \mu\Omega \text{ cm T}^{-1}$.

6.4 Switching response in $\text{Co}_{1/3}\text{TaS}_2$

Using focused ion beam techniques described in Section 3.2 we fabricated 8-prong switching devices. The device comprises of two perpendicular bars for the application of dc pulse currents and two thinner bars at 45° for the application of small ac excitation current and transverse resistance readout. The device shown in Figure 6.2 (a) is designed so that current can be pulsed along the crystallographic a directions. Specifically current is pulsed along the [100] ‘A’ direction (blue) and the [010] ‘B’ direction shown in (red), which are along the helical axis depending on the domain. This defines the ‘write’ state, which we suppose will orient or couple to the helical domains, resulting in a re-orientation of the principal axes of conductivity. The effect is the measured by passing a ‘read’ current along the narrow contacts which picks the resulting transverse signal labeled $R_{\perp}$. We note that this is not necessarily the same as the transverse signal resulting from the AHE, which is odd in field. Figure 6.2 (a) shows a typical switching protocol: a small dc current pulse is applied along the ‘A’ bar (red), the transverse resistance is measured and then a second small dc current pulse is applied along the ‘B’ bar (blue) and the transverse resistance is measured. A typical switching response is shown in Figure 6.2 (b). An ac excitation current of $50 \mu\text{A}$ was used to probe the system and typical dc pulse currents used range from 10^4 A cm^{-2} up to 10^5 A cm^{-2} .

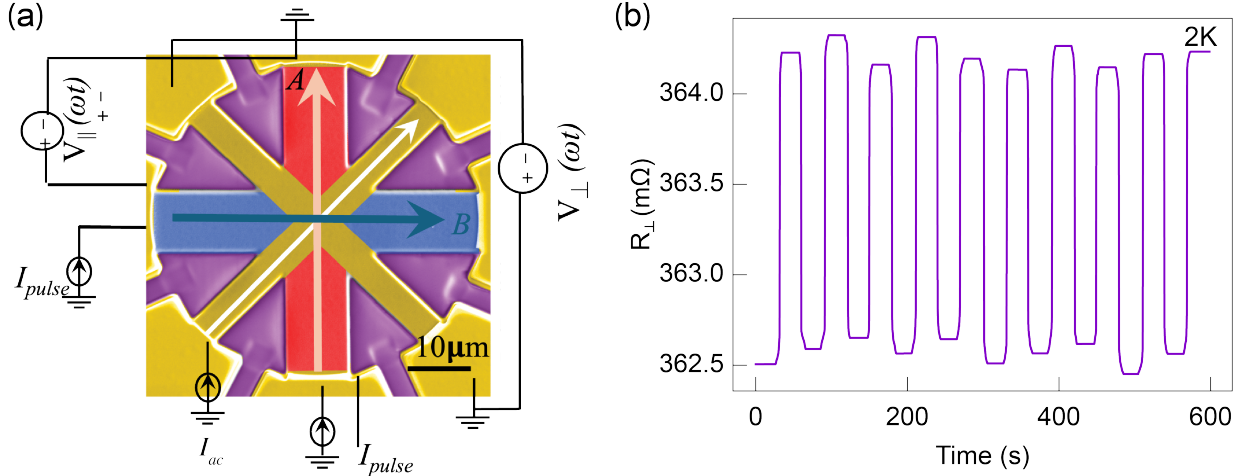


Figure 6.2: **Switching response in a $\text{Co}_{1/3}\text{TaS}_2$ device** (a) False colored SEM micrograph of a $\text{Co}_{1/3}\text{TaS}_2$ 8-prong device. A small ac current is used for electronic readout (white arrow) and two DC currents are pulsed along red and blue bars. The transverse resistance is readout along the gold bars labeled as $V_{\perp}(\omega t)$. (b) The transverse resistance $R_{\perp}$ response to the pulsed DC currents at 2 K. The first pulse brings the system to the low resistance and the second pulse brings the system to the high resistance state.

with pulse durations from 1 m sec to 100 m sec. The time between pulses is generally 30 s.

Figure 6.3 (a) shows the pulse trains for different pulse currents in A and B directions, as measured in the transverse channel at 2 K. The switching response $\Delta R_{\perp}$ was calculated by subtracting the background resistance (R_{avg}) from the measured signal $R_{\perp}$.

$$\Delta R_{\perp} = R_{\perp} - R_{avg} \quad (6.1)$$

The background resistance is determined by finding the average value of between the resistance values due to A pulse and B pulse given by

$$R_{avg} = \frac{1}{2}[R_{\perp}(I_A) + R_{\perp}(I_B)] \quad (6.2)$$

where $R_{\perp}(I_A)$ and $R_{\perp}(I_B)$ is the resistance due to the A-pulse current and to the B-pulse current, respectively. The switching response exhibits a non-monotonic dependence on the applied current density, with a well-defined window where switching is observed. The response emerges at $6.7 \times 10^4 \text{ A cm}^{-2}$, increases to a maximum near $7.5 \times 10^4 \text{ A cm}^{-2}$ and subsequently decreases, becoming fully suppressed for current densities exceeding $8.3 \times 10^4 \text{ A cm}^{-2}$. The critical current are remarkably low $\sim 10^4 \text{ A cm}^{-2}$ - 10^5 A cm^{-2} , around two orders of magnitude below what has been used in prior studies of chiral switching [55, 97], and comparable

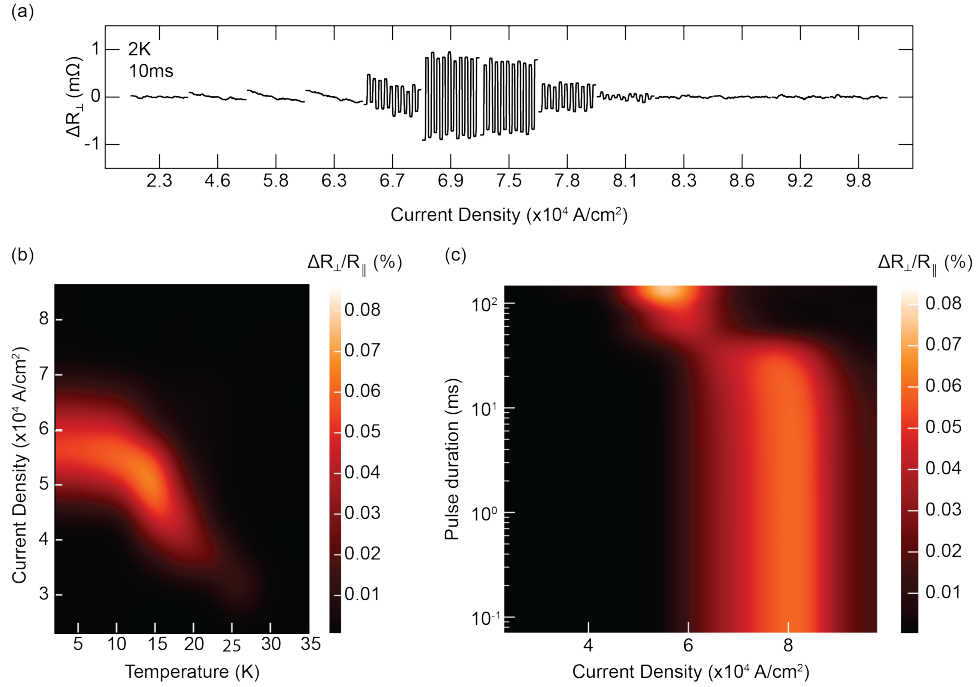


Figure 6.3: **Pulse parameters and temperature dependence of $\text{Co}_{1/3}\text{TaS}_2$.** (a) Pulse current density dependence of the switching response ($\Delta R_{\perp}$) at 2 K and with a pulse duration of 10 ms. There is a small window in current densities where the switching response turns on and switches off thereafter. (b) Pulse current density and temperature dependence of the switching response. The intensity is the amplitude of the relative switching response ($\Delta R_{\perp}/R_{\parallel}$) from peak-to-peak. (c) Pulse current density and pulse duration dependence of the switching response. The intensity is the amplitude of the relative switching response ($\Delta R_{\perp}/R_{\parallel}$) from peak-to-peak. This measurement was performed on a second device, which leads to the switching window appearing at higher current densities, likely due to device-to-device variations such as differences in geometry, contact resistance, or thermal coupling.

only to those seen in the system $\text{Fe}_{1/3}\text{NbS}_2$ [98]. Remarkably, there is not only a critical current at which the switching turns on, there is also a critical current at which the switching turns off. In this case $j_{on} \approx 6.7 \times 10^5$ A cm⁻² and $j_{off} \approx 8.1 \times 10^5$ A cm⁻².

Temperature Dependence

The temperature dependence of the switching response as a function of current density is shown in Figure 6.3 (b). The plot highlights how the switching amplitude evolves with increasing current density and temperature, revealing the onset and suppression regions of the response. The amplitude of the relative switching response, ($\Delta R_{\perp}/R_{\parallel}$ %), at ($6.7 \times$

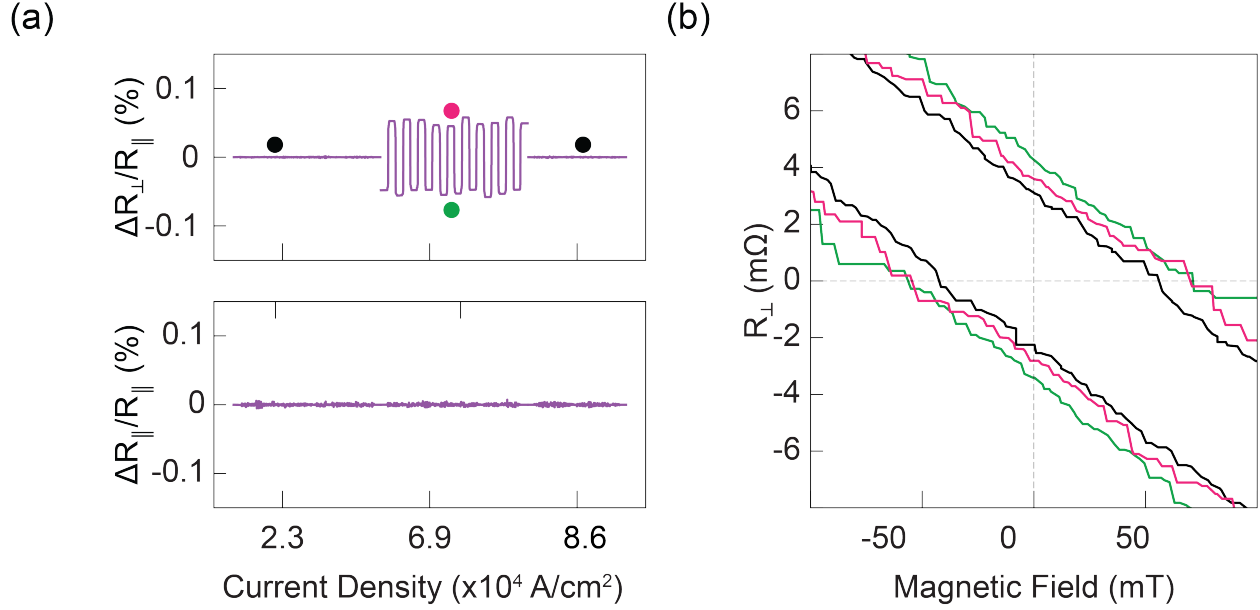


Figure 6.4: **Hysteresis dependence on pulse currents in a $\text{Co}_{1/3}\text{TaS}_2$ device** (a) The relative switching response at different pulse currents in transverse and longitudinal channels. (b) Transverse Hall resistance ($R_{\perp}$) after pulse measurements in (a) Rose color corresponds to high resistance state and green to the low resistance state.

10^4 A cm⁻²) increases from approximately (0.17%) at 2 K to about (1.21%) at 15 K. This enhancement with temperature suggests increased spin-torque efficiency or domain mobility within the antiferromagnetic state. As the temperature approaches 20 K, the window of current densities exhibiting a switching response narrows, reducing to a single observable current density of (5.8×10^4 A cm⁻²). The suppression of switching near this temperature likely reflects thermal destabilization of the antiferromagnetic order or reduced torque efficiency as the system approaches the Néel transition. For temperatures above 30 K, the switching response is fully suppressed at all current densities, consistent with the loss of antiferromagnetic order beyond the transition temperature. Figure 6.3 (c) is a 2D heat map showing how the normalized transverse resistance change, ($\Delta R_{\perp}/R_{\parallel}$ %), depends on current density and pulse duration. The non-monotonicity behavior of the switching response also extends to pulse duration dependence. At low current densities ($\lesssim 5 \times 10^4$ A cm⁻²), the map is mostly dark, indicating a negligible resistance change over all pulse durations. A sharp onset threshold appears around $6 - 7 \times 10^4$ A cm⁻², where the resistance change becomes appreciable. At higher current densities ($\sim 7 - 8.5 \times 10^4$ A cm⁻²), a broad vertical band of elevated signal emerges, persisting nearly the entire pulse-duration range. This suggests the response is primarily current-duration rather than strongly time-limited. A localized maximum, appears at intermediate currents ($\sim 5 - 6 \times 10^4$ A cm⁻²) and long pulse durations

(~ 100 s), indicating enhanced sensitivity when long pulses are applied near the threshold. The map indicates a current density threshold for switching or reconfiguration, with pulse duration playing a secondary role once the threshold is exceeded. This behavior is consistent with a current-induced mechanism (e.g., domain reorientation or electrically driven magnetic/structural changes) rather than purely thermal accumulation.

Magnetic Field Dependence

We track the hysteresis loop at each point in the on and off behavior of the switching response and as well in the high and low resistance states each of these points are illustrated by the colored circles in Figure 6.4 (a). Each device is initialized at low temperature by polarizing the system in fields up to 9 T before a pulse sequence begins. This means we always begin on one side of the hysteresis loop. While the size of the hysteresis loop changes slightly in the high, low and off state, in every case we observe that we are always on the same side of the hysteresis loop as the initialized state. The switching preserves its out-of-plane magnetization, but re-orientes the chiral order in-the-plane. This data also serves to show that the system is not being heated above its transition temperature, which would vanish the magnetization. On the contrary, the hysteresis loop tends to grow, suggesting a switching-induced-enhancement of the stiffness of the ferromagnetic moment.

The switching behavior is robust to the application of in and out of plane fields. In Figure 6.5 we illustrate four situations; zero-field switching after initialization at ± 9 T, and in-field switching at ± 9 T. The critical current for the on and off state is almost completely unchanged. The magnitude of the switching slightly, but measurably increases at 9 T. This should be contrasted to other AFM switching experiments, where switching is not possible when the spin texture is polarized by field. Even in $\text{Fe}_{1/3}\text{NbS}_2$ switching is not possible at high magnetic fields, even though the system may maintain memory of the zero-field state [98]. This illustrates an extraordinary robustness of the switching response to the magnetic field. Furthermore, a detailed field dependence on the switching response was done at varying current densities. Figure 6.6 presents an activation map of the absolute resistance response, $\Delta R_{\perp}/R_{\parallel}(\%)$, as a function of current density and magnetic field. A narrow current threshold near $J \approx 6 - 7 \times 10^4 \text{ A cm}^{-2}$ produces a strong, field-robust response, consistent with current-driven manipulation of the antiferromagnetic spin texture. The persistence of the signal across both positive and negative fields is indicative of a spin-orbit-torque-like mechanism acting on noncollinear antiferromagnetic domains rather than a purely field-controlled effect.

Discussion

When a current is applied $\mathbf{j} \parallel \mathbf{q}$, the helix deforms by a finite cone angle ϕ as mentioned in section 1.3. The spin transfer torque is governed by the Landau-Lifshitz-Gilbert (LLG) equation, modified to include a spatially varying magnetization given by Equation 1.17. On large timescales, the initially multidomain helical phase transforms into a monodomain phase

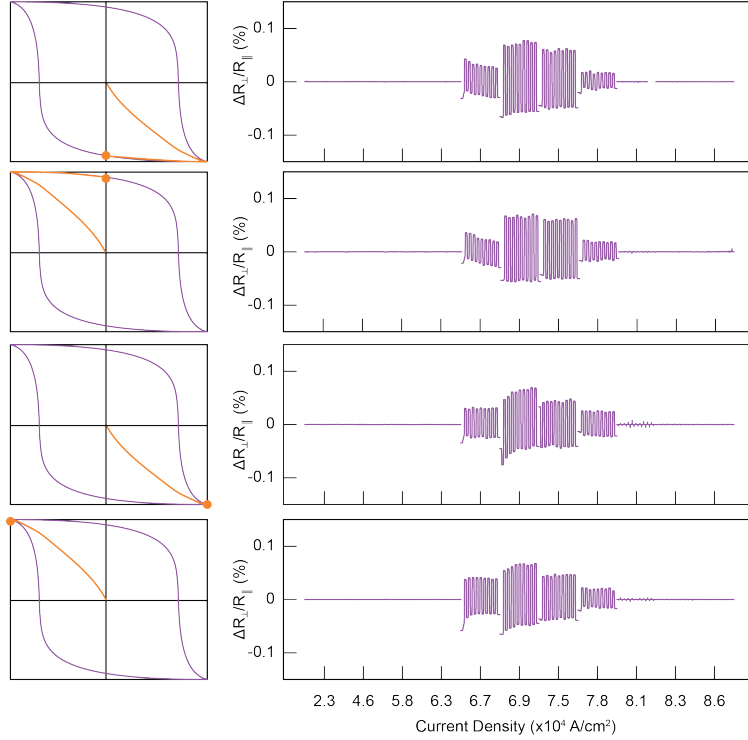


Figure 6.5: **Switching response in $\text{Co}_{1/3}\text{TaS}_2$ to magnetic field training and with an applied field of ± 9 T** (a) The magnetic field was swept to $+9$ T and back to 0 T to polarize the device. The right panel consecutive pulse trains after the field polarization. (b) The magnetic field was swept to -9 T and back to 0 T to polarize the device. The right panel consecutive pulse trains after the field polarization. (c) The magnetic field was swept to $+9$ T and parked there. back to 0 T to polarize the device. The right panel consecutive pulse trains after the field polarization. (d) The magnetic field was swept to -9 T and parked there. The right panel consecutive pulse trains after the field polarization.

with $\mathbf{j} \parallel \mathbf{q}$. This ordering process is driven by the dynamics of topological defects in the helical texture which carry a non-quantized topological charge,

$$Q = \iint_{\Omega} \hat{\mathbf{m}} \cdot \left(\frac{d\hat{\mathbf{m}}}{dx} \times \frac{d\hat{\mathbf{m}}}{dy} \right) d\mathbf{r} \quad (6.3)$$

where $\hat{\mathbf{m}} = \mathbf{M}/M_s$ is the normalized magnetization. As mentioned above, these topological charges arise naturally at interfaces of differently oriented helices, namely domain walls. In $\text{Co}_{1/3}\text{TaS}_2$, there are three favored orientations to these domains, so that two perpendicular directions of applied current act differently. For currents applied parallel to a principal axis, a single domain is favored, while perpendicular to it two domains are favored, so that the

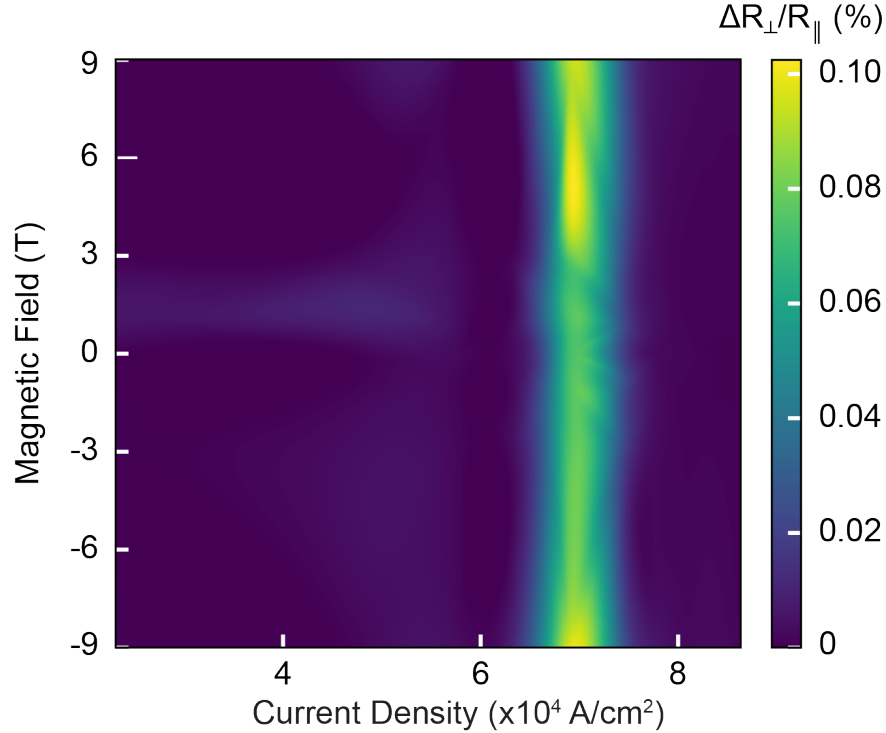


Figure 6.6: **Current-field activation map of the transverse resistance response.** Color map of the absolute normalized transverse resistance change, $(\Delta R_{\perp}/R_{\parallel}(\%))$, as a function of applied current density and magnetic field. A pronounced vertical ridge centered at $J \approx 6-7 \times 10^4 \text{ A cm}^{-2}$ marks the onset of a strong electrically driven response that persists over the full magnetic-field range, indicating a predominantly current-controlled switching regime.

formation of topological defects of a specific domain configuration is favored. This helps explain the data shown in Figure 6.7 (a); when the pulse is parallel to the principal axis, the switching reveals one resistance, while the perpendicular direction each pulse systematically changes the resistance as the topological defects form. The resistance state is not simply a measure of the conductivity anisotropy of a monodomain of the helical order, it is an indicator of the number of topological defects at the interface of helical domains.

One the remarkable observation of the current study is that we also observe a switching “off” current. It is known theoretically that at sufficiently large current the cone angle $\phi \rightarrow 0$, which will arrest the motion of domains walls in analogy to Walker breakdown. Due to the anisotropy of the collective modes, the critical current at which Walker breakdown occurs (j_W) depends on the projection on $\mathbf{q}$, so that for $\mathbf{j} \perp \mathbf{q}$ the spin order is already soft. We expect therefore that

$$\mathbf{j}_W(\mathbf{r}) \approx \mathbf{j}_W(\mathbf{q}) \cos \theta \quad (6.4)$$

where θ is the angle between $\mathbf{r}$ and $\mathbf{q}$. This answers two interesting observations. Firstly, currents along a principal axis, will reach the Walker breakdown before current perpendicular ($\mathbf{j}^B$ is oriented $\pi/3$ to the other principal axes), so that switching in the former will cease before switching in the latter. This is consistent with our data (see Figure 6.7 (b)) where the switching along the A direction appears to stop being effective, while switching in the B direction continues. The B direction reaches the Walker breakdown roughly $1.15\mathbf{j}_W^A$, which is roughly $\mathbf{j}_W^A/\cos(\pi/3)$. Secondly, the same argument can be used for the switching “on” current, because the A will begin to switch before the perpendicular direction, just as observe. When both A and B pulse directions are aligned with the principal axes, both directions switch “on” and “off” at exactly the same critical current.

The currents we see are much more comparable to the AFM switching in $\text{Fe}_{1/3}\text{NbS}_2$. In that case, it was suggested that the low current densities are due to the intrinsic frustration of the system. Specifically, as a function of iron composition x , $\text{Fe}_{1/3}\text{NbS}_2$ favors two different order parameters, can even coexist coexist near $x = 1/3$. In $\text{Co}_{1/3}\text{TaS}_2$ a similar phenomenon may be occurring. Park *et al.* showed that the composition dependence connects an order parameter of scalar spin chirality to one of helical order [81]. Near the compositions studied here $x \sim 1/3$, the system may be feeling a similar level of frustration that leads to a softening of the spin order, enabling electrical control at low current densities. Notably, this may also explain the extremely low Walker breakdown currents, which are theoretically thought to be orders of magnitude larger than the current needed to comb the helical order. The intrinsic frustration of the system may lead to such gross softening that switching is observed in a narrow window of current.

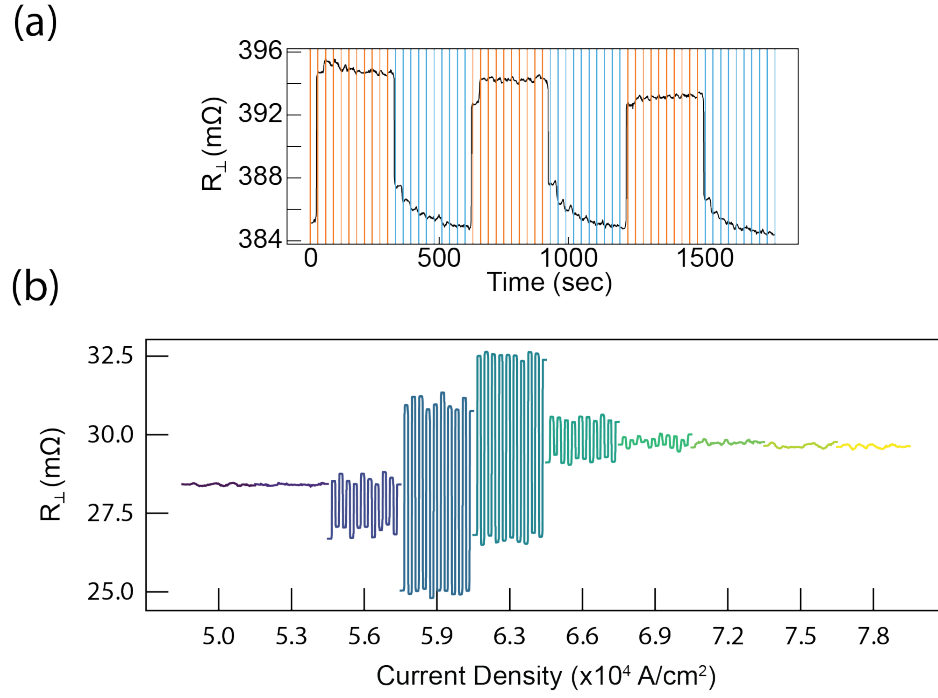


Figure 6.7: **Pulse direction dependence on switching response** (a) Repeat triggers along A pulse direction in red and along B pulse direction in blue. (b) Pulse current density dependence on the transverse direction $R_{\perp}$ illustrating pulse along A direction reaches the Walker break down before B pulse direction. Both measurements were done at 2 K.

Chapter 7

Conclusion and Outlook

“The grace of the Lord Jesus be with God’s people. Amen.”

— Revelation 22:21

The work presented in this dissertation explored the relationship on how the intercalation level would influence the magnetic properties of the intercalated transition metal dichalcogenides and how an electrical stimulus would effect the transport properties. The motivation stemmed from previous work conducted on $\text{Fe}_{1/3}\text{NbS}_2$ a c-axis antiferromagnet which shows a distinct response to electrical stimulus. In part we explored two systems that are in the family, and ferromagnet and and antiferromagnet.

Chapter 4 presented the response of $\text{Ni}_{1/4}\text{TaSe}_2$ to an electrical stimulus. This material platform is a c-axis itinerant ferromagnet with a small in-plane moment. The original motivation was to see if the anomalous Hall effect in this material could be electrically switched via electrical pulses. None was found. However, this material platform showed to be a novel variant of a heat-assisted magnetization recording device, in which magnetization reversal is done via an electrical pulse and not a laser and with an integrated electronic readout. Thus making this device an all-electronic read-write scheme for magnetization recording. A possible direction is the exploration at the intercalation level of $\text{Ni}_{1/3}\text{TaSe}_2$ which to the time of the writing of this dissertation has not been synthesized. Both for its magnetic properties and device applications and as a candidate material for the elusive altermagnetism [99].

Chapter 5 presented the intercalation dependence of Co_xTaS_2 over the range $x \sim 0.25 - 0.33$. We find that the anomalous Hall effect (AHE) is large near $x = 1/4$, is strongly suppressed for intermediate concentrations $0.25 < x < 0.30$, and increases again around $x \sim 0.325$, where the AHE is attributed to scalar spin chirality. Upon approaching the commensurate composition $x = 1/3$, the AHE is again dramatically reduced. While the magnetic order is well established for $x \geq 0.325$, the nature of the magnetic ground state

at $x = 1/4$ remains unknown. Careful attention must be paid to how intercalation influences the magnetic order in this family of materials. It has been shown that superstructure formation can stabilize both majority and minority phases corresponding to the commensurate intercalation levels $x = 1/4$ and $x = 1/3$ within the same crystal, leading to phase coexistence within a single sample [100].

Chapter 6 presented measurements on the helical antiferromagnet $\text{Co}_{1/3}\text{TaS}_2$, which was found to be electronically switchable at surprisingly low current densities similar to the current densities found in $\text{Fe}_{1/3}\text{NbS}_2$. The combing of the helical order via electrical currents, writes information into the device and the change in the transverse resistance is the readout. An all-electrical switching of the scalar spin chirality has recently been observed in Co_xTaS_2 (with $x \sim 0.325$) via pulse currents up to $8 \times 10^6 \text{ A cm}^{-2}$ [101], several orders of magnitude higher than pulse currents used in $\text{Ni}_{1/4}\text{TaSe}_2$ and $\text{Co}_{1/3}\text{TaS}_2$. As discussed in Chapter 4, the applied current pulses can generate a substantial amount of Joule heating in the device. Although switching mechanisms described in the introduction, such as spin-orbit torque, may be present, their contribution could be significantly weaker than the thermal effects produced by the pulse currents. In this case, the dominant energy scale may arise from Joule heating, which could raise the device temperature above the magnetic transition and thereby drive the observed switching. Therefore, it is essential to establish an experimental method to quantify the extent of heating introduced into the device by the current pulses used to induce switching in these material platforms.

A promising future direction is a systematic study of pulse-driven switching as a function of the cobalt intercalation ratio in Co_xTaS_2 . Because intercalated cobalt ions introduce localized magnetic moments and modify the electronic structure of the host lattice, varying the intercalation level provides a powerful means of tuning the magnetic exchange interactions, and scattering environment to be tuned. These changes can strongly influence both thermally driven switching and current-induced mechanisms such as spin-orbit torques. By exploring the switching behavior across a broader range of intercalation levels, it may be possible to disentangle these competing mechanisms and identify the dominant energy scales governing the switching process. Such studies could also provide insights into how magnetic intercalation can be used to engineer current-driven phenomena in layered materials, with potential implications for the design of future spintronic devices based on transition-metal dichalcogenides. Overall, this work highlights how magnetic intercalation in layered materials provides a versatile platform for exploring the interplay between electronic transport, magnetism and current-driven phenomena in low-dimensional quantum materials.

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